

FY 1997 Project for the Advise ment of Sustainable Development Support  
Commissioned by Environment Agency, Government of Japan

# Air Pollution Control Technology Manual

March 1998

Overseas Environmental Cooperation Center

## Preface

Japan has manifested, both domestically and internationally, a policy of strengthening support for environmental conservation in developing countries, and it is hoped that Japan will play a major role in technological cooperation by translating this policy into more "visible assistance" and dispatching capable personnel to developing countries so that specialists from both Japan as well as developing countries will be able to collaborate in problem solving.

A distinguishing feature of the environmental problems in developing countries is that environmental deterioration on a global scale is occurring at the same time as serious environmental pollution like that Japan experienced during its high growth period. In addition, regardless of the present situation, the pressures involved in the economic development that these countries are seeking is growing all the more while they continue to neglect paying sufficient consideration to the environment.

When Japanese specialists are dispatched to developing countries that are facing such circumstances, they are often questioned about a wide-range of issues concerning environmental conservation which are not limited to their personal fields of specialty. An obvious need has arisen to provide materials with such a wide range of content in order to support these types of specialists.

With this situation in mind, extensive materials on conservation technology, within the field of air quality conservation, which has become a serious issue and here has been much demand for technology transfer from developing countries, have been put together in this "Air Pollution Control Technology Manual". It would give me great pleasure for this manual to be utilized not only for the transfer of technology concerning the environmental field from Japan to these developing countries but also as research material for the training of international environmental cooperation specialists.

This manual was written by persons of learning and experience in the field of air pollution control in Japan, and it was put together by various specialists in each field with advice from the members of The Air Pollution Control Technology Manual Committee. I would also like to thank Professors Yasuji Himi and Yoshio Saiki for taking time out from their busy schedules to proofread the manual. I would like to extend my greatest appreciation to all those who have been involved in the making of this manual.

March, 1998

Osamu Watanabe, Managing Director  
Overseas Environmental Cooperation Center, Japan

# Contents

<b>Chapter 1 An Introduction to Air Pollution</b> .....	<b>1</b>
1.1 An Outline of the "Atmosphere" .....	1
1.1.1 Composition of the Atmosphere .....	1
1.1.2 Structure of the Atmosphere .....	2
1.1.3 Solar Radiation .....	3
1.2 Definition of Air Pollution and Air Pollutants .....	5
1.2.1 Foreword .....	5
1.2.2 The Sources and Types of Air Pollutants .....	5
1.2.3 Air Pollutants .....	5
1.2.4 Other Air Pollutants .....	6
1.2.5 New Air Pollutants .....	7
1.3 Air Pollution and the Issue of Resources .....	8
1.3.1 Energy and Air Pollution .....	8
1.3.2 Air Pollution Control and Resource Recovery .....	9
1.3.3 Resource Development and Environmental Issues .....	9
1.3.4 Securing and Treating Water for Industrial Use and Transportation Issues .....	10
1.3.5 Environment and Resource Issues .....	10
1.4 Global environmental problems .....	11
1.4.1 Introduction .....	11
1.4.2 Global warming .....	11
1.4.3 Depletion of the stratospheric ozone layer .....	12
1.4.4 Acidification of the tropospheric air (acid rain) .....	12
<b>Chapter 2 History of Air pollution</b> .....	<b>15</b>
2.1 Historical Review of Air pollution around the World .....	15
2.1.1 Introduction .....	15
2.1.2 England .....	16
2.1.3 America .....	17
2.1.4 Air pollution in the Megalopolises around the world .....	18
2.2 Historical Retrospective of Air Pollution in Japan .....	20
2.2.1 Air Pollution, the First Stage .....	20
2.2.2 Air Pollution, the Second Stage .....	23
2.2.3 Air Pollution, the Third Stage .....	28
<b>Chapter 3 Background to air pollution regulations in Japan</b> .....	<b>35</b>
3.1 Overview .....	35

3.2	Impetuous and background to establishment of regulations .....	35
3.3	Technical aspects behind the implementation of regulations .....	35
3.4	Social aspects behind the implementation of regulations .....	36
3.5	Administrative structure as an aspect of implementation of regulations .....	37
3.6	Background to the era .....	38
<b>Chapter 4</b>	<b>Weather Conditions, and the Variation and Distribution of the Concentration of Air Pollutants .....</b>	<b>43</b>
4.1	Effect of Weather Conditions .....	43
4.1.1	Preface .....	43
4.1.2	Atmospheric Boundary Layer .....	43
4.1.3	Vertical Variation of Air Temperature .....	43
4.1.4	Vertical Distribution of Temperature and Diffusion .....	44
4.1.5	Vertical Distribution of Wind .....	45
4.1.6	Urban Wind and Air pollution .....	45
4.1.7	Urban Boundary Layer and Air Pollution .....	46
4.1.8	Synoptic Meteorology and Local Meteorology .....	47
4.2	Formation Mechanism of Air Pollutants .....	49
4.2.1	Introduction .....	49
4.2.2	Photochemical Reaction of Ozone Formation .....	49
4.2.3	Reaction of Hydrocarbons .....	50
4.2.4	Atmospheric Chemical Reaction Associated With Acidification of Tropospheric Air (Acid Rain) .....	52
4.3	Concentration Variation of Air Pollutants .....	54
4.3.1	Introduction .....	54
4.3.2	Diurnal Variation of Air Pollutant Concentration and the Mutual Relationship Among Pollutants .....	54
4.3.3	Seasonal Variation of Air Pollutant Concentration .....	56
4.3.4	Annual Variation of Air Pollutant Concentration and Its Statistical Analysis .....	56
4.4	Distribution of Air Pollutants .....	57
4.4.1	Introduction .....	57
4.4.2	Flow of Air Pollution from City to Suburbs .....	57
4.4.3	3D Distribution of Air Pollution .....	58
4.4.4	Distribution of Wide Area Air Pollution .....	62
4.4.5	Measurement of Distribution and Monitoring of Air Pollution .....	63
<b>Chapter 5</b>	<b>Effect of Air Pollutants .....</b>	<b>67</b>
5.1	Health effects on human .....	67
5.1.1	Introduction .....	67
5.1.2	Respiratory system .....	67



5.1.3	What respiratory diseases are induced by air pollutants? .....	67
5.1.4	Health effects of typical air pollutants .....	69
<b>5.2</b>	<b>Effect on Vegetation .....</b>	<b>72</b>
5.2.1	Introduction .....	72
5.2.2	Expressions of visible plant damage .....	72
5.2.3	The effect on plant's physiological function and yield .....	75
<b>5.3</b>	<b>The effects of air pollution on materials and cultural properties .....</b>	<b>78</b>
5.3.1	Introduction .....	78
5.3.2	The effects of air pollution relating to raw materials .....	78
5.3.3	Mechanisms for the effects of atmospheric pollution .....	81
5.3.4	Conclusion .....	81
<b>5.4</b>	<b>Visibility degradation .....</b>	<b>82</b>
5.4.1	Introduction .....	82
5.4.2	Causes of visibility degradation .....	82
5.4.3	Measuring visibility .....	84
5.4.4	Effects of visibility degradation .....	84
<b>5.5</b>	<b>Effects on senses (sense of smell) .....</b>	<b>86</b>
5.5.1	Sense of smell .....	86
5.5.2	Features of the sense of smell .....	87
5.5.3	Effects on the physiological functions of odors .....	89
5.5.4	Effects of odors and their uses .....	90
<b>Chapter 6</b>	<b>Overview of Sources of the Air Pollutants .....</b>	<b>95</b>
<b>6.1</b>	<b>Stationary Combustion Facilities .....</b>	<b>95</b>
6.1.1	Introduction .....	95
6.1.2	Formation of Major Air Pollutants .....	96
6.1.3	Thermal Power Station .....	97
<b>6.2</b>	<b>Source of Dust .....</b>	<b>101</b>
6.2.1	Introduction .....	101
6.2.2	Pulverization or Crushing Process .....	101
6.2.3	Polishing Process .....	102
6.2.4	Sieving of Particles .....	102
6.2.5	Transportation of Particles .....	102
6.2.6	Pile of Particles .....	103
<b>6.3</b>	<b>Chemical Process .....</b>	<b>104</b>
6.3.1	Introduction .....	104
6.3.2	Cement Industry .....	104
6.3.3	Sulfuric Acid Industry .....	106

6.3.4	Nitric Acid Industry .....	107
6.3.5	Chlorine Production .....	107
<b>6.4</b>	<b>Metal Refinery .....</b>	<b>109</b>
6.4.1	Introduction .....	109
6.4.2	Integrated Steel Works .....	109
6.4.3	Copper Refinery .....	112
6.4.4	Zinc Refinery .....	113
6.4.5	Aluminum Refinery .....	115
6.4.6	Refinery of Titanium and Silicon .....	115
<b>6.5</b>	<b>Petroleum Refinery .....</b>	<b>116</b>
6.5.1	Petroleum .....	116
6.5.2	Petroleum Refinery .....	117
6.5.3	Desulfurization of Heavy Oil .....	119
6.5.4	Air Pollutants .....	121
<b>6.6</b>	<b>Transportation .....</b>	<b>123</b>
6.6.1	Overview .....	123
6.6.2	The present situation of the motor vehicles as a facilities of transportation .....	123
6.6.3	The present situation of the motor vehicles as a pollution source .....	123
6.6.4	Other modes of transportation .....	126
6.6.5	Indicated problems .....	127
<b>6.7</b>	<b>Sources of offensive odors .....</b>	<b>128</b>
6.7.1	State of offensive odor complaints .....	128
6.7.2	Worksites generating specific offensive odor substances .....	128
6.7.3	Impact of the main offensive odor generating worksites .....	129
6.7.4	Main worksite offensive odor sources .....	130
<b>6.8</b>	<b>Emission factors for atmospheric pollutants .....</b>	<b>133</b>
6.8.1	Outline .....	133
6.8.2	Air pollutant emission factors from stationary sources .....	134
6.8.3	Air pollutant emission factors from mobile sources .....	137
<b>Chapter 7</b>	<b>Air Pollution Control Technology .....</b>	<b>145</b>
<b>7.1</b>	<b>Basic Ideas Behind Air Pollution Control .....</b>	<b>145</b>
7.1.1	Overview .....	145
7.1.2	Basic considerations on air pollution prevention .....	145
7.1.3	Air Pollution Control Equipment .....	145
7.1.4	The establishment of an air pollution control organization .....	146
<b>7.2</b>	<b>Transitions in Countermeasures to Air Pollution .....</b>	<b>148</b>
7.2.1	Introduction .....	148

7.2.2	Examples of instruction and agreements .....	148
7.2.3	Gradual introduction of restrictions and results. ....	148
<b>7.3</b>	<b>Better fuel countermeasure .....</b>	<b>153</b>
7.3.1	Introduction .....	153
7.3.2	The necessity of better fuel countermeasure. ....	153
7.3.3	Fuel usage standards as a Air Pollution Control Law .....	153
7.3.4	The effects of using better fuel .....	153
7.3.5	Supply of high-quality fuels .....	154
<b>7.4</b>	<b>Combustion control and energy saving .....</b>	<b>156</b>
7.4.1	The meaning of combustion control .....	156
7.4.2	Trends in combustion control and energy saving in Japan .....	156
7.4.3	Items to mention .....	159
<b>7.5</b>	<b>Process Management .....</b>	<b>160</b>
7.5.1	Overview .....	160
7.5.2	Method through Cleaning of a Workplace .....	160
7.5.3	Method through Management of Raw Materials and Fuels Used during the Process .....	160
7.5.4	Strengthening of the Process Maintenance and Management .....	161
7.5.5	Improving Processes .....	161
7.5.6	Rationalizing Processes Through Combination .....	161
<b>7.6</b>	<b>Dust Collector .....</b>	<b>163</b>
7.6.1	Introduction .....	163
7.6.2	Changes in the dust collecting techniques in Japan .....	163
7.6.3	Classification of dust collector .....	163
7.6.4	Development of new technology .....	164
7.6.5	Conclusion .....	167
<b>7.7</b>	<b>Flue Gas Desulfurization Equipment and By Products .....</b>	<b>168</b>
7.7.1	Various Processes of Desulfurization .....	168
7.7.2	Future Trends for Flue Gas Desulfurization .....	174
<b>7.8</b>	<b>Improving combustion as a countermeasure to NOx .....</b>	<b>175</b>
7.8.1	Introduction .....	175
7.8.2	History of combustion improvement as a countermeasure against NOx in Japan .....	175
7.8.3	Low NOx combustion technology in Japan .....	175
7.8.4	Indicated problems .....	178
<b>7.9</b>	<b>Denitrification of Exhaust Gas .....</b>	<b>179</b>
7.9.1	Introduction .....	179
7.9.2	History of Flue Gas Denitrification Techniques in Japan .....	179
7.9.3	Recent trends in Flue Gas Denitrification Technology in Japan .....	181
7.9.4	Specified Issues .....	183

7.10 Toxic Gas Treatment .....	184
7.10.1 Adsorption Method .....	184
7.10.2 Absorption Method .....	188
7.11 Automobile Exhaust Gas Control Measures .....	189
7.11.1 Introduction .....	189
7.11.2 Measure for gasoline fueled vehicle exhaust gas control .....	189
7.11.3 Diesel powered vehicles exhaust gas emission control measures .....	191
7.11.4 Specified Issues .....	194
7.12 Hydrocarbon Species Countermeasures .....	195
7.12.1 Introduction .....	195
7.12.2 Hydrocarbon Species Emission Sources .....	195
7.12.3 Countermeasures to Hydrocarbon Species in Combustion Emission Gas .....	195
7.12.4 Gasoline Vapor Control Measures .....	196
7.12.5 Hydrocarbon Species Treatment Apparatus .....	197
7.12.6 Countermeasures of hydrocarbon included chlorine .....	198
7.12.7 Chlorofluorocarbon .....	198
7.13 Offensive Odor Control Measures .....	199
7.13.1 Planning for Controlling Offensive Odor Deodorization Method .....	199
7.13.2 Deodorizing technologies .....	199
7.14 Development Planning and Examples of Countermeasure .....	203
7.14.1 Overview .....	203
7.14.2 The Negishi Bay Industrial Area Development and the Yokohama Type Agreement on Pollution Control .....	203
7.14.3 The Yokkaichi Petroleum Complex Plan and the Air Pollution Problem .....	204
7.14.4 The Significance and Air Pollution Counter Measures of the Ogishima Project .....	205
7.15 The cost of air pollution control technology .....	207
7.15.1 Changes in plant and equipment investment capital .....	207
7.15.2 Sulfur Oxides Countermeasures .....	207
7.15.3 The countermeasure cost of individual investment- the example of heavy oil power plant .....	209
7.15.4 The economic influence of sulfur countermeasures .....	209
<b>Chapter 8 Measuring method of air pollutants (emission sources) .....</b>	<b>215</b>
<b>8.1 Measuring and analytical method of air pollutants in exhaust gas .....</b>	<b>215</b>
8.1.1 Overview .....	215
8.1.2 Particle concentration measuring method .....	216
8.1.3 Gaseous pollutants concentration measuring methods .....	218
8.1.4 Calculation of quantities of air pollutant emissions .....	219
8.1.5 Easy measuring methods of black smoke emission using Ringleman's smoke chart .....	220

8.2	Methods for measuring and analyzing the main components of combustion exhaust gas (flue gas) .....	221
8.2.1	Overview .....	221
8.2.2	Sampling methods of sample gas .....	221
8.2.3	Orsat analyzer .....	222
8.2.4	Methods using measuring equipment .....	222
8.3	Automatic exhaust gas air pollutant measurer .....	225
8.3.1	Overview .....	225
8.3.2	Composition of the continuous measuring system .....	225
8.3.3	Continuous sulfur oxides analyzer in flue gas .....	225
8.3.4	Continuous nitrogen oxides analyzer in flue gas .....	226
8.4	Fuel testing methods .....	228
8.4.1	Overview .....	228
8.4.2	Analytical method of sulfur in fuelsulfur in fuel .....	228
8.4.3	Analytical method of nitrogen in fuel .....	228
8.4.4	Analytical method of carbon and hydrogen in fuel .....	229
8.4.5	Testing method on Calorific value of fuel .....	229
8.5	Air Pollutant Control Equipment .....	230
8.5.1	Overview .....	230
8.5.2	Dust collector testing methods .....	230
8.5.3	Desulfurization and denification equipment for flue gas testing method .....	234
8.6	Exhaust Gas Measurement for Motor Vehicles .....	235
8.6.1	Introduction .....	235
8.6.2	Measuring equipment overview .....	235
8.6.3	Procedure for testing motor vehicle exhaust gas .....	240
8.6.4	Indicated problems .....	241
Chapter 9	Air pollution measuring methods (atmosphere) .....	245
9.1	Basis of air pollution measurement .....	245
9.1.1	Introduction .....	245
9.1.2	Measurement methods classification and sampling .....	245
9.1.3	Manual analysis methods .....	247
9.1.4	Concentration meter .....	249
9.1.5	Continuous analyzers .....	249
9.1.6	Measurement accuracy and measurement management .....	250
9.1.7	Laboratory management .....	251
9.2	Continuous analysis methods .....	252
9.2.1	Introduction .....	252

9.2.2	Continuous analyzer .....	252
9.2.3	Objectives and types of ambient air measurements .....	253
9.2.4	Construction and facilities of measuring stations .....	253
9.2.5	State and responses to international standards .....	254
9.3	Planning and managing air pollution monitoring systems .....	259
9.3.1	Introduction .....	259
9.3.2	Planned disposition of the monitoring system .....	259
9.3.3	Measurement sites and area of measuring station .....	261
9.3.4	Installation conditions of measuring equipment .....	261
9.3.5	Maintenance and management of measuring equipment .....	262
9.4	Dust fall measurement methods .....	263
9.4.1	Introduction .....	263
9.4.2	Sampling method .....	263
9.4.3	Analysis .....	264
9.5	Measurement Techniques of Acid Deposition .....	266
9.5.1	Introduction .....	266
9.5.2	Points for Wet Deposition Measurement Techniques .....	266
9.5.3	Summary .....	269
9.6	Methods of analyzing airborne particulate matter .....	270
9.6.1	Introduction .....	270
9.6.2	Atomic absorption photometry .....	270
9.6.3	Radio activation analysis .....	272
9.7	Measurement methods for polycyclic aromatic hydrocarbons in suspended particles .....	274
9.7.1	Introduction .....	274
9.8	Methods of Analyzing Toxic Airborne Compounds (Volatile Organic Compounds) .....	276
9.8.1	Introduction .....	276
9.8.2	Methods of analyzing benzene and other volatile organic compounds (VOCs) .....	276
9.8.3	Formaldehyde and acetaldehyde analysis methods .....	280
9.9	Dioxin analysis methods .....	283
9.9.1	Introduction .....	283
9.9.2	Sampling .....	283
9.9.3	Analytical operations .....	284
9.9.4	Indication of concentration .....	286
9.10	Simple NO, NO <sub>2</sub> measuring methods .....	287
9.10.1	Introduction .....	287
9.10.2	Sampler construction and collection method .....	287
9.10.3	Sampler preparations .....	288
9.10.4	Sampler assembly .....	288

9.10.5	Collection of nitrogen oxides .....	288
9.10.6	Analysis methods .....	289
9.11	Offensive odor measuring methods .....	291
9.11.1	Overview of offensive odor measuring methods .....	291
9.11.2	Instrumental analysis methods .....	291
9.11.3	Sensory test methods .....	295
9.12	Surveys into the impact on health (epidemiological survey methods) .....	299
9.12.1	Introduction .....	299
9.12.2	Steps of epidemiologically evaluating air pollution .....	299
9.12.3	Actual process of epidemiological survey .....	299
9.13	Field survey of effects on vegetation .....	302
9.13.1	Methods of indexing the appearance of visible damage (plant indicator) .....	302
9.13.2	Open top chamber method .....	302
9.13.3	Phytometer .....	303
9.13.4	Survey method for plants growing in the field .....	304
9.14	Survey on the Impact on Materials and Cultural Assets .....	306
9.14.1	Visibleobservation .....	306
9.14.2	Methods of measuring and analyzing deterioration .....	307
9.15	Meteorological observation measurements .....	308
9.15.1	Introduction .....	308
9.15.2	Surface meteorological observation .....	308
9.15.3	Upper air meteorological observation .....	308
9.15.4	Continuous remote sensing of upper atmosphere meteorology .....	310
9.15.5	Meteorological network .....	310
Chapter 10	Air Pollution Estimating Techniques .....	315
10.1	The Theory of Atmospheric Diffusion .....	315
10.2	Outline of the Model to Estimate Atmospheric Diffusion .....	320
10.2.1	Plume and Puff Models .....	320
10.2.2	The Numerical Simulation Model .....	321
10.2.3	The particle method model of diffusion .....	323
10.2.4	The model of the plume rise .....	324
10.3	Investigation of atmospheric diffusion .....	325
10.3.1	Meteorological conditions investigation .....	325
10.3.2	Investigation of the diffusion condition .....	327
10.4	The Practical Method of Estimation Air Pollution .....	329
10.4.1	Estimating the long-term concentration .....	329
10.4.2	Estimating the short-term average concentration .....	333

10.4.3	Estimating chemically reactive substances .....	334
10.4.4	Estimating from wind tunnel tests .....	335
10.5	The Statistical Forecasting Model .....	338
10.6	CMB Method .....	340
10.6.1	Overview .....	340
10.6.2	The CMB model .....	340
10.6.3	Index elements .....	344
<b>Chapter 11</b>	<b>System of Air Quality Conservation Laws .....</b>	<b>347</b>
11.1	Overview .....	347
11.1.1	Laws and Ordinances .....	348
11.1.2	Conventions, Protocols, and Declarations .....	348
11.2	The Basic Environment Law .....	349
11.2.1	Overview .....	349
11.2.2	Environmental Quality Standards .....	349
11.2.3	Environmental Pollution Control Programs .....	350
11.2.4	International Cooperation .....	350
11.3	Air Pollution Control Law .....	351
11.3.1	Overview .....	351
11.3.2	Smoke and Soot .....	354
11.3.3	Particulate dust .....	359
11.3.4	Motor Vehicle Exhausts .....	359
11.3.5	Controls on Harmful Air Pollutants .....	359
11.4	Law Concerning Special Measures to Reduce The Total Mass of Nitrogen Oxides Discharged by Motor Vehicles in Specified Areas (The Motor Vehicle NO <sub>x</sub> Law) .....	361
11.4.1	Background .....	361
11.4.2	Overview .....	361
11.5	Offensive Odor Control Law .....	365
11.6	Law Concerning the Improvement of Pollution Prevention Systems in Specific Factories .....	365
11.7	Environmental Impact Assessment Law .....	366
11.8	Law Concerning Rational Use of Energy (Energy Conservation Law) .....	366
11.9	Conventions, Protocols and Associated Laws .....	367
11.9.1	Overview .....	367
11.9.2	Conventions and Protocols .....	367
11.9.3	Promulgation of Domestic Laws .....	367
<b>Chapter 12</b>	<b>Administrative Outline of Air Quality Conservation .....</b>	<b>369</b>
12.1	Air Quality Conservation Administrative Framework and Enforcement System .....	369



12.2 Atmospheric Environment Monitoring System .....	372
12.2.1 Overview .....	372
12.2.2 Information Flow in the Atmospheric Environment Monitoring System .....	372
12.2.3 Atmospheric Environment Monitoring System Applications .....	372
12.3 Factory Inspections .....	376
12.3.1 Overview .....	376
12.3.2 Items Targeted for Inspection .....	376
12.3.3 Inspections .....	378
12.4 Environmental Impact Assessment System .....	379
12.4.1 The Process of Establishing the Environment Impact Assessment System .....	379
12.4.2 Environmental Impact Assessment Law and An Overview of the System .....	379
12.4.3 Assessment concerning Air Quality Conservation .....	381
12.5 System of Pollution Control Manager, etc. ....	385
12.5.1 Background Circumstances .....	385
12.5.2 Specified Factories .....	385
12.5.3 Qualification Acquisition .....	385
12.5.4 Job Specifications .....	385
12.5.5 Other Systems Related to Air Quality Conservation .....	387
12.6 Supporting System of Pollution Control Measures .....	388
12.6.1 Governmental Supports for Commercial Pollution Control Investment .....	388
12.6.2 Effects of Supporting Measures .....	389
12.7 Systems of Compensation and Prevention for Pollution-Related Health Damage .....	391
12.7.1 Frequent Incidents of Health Damage Followed by the Yokkaichi Pollution Lawsuit, and the Establishment of the System for Pollution-related Health Damage compensation .....	391
12.7.2 Recognition of the victims and Compensation, and the Expense Bearing .....	391
12.7.3 System Reform Accompanying Improved State of Pollution .....	392
12.7.4 From Individual Patient Compensations to Health Damage Prevention Programs Directed At Entire Areas .....	393

## The List of Authors

Name	Title/Organization	Sections written by the author
Fujimura, Mitsuru	Manager, Environmental Consulting Division, Green Blue Corporation	9.9
Fukuoka, Saburo	Technical Adviser, Suuri-Keikaku Co.ltd.	6.6, 7.11, 8.6
Hara, Hiroshi	Senior Research Scientist, National Institute of Public Health	9.5
Hatakeyama, Siro	Research Program Manager, Center for Global Environmental Research National Institute for Environmental Studies	1.4, 4.2
Himi, Yasuji	Lecturer, Faculty of Life Science, Tokyo University of Pharmacy Technical Adviser Japan Environmental Sanitation Center	2.2, 3, 6.1~6.5, 6.8, 7.1, 7.5, 7.14, 8.1~8.5, 11, 12.1
Hishida, Kazuo	President, Hishida Office Environmental Planning, Senior Technical Adviser, Overseas Economic Cooperation Fond	2.1, 7.2, 7.3
Hori, Masahiro	Associate Professor, Faculty of Education and Human Sciences, Yokohama National University	9.1
Imura, Hidefumi	Professor, Institute of Environmental Systems, Kyusyu University	7.15, 12.7
Kato, Seitaro	Lecturer, Chuo University	7.7, 7.10
Kido, Nobuo	General Research Counselor, National Institute for Resources and Environment	1.3, 6.1, 6.4, 6.5, 7.4, 7.6, 7.8, 7.9
Kitabayashi, Kouji	General Director, National Institute for Resources and Environment	10.1~10.5
Kogure, Nobuyuki	Senior Researcher, Air Quality Measurement Division, Atmospheric Environmental Protection Department, National Institute for Resources and Environment	7.6
Komeiji, Tetuto	Chief Researcher, Applied Research Division, The Tokyo Metropolitan Research Institute for Environmental Protection	5.3, 9.14
Mizohata, Akira	Associate Professor, Research Institute for Advanced Science and Technology	10.6
Mori, Masaki	Senior Adviser, DKK Corporation	9.2
Morishima, Akira	Director, Japan Environment Corporation	12.6
Nakayama, Sinji	Japan Environmental Sanitation Center	5.5, 6.7, 7.13, 9.11
Nihei, Hisao	Technical Adviser, Suuri-Keikaku Co.ltd.	6.8, 9.3, 12.2, 12.3, 12.5
Otoshi Tunehiko	Japan Environmental Sanitary Center	9.6
Samata, Mitsuo	Yokohama Environmental Research Institute	7.12
Sato, Sizuo	Director, Kawasaki Municipal Research Institute for Environmental Protection	9.4, 9.6~9.8, 9.10
Totsuka, Tumugu	Professor, Department of Environment and Information Faculty of Sociology, Edogawa University	5.2, 9.13
Uchiyama, Iwao	Director, National Institute of public Health Department of Occupational Health	5.1, 9.12
Wakamatsu, Sinji	Head, Urban Air Quality Research Team, Regional Environment Division, National Institute Environmental Studies	1.2, 4.1, 4.3, 4.4, 5.4, 9.15
Washida, Nobuaki	Director of the Atmospheric Environment Division, The National Institute for Environmental Studies	1.1

Name: alphabetical order

**The Investigation Committee for  
Manual for Transfer of Air Pollution Control Technology**

Name	Title/Organization
Himi, Yasuji	Lecturer, Faculty of Life Science, Tokyo University of Pharmacy, Technical Adviser, Japan Environmental Sanitation Center
Hishida, Kazuo	President, Hishida Office Environmental Planning, Senior Technical Adviser, Oversea Economic Cooperation Fund
Kato, Saburo	Executive Managing Director, Overseas Environmental Cooperation Center, Japan
Nihei, Hisao	Technical Adviser, Suuri-Keikaku Co. Ltd.

**Cooperated Supervisor**

Name	Title/Organization
Himi, Yasuji (Japanese edition)	Lecturer, Faculty of Life Science, Tokyo University of Pharmacy, Technical Adviser, Japan Environmental Sanitation Center
Saiki, Yoshio (English edition)	Nihon University, College of Science and Technology

# Chapter 1 An Introduction to Air Pollution

## 1.1 An outline of the “Atmosphere”

### 1.1.1 Composition of the Atmosphere

The earth’s “atmosphere” consists of an abundance of chemical elements. Its main constituents are nitrogen (volume ratio 78.1%) and oxygen (21.0%). Water vapor is also highly prevalent in the atmosphere though quantities vary. Other than this, existing in extremely minor quantities, are trace gases. Table 1.1.1 illustrates the chemical make-up of the atmosphere. Should wind shift in the atmosphere be weakly defined, gaseous components would likely undergo a diffusive separation. This is due to the fact that individual elements differ in mass from one another, resulting in heavier elements sinking to lower regions, while lighter elements would rise to regions higher in the atmosphere. However, in reality, due to the existence of wind conditions in the regions below the tropopause, 11 kilometers above ground, the elements are well mixed. The mixing ratio is relatively the same. However traces gases, excluding noble gases, are not equally distributed throughout the atmosphere by source or photochemical reaction within the atmosphere. But rather, their concentrations and distribution differ, effected by such factors as altitude, latitude, longitude, and time of year. Concentration levels also differ depending on place, such as in urban areas.

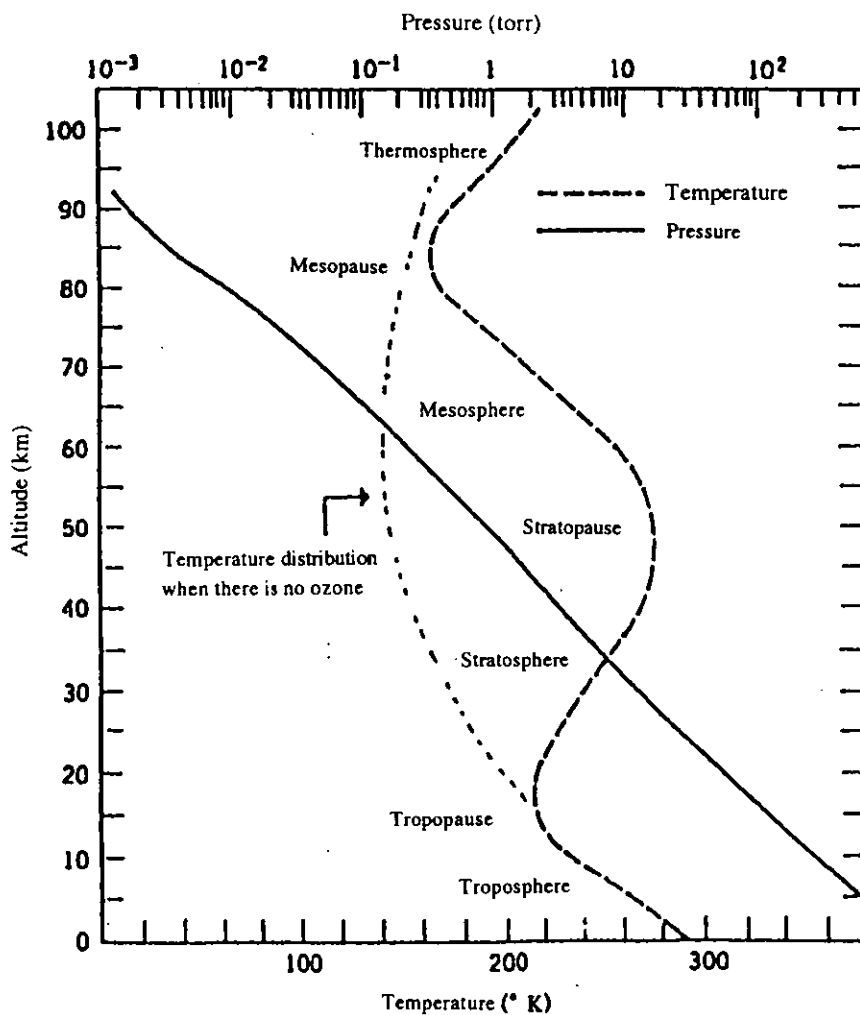
Table 1.1.1 Atmospheric Composition Around the Earth’s Surface

Component	Molecular Formula	Molecular Weight	Existence Ratio (%)	
			Volume Ratio	Weight Ratio
Nitrogen molecules	N <sub>2</sub>	28.01	78.11	75.53
Oxygen molecules	O <sub>2</sub>	32.00	20.96	23.14
Argon	Ar	39.94	0.9343	1.280
Carbon dioxide gas	CO <sub>2</sub>	44.01	0.03	0.045
Carbon monoxide	CO	28.01	1 × 10 <sup>-3</sup>	1.2 × 10 <sup>-5</sup>
Neon	Ne	20.18	1.8 × 10 <sup>-3</sup>	1.2 × 10 <sup>-5</sup>
Helium	He	4.00	5.3 × 10 <sup>-4</sup>	7.3 × 10 <sup>-5</sup>
Methane	CH <sub>4</sub>	16.05	1.52 × 10 <sup>-4</sup>	8.4 × 10 <sup>-3</sup>
Krypton	Kr	83.7	1 × 10 <sup>-4</sup>	3 × 10 <sup>-4</sup>
Nitrous oxide	N <sub>2</sub> O	44.02	5 × 10 <sup>-5</sup>	8 × 10 <sup>-5</sup>
Hydrogen molecules	H <sub>2</sub>	2.02	5 × 10 <sup>-5</sup>	3 × 10 <sup>-6</sup>
Ozone	O <sub>3</sub>	48.0	2 × 10 <sup>-5</sup>	3 × 10 <sup>-6</sup>
Water vapor	H <sub>2</sub> O	18.02	variable	variable

### 1.1.2 Structure of the Atmosphere

The temperature of the atmosphere usually decreases in line with an increase in altitude. The major source of heat for these regions is solar radiation. Solar radiation is generated by the sun’s rays which are initially absorbed into the earth’s surface only to be reflected once again into the atmosphere. This warm air then rises to the upper

layers of the atmosphere. As illustrated in Fig. 1.1.1, air density (pressure) declines toward the upper layers of the atmosphere. As a result, as the air from the lower layers is carried up to higher regions, the energy contained by the elements in the atmosphere decreases due to the effects of adiabatic expansion, causing the temperature to drop. The atmospheric region or shell from the earth's surface to an altitude of 11 km is known as the troposphere. This comes from the fact that temperature distribution is determined by adiabatic expansion in this region of the atmosphere which has many convection currents. Above the troposphere, is the ozonosphere, in which solar radiation occurs. Ozone, plays an important part in the radiative balance of the atmosphere, preventing temperatures from declining by converting sunlight to heat and actually causing the temperature to rise gradually. Atmospheric stability is realized from the standpoint of temperature in regions where there convection currents do not occur, as warm air remains above cool air.



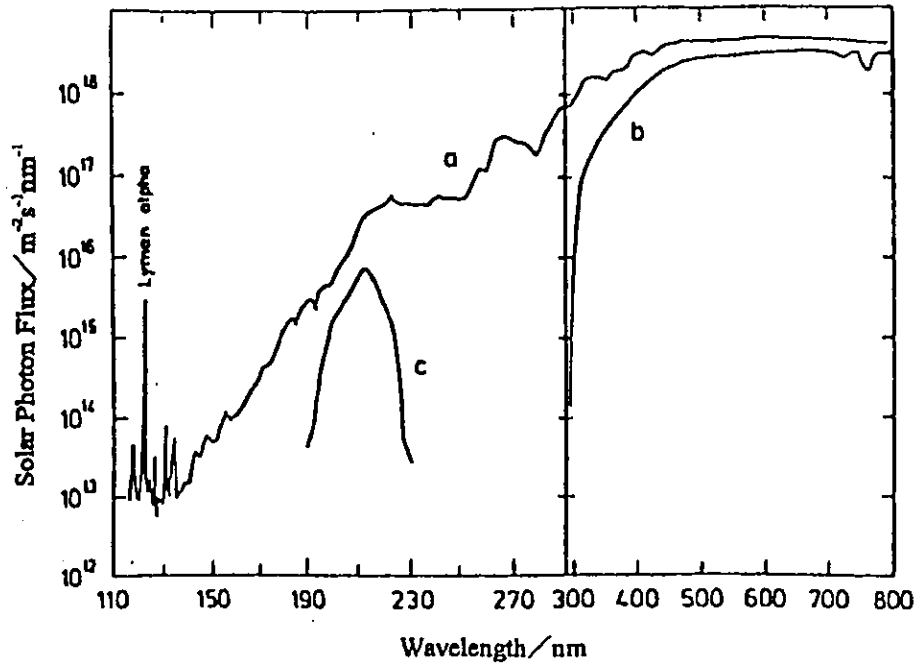
Note: temperature distributions predicted for when no ozone is present.

Fig. 1.1.1 Vertical structure of the atmosphere including atmospheric layer names, perpendicular distribution of temperatures and pressures

This atmospheric region is known as the stratosphere. The tropopause is the boundary between the troposphere and stratosphere where the temperature once again begins to elevate with altitude. Within the stratosphere, temperature is determined by radiation equilibrium, a balance between the ozone working to absorb the sun's UV radiation and cooling caused by infrared radiation. Furthermore, the fact that air density and ozone concentrations decrease at higher altitudes results in a decline in temperature. Temperatures reach maximum levels at an altitude of about 50 km. The altitude at which this temperature reaches its maximum, commonly defines the boundary of the stratosphere, and is the region known as the stratopause. The atmospheric shell above the stratopause is known as the mesosphere and is characterized by a temperature that typically declines with altitude. However, the temperature begins to increase once again at an altitude of nearly 80 km. Above this region is the shell known as the thermosphere. The mesopause exists between the mesosphere and the thermosphere. It is chiefly within the thermosphere that oxygen molecules absorb the sun's UV rays. This, along with the repeated process of photodissociation and recombination are what cause the sun's UV rays to be converted into heat and cause temperatures to rise. It is in this manner that the atmospheric structure is determined by physical and photochemical processes caused by the sun and related energy.

### 1.1.3 Solar Radiation

The sun emits a variety of electromagnetic waves from shortwaves (high energy), such as X-rays and UV rays, to longwaves (low energy) such as infrared, micro, or radio waves. Since the temperature of the sun's surface is 6,000 K, the energy distributed by solar radiation corresponds to that of a black-body at 6,000 K and the intensity maximum around the blue area at 480 nm. Fig. 1.1.2 illustrates the solar flux spectrum in the 120-800 nm wavelength region. The difference between (a) outside the earth's atmosphere and (b) at sea level, shows the absorption by gases in the atmosphere. In this absorption, the solar flux below 200 nm is absorbed mainly by the oxygen molecules and the flux between 200-300 nm is absorbed by the ozone in the stratosphere. The distribution of the solar flux changes depending upon the concentrations of ozone according to the altitude of the stratosphere. Fig. 1.1.2 (c) shows the solar photon flux at 30 km altitude to show the atmospheric window in the 180-220 nm wavelength region, where the valley between the absorption by oxygen below 200 nm and ozone around 250 nm. We can see that the composition of atmospheric trace gases and the structure of the atmosphere are controlled by the photochemical reactions due to solar radiation at 180-220 nm in the stratosphere and above 400 nm in the troposphere. Therefore, when considering the problems of the atmosphere, it is necessary to think of the physical movements in the atmosphere and the chemical (photochemical) reactions in the atmosphere. We recommend the reference materials listed below to those who wish to learn more about atmospheric physics and chemistry.



Note: (a) outside the earth atmosphere, (b) at sea level, and (c) at about 30km altitude to show the atmosphere window in the 185-215 nm wavelength region.

Fig.1.1.2 The solar flux spectrum in the 120-800nm wavelength region

## 1.2 Definition of Air Pollution and Air Pollutants

### 1.2.1 Foreword

When the substances which have been emitted into the atmosphere exist through out the atmosphere, over and above the natural physical diffusion and deposition functions, chemical elimination functions, and biological purification functions, is dispersed throughout the atmosphere and their volume exceeds natural conditions, they have a direct or indirect influence on living beings, including human being. This is called airpollution, air pollutants are generally defined as those substances which alter the composition of the natural atmosphere.

### 1.2.2 The Sources and Types of Air Pollutants

Sources of air pollutants are divided into those which derive from natural causes and those which are brought about by human power. These are called natural sources and anthropogenic sources. Some of the major natural sources include volcanic emissions, forest fires, pollen scattering, sandstorms and yellow sand being blown up from the surface of the earth by winds, sea salt particles being blown up from the surface of the sea by winds, and ozone which has subsided from the stratosphere to the troposphere. Some of the major anthropogenic sources include factories and thermal power stations, substances emitted due to the burning of fossil fuels in engines like those of automobiles, gases and particulate matter created in the production process, and suspended particulate matter and chemical substances created in the process of waste disposal.

Air pollutants are divided into primary pollutants like carbon monoxide, sulfur dioxide, hydrocarbon species, dust and soot, which are emitted directly by air pollutant sources, and secondary pollutants like nitrogen dioxide, photochemical ozone, and aerosols, which are created by chemical changes which occur in the atmospheric environment. Physically, they are also divided into gas particles and particulate matter.

### 1.2.3 Air Pollutants

Environmental quality standards have been laid down for the major air pollutants which have an effect on the health of human beings, and the figures regarding their concentrations are monitored so that they will not exceed these standards. Air pollutants which are controlled by environmental quality standards and guideline figures in Japan are indicated below.

(1) Sulfur Dioxide (SO<sub>2</sub>): Sulfur dioxide is formed when the sulfurous components contained within fossil fuels like coal and oil develop when they are oxidized during the combustion process. Sulfur dioxide brings about air pollution in the region environment, and since it has a long lifetime in the atmospheric environment and can be carried long distances, it has an influence on a wide area as acidic deposition.

(2) Nitrogen Dioxide (NO<sub>2</sub>): Nitrogen dioxide is formed when fossil fuels like coal and oil are burned and the nitrogen



in the air is oxidized to form nitric oxide (NO) and then oxidized again to form nitrogen dioxide (NO<sub>2</sub>). The sum total of nitric oxide and nitrogen dioxides are called nitrogen oxides. Nitrogen oxides contain poisonous gas in themselves, and they are substances which cause photochemical air pollution.

(3) Suspended Particulate Matter (spm): Suspended particulate matter consists of particles of under 10  $\mu$  m in particle size which are suspended in the atmosphere. They come from a mixture of sources including anthropogenic and natural origins, and they are either emitted directly from their source or are secondarily created by being particularized from gas particles in the atmosphere. Suspended particulate matter not only decreases visibility but minute particles in particular have a bad effect on people's health as they coexist with other gaseous air pollutants.

(4) Carbon monoxide (CO): Carbon monoxide is a highly poisonous, gaseous substance which develops from the incomplete combustion of the carbon contained in fuels, and the major source of origin is automobile exhaust gas. Highly concentrated pollution occurs on roads and parking lots.

(5) Photochemical oxidants (Ox): Photochemical oxidants are formed by atmospheric photochemical reactions which occur to nitrogen oxides and volatile organic compounds which are emitted directly from sources like factories, enterprises and automobiles and hydrocarbon species from biogenic sources. Most all photochemical oxidants are ozone (O<sub>3</sub>).

(6) Non-Methane Hydrocarbon Species (NMHC): This is the name given to all hydrocarbon components aside from methane. These substances give rise to photochemical oxidants. Concentrations are expressed as ppmC, using the ppm concentrations for methane conversions.

In general, acidic deposition that is generally called acid rain is also categorized as an air polluting substance. Acidic deposition is divided into wet deposition by rain and dry deposition as gas and aerosols fall directly on the ground.

#### 1.2.4 Other Air Pollutants

Some gaseous air pollutants other than those mentioned above are nitric oxide, gaseous nitric acid, PAN, gaseous fluorine, and hydrogen chloride. Other offensive odorous substances like ammonia, methyl mercaptan, hydrogen sulfide, methyl sulfide, trimethylamine, dimethyl sulfide, aldehyde, and styrene are also considered to be gaseous air pollutants.

In addition to suspended particulate matter, dust fall is also considered to be particulate air pollutant. It is the volume of dust fall, and it's also called particulate fall which falls in units of time on units of area. Some natural sources are soil (including substances like yellow sand) which is scattered from the ground, volcanic emissions, sea salt particles, and biological sources like hydrocarbon substances which have been particularized in the atmosphere. Artificial sources include substances which have been emitted in factory production processes (including combustion),

automobile emissions, substances which have blown up off roads by traffic, and substances from the incineration of waste materials.

#### 1.2.5 New Air Pollutants

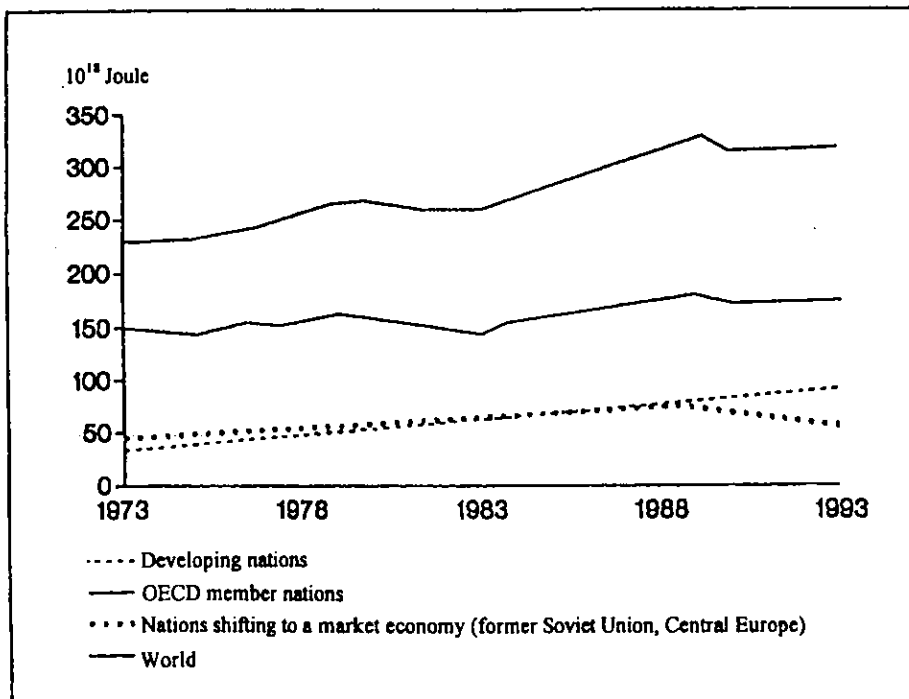
Today, we are facing new issues related to environmental pollution. One is the problem of the global environment. Another is the problem of harmful chemical substances. Rapid increases in the consumption of fossil fuels are causing increasing concentrations of carbon dioxide in the atmosphere and contributing to global warming. The volume of ultraviolet rays which reach the ground surface is also increasing due to the fluorocarbons which are destroying the stratospheric ozone layer. Rises in temperature and increases in ultraviolet rays lead to the increase of photochemical air pollution, and because of this, the pollution situation is worsening in metropolitan areas. Carbon dioxide and fluorocarbons on the ground level are not air pollutants in and of themselves, but they can be called indirect air pollutants. On the other hand, harmful trace pollutants have also become a major issue. Highly cancer-causing substances are included among these, and obtaining information about their dynamics in the environment and conducting risk assessments are of vital importance at the present time. One major characteristic of these new forms of air pollution is that there are no clear distinctions, as there were before, between the polluters and the pollution victims. In other words, the major characteristic is that instances of urban air pollution where an unspecified large number of citizens are both polluters and pollution victims have come to command the majority of pollution cases. Various air pollution problems are becoming even more serious in major metropolitan areas, in particular, due to new population concentrations in the cities and to the changes in lifestyles which accompany this phenomenon, and automobile pollution problems are becoming worse than ever. These problems give rise to not only air pollution from nitrogen dioxide, photochemical ozone, and suspended particulate matter, but also harmful chemical substances from things like benzene. The incineration of plastics which have been thrown away as urban refuse has also given rise to highly carcinogenic substance like dioxins.

## 1.3 Air Pollution and the Issue of Resources

### 1.3.1 Energy and Air Pollution

Air pollution is brought about by substances like sulfur oxides, nitrogen oxides, soot and dust, hazardous substance, dust, carbon monoxide, and hydrocarbon species emitted from stationary source like factories and mobile sources like motor vehicles. Most air pollution is accounted for by combustion reactions to fossil fuels, but much air pollution caused by improvements made in combustion has been eliminated. Air pollution problems are closely related to problems with energy resources, and those material industries which directly employ these resources have become a major issue.

Energy consumption in developing nations, as shown in Fig. 1.3.1, has been increasing rapidly in recent years. Energy consumption levels in 1993 were nearly three times those in 1973. Emissions of air pollutants like sulfur oxides and nitrogen oxides have also increased along with the increases in energy consumption levels. To be more specific, there are countries which do not have flue-gas desulfurization equipment and flue-gas denitrification equipment, countries like China which are generating grave air pollution problems, and countries which, if left alone, may develop problems in the future.



Note: Indicates OECD member nations and economic cooperation and development system members.

Source: United Nations (U.N.) Statistical Division, 1993 Energy Statistics Yearbook (U.N., New York, 1995)

Fig. 1.3.1 World Energy Consumption, 1973-1993

In order to promote air pollution control measures in developing countries, steps must be taken which are in accord with the energy situation in each particular country. It must be noted that most developing countries are

expected to have increases in the consumption of coal, which is the richest of all fossil fuels. For example, China, which uses low quality coal containing high quantities of sulfur for 80% of its primary energy, has experienced serious damage due to acid rain.

Next, it must also be mentioned that the implementation of energy conservation measures are becoming most important for controlling air pollution. The implementation of energy conservation measures not only helps to conserve valuable energy resources, but the emission levels of air pollutants can be controlled by curtailing fuel consumption. In countries which do not have highly effective ways to use energy, there are more opportunities than in Japan to apply energy-saving technology, and if implemented, the results will be all the greater. It is hoped that energy conservation technologies like starved air combustion and waste heat recovery will become more widespread.

### 1.3.2 Air Pollution Control and Resource Recovery

Many of the flue-gas desulfurization facilities in Japan use methods which take the environment and resources into consideration. Most of these facilities utilize the abundant, good-quality limestone available in Japan in a process called the lime-gypsum method, which neutralizes and stabilizes the crude oil and large quantities of sulfur which are brought into the process in order to create the secondary product, gypsum, a resource which is rare in Japan and is used to produce nearly 2,300,000 tons of cement retarder, gypsum board, plaster, and calcinated gypsum per year.

At the present time, however, China has no prospects for a demand for such desulfurized gypsum, Korea does not have sufficient quantities of good-quality limestone, and the United States and Thailand, which have abundant gypsum resources, have not progressed much with applications of secondary desulfurized gypsum. Flue-gas desulfurization has not become very widespread in China, but if China were to develop methods to both reclaim sulfur oxides and nitrogen oxides at a low cost and obtain high-quality fertilizers like ammonium sulfate and ammonium nitrate and agricultural soil conditioners, then they would be able to kill two birds with one stone and stimulate improvements in agricultural production at the same time.

Treatment of the ash which derives from the coal combustion has also become a problem, and it is hoped that more effective utilization technologies will be developed because of the difficulties in securing disposal sites. It is effective to make practical use of desulfurized gypsum as a material resource, and it is being used as raw material for products like cement, cement compounds, roadbed materials, artificial light aggregates, fertilizers, snow melting materials, and materials for man-made gathering-places for fish, but more technological development is needed to make use of the vast quantities of limestone available. Fly ash is already being used in Japan as raw material for cement.

### 1.3.3 Resource Development and Environmental Issues

Active development of resources has accompanied economic progress in many countries, but there is a growing need for each of these countries to make further considerations for the environmental conservation. For example, in the processes of probing, development, operations, refining, and mine closure in the development of metal

resources, the world is now calling for resource development and environmental pollution control measures which take the environment into consideration. Most necessary are resource development plans which consider everything from the treatment of waste water after mine closures to post-closure measures and measures to restore land in regions undergoing development.

#### 1.3.4 Securing and Treating Water for Industrial Use and Transportation Issues

Japan possesses an enormously long coastline and securing water for industrial use has not been difficult. Accordingly, there have been few problems operating wet flue-gas desulfurization facilities which require vast amounts of industrial-use water and limestone. In countries which have most of their factories located inland, however, it has been difficult to secure sufficient industrial-use water at times and it has not been easy to transport the limestone and secondary gypsum, so it has been difficult to increase the number of wet flue-gas desulfurization facilities like those in Japan. An enormous amount of energy is required for drainage water treatment and operations even if the water used is circulated.

#### 1.3.5 Environment and Resource Issues

The above air pollution problems are deeply related to the problem of resources, and it is necessary to implement these measures so as to keep in line with the actual situations of resources and technology in the source regions. International cooperation is necessary in air pollution control measures, but it is even more necessary to have an understanding of the resource issues faced by each region.

## 1.4 Global environmental problems

### 1.4.1 Introduction

As global environmental problems concerning air, we have global warming, depletion of the ozone layer and acidification of the tropospheric air (so-called acid rain). It was when the public at large were confronted with these problems that they started to be aware of global environmental problems in general.

### 1.4.2 Global warming

Global warming is the phenomenon in which, as human activities have expanded, some components of the air existing in extremely small quantities (such as carbon dioxide generated in consumption of fossil fuels, methane, ozone and fluorocarbons) have increased so much that they have come to trap and accumulate more heat within the atmosphere than the old green-house gases (so called because they prevent heat from escaping out into space by absorbing infrared rays reflected from the earth's surface, like the glasses of the green house) including vapor did formerly, resulting in increased atmospheric temperatures on the average. The Intergovernmental Panel on Climate Change, (IPCC), forecasts that, if the new green-house gases continue to increase at the current rate, the average air temperature of the earth will rise by about 3°C by the end of the 21st century. Fears are entertained that this will translate into various unfavorable effects upon environment including the rising surface of the sea and less rain/drought in some regions of the earth. In addition to these global problems, it is apprehended that the rise in air temperatures will produce effects seriously affect the civic life such as aggravation in air pollution by photochemical ozone<sup>1)</sup>. It can be said that its effects will be truly variegated.

Among the extremely small components of the air contributing to global warming, the most problematic is carbon dioxide (CO<sub>2</sub>) generated when fossil fuels are burned. Methane and nitrous oxide (N<sub>2</sub>O) follow. Recently, it has been pointed out that ozone within the troposphere is also important. Fluorocarbons, therefore, play a significant role in causing not only the problem of the stratospheric ozone layer but global warming. Although such miscellaneous tiny components of the air as methane are in lower concentrations in comparison with vapor or carbon dioxide, they have nonetheless high capacities for enhancing global warming because they absorb infrared rays of other wavelength ranges than that absorbed by vapor or carbon dioxide (so-called window ranges). However, more researches should be necessary because, even when narrowing down to carbon dioxide alone, there are still other problems whose effects upon concentration of the substance are yet to be clearly determined, such as deforestation and the problem of CO<sub>2</sub> absorptivity of the sea, than the burning of fossil fuels.

As major part of international efforts to cope with global warming, IPCC mentioned above has issued this latest finding about forecasts, assessed effects and measures to be taken: measures to counter global warming, therefore, have been given the scientific basis. Also in 1992, the "United Nations Framework Convention on Climate Change" was concluded to promote international measures to check global warming. For the purpose of stabilizing the concentration of green-house gases in the air, the signatory powers to the convention have been saddled with the responsibility for preparing inventories of emissions/absorptions of green house gases, formulating and

executing a state program of coping with global warming and so on.

#### 1.4.3 Depletion of the stratospheric ozone layer

The ozone layer in the stratosphere has been brought into existence as oxygen, which has since ancient times been produced by plants through their photosynthesis and accumulated in the air, has shown photochemical reactions in the stratosphere. It has served as a precious barrier protecting living things on earth against high-energy ultraviolet rays. That this ozone layer is in danger of being depleted by chlorine atoms produced as fluorocarbons, which are emitted by human beings, undergo photolysis in the air was pointed out for the first time in 1974 by Molina and Rowland<sup>2)</sup>. Later when it was ascertained that the ozone hole above the Antarctic Zone had occurred and expanded as the result of this reaction of ozone, people on the earth were all shocked. Fears have since been entertained that, when the ozone layer above is depleted, ultraviolet rays of shorter wavelengths that have higher energy should be poured straightly down, not only affecting human bodies directly by increasing the incidence of cutaneous cancer and cataract but damaging all animals and plants in the region seriously.

International endeavors to seek to protect the ozone layer started relatively early. In 1985 the "Vienna Convention for the Protection of the Ozone Layer" was concluded and in 1987 the "Montreal Protocol on Substances that Deplete the Ozone Layer" was adopted. This protocol contained regulatory measures such as the one to cut productions of substances depleting the ozone layer. However, it was reviewed three times later in 1990, 1992 and in 1995 because it came to be known that depletion of the ozone layer was progressing at a rate greater than initially expected as evidenced by, for example, the rapid expansion of the ozone hole. The regulations have been strengthened by stages, consequently, with new entries having been made in the list of substances to be regulated and the regulation schedule having been accelerated. Recently, it has been observed that under these regulations concentration of fluorocarbons in the air has been on the decrease.

#### 1.4.4 Acidification of the tropospheric air (acid rain)

While increasing quantities of fossil fuels have been consumed on the earth, also expanding quantities of sulfur oxides, or SO<sub>x</sub>, and nitrogen oxides, or NO<sub>x</sub>, as part of the exhaust gas from the burning fuels have been emitted into the air. Also, as NO<sub>x</sub> and hydrocarbon species, which have been emitted together, have shown photochemical reaction, radicals, ozone and other active species called oxidants have been generated that are very highly oxidizing. By oxidizing sulfur dioxide and NO<sub>x</sub> in the air, they have generated sulfuric acid and nitric acid. Consequently, the tropospheric air has become very much more acid than before the industrial revolution. When these acid substances dissolve in water drops in the air and fall onto the earth as rain, it is the phenomenon of the so-called acid rain. It is considered that acid rain has acidified rivers, lakes and marshes, damaging aquatic life living in them, and that it has also affected forests and cultural properties and other artificial constructions.

Of the substances causing the acidification of the air, SO<sub>x</sub> originate in sulfuric contents of coal and heavy oil. Therefore, measures to reduce their emissions can be comparatively realistic. However, in countries such as China where huge quantities of coal are used and where it is not easy to switch to low-sulfur light oil or natural gases, a

long-sustained effort should be needed in the future in coping with the problem. In the case of NO<sub>x</sub> emissions, on the other hand, we have been yet very much behind in our devising and executing measures. Particularly in advanced industrialized countries/regions including Western countries and Japan, it is expected that NO<sub>x</sub> measures will be the focus in settling the problem of acid rain.

It can be said that acid rain is a continental rather than global problem because the phenomenon occurs in areas a little smaller than those affected by global warming or the problem of the ozone layer. Therefore, international efforts to tackle acid rain have been made by individual continents. In Asia, the efforts have been made mainly in the East Asia, where Japan and Korea, China, Taiwan and other newly industrialized countries/economies concentrate. One of them is the "Acid Rain Monitoring Network in East Asia" advocated by Japan's Environment Agency.



#### (1.1) References

- 1) P. Warneck, *Chemistry of the Natural Atmosphere*. Academic Press (1987)
- 2) R. P. Wayne, *Chemistry of Atmospheres*. Claredon Press (1991)
- 3) B. J. Finlayson-Pitts and J.N. Pitts, Jr., *Atmospheric Chemistry: Fundamentals and Experimental Techniques*. John Wiley & Sons (1986)
- 4) J. Heicklen, *Atmospheric Chemistry*. Academic Press (1976)
- 5) Okabe, H., *Photochemistry of Small Molecules*. Wiley Intersci (1978)
- 6) Shimazaki, Tatsuo, *Stratospheric Ozone*. Tokyo University Press (1989)
- 7) Washida, Nobuaki and Hajime Akimoto, *Kagaku Sosetsu No. 39: Photochemical Reactions in Gas Phase: Inorganic Photochemistry*, Japan Chemical Association Edition. Gakkai Publishing Center (1983), pp.14-38.

#### (1.4) Literature cited

- 1) Hatakeyama, S., Akimoto, H., and Washida, N.; *Environ. Sci. Technol.*, 25, 1884-1890 (1991)
- 2) M. J. Molina and F. S. Rowland, *Nature*, 249, 810-812, 1974

## Chapter 2 History of Air pollution

### 2.1 Historical Review of Air pollution around the World

#### 2.1.1 Introduction

Adverse consequences to the human body due to air pollution were already of evident concern in 14th century England. People's lives were maligned by dirtied skies, a direct consequence of increased coal usage due to England's industrial development during the era and also resulting from methods used for fueling household heaters. For this reason, in 1306, craftsmen were banned from burning coal in their furnaces.

Table 2.1.1 Eminent Episodes of Air pollution

	Meuse (Belgium) 1930 (Dec.)	Donora (USA) 1948 (Oct.)	London (UK) 1952 (Dec.)	Los Angeles (USA) 1944~present	Pozarica (Mexico) 1950 (Nov.)
Environment	Ravines Calm condition Temperature - inversion Generate haze Factory district steel works 3 metal works 3 glass works 4 zinc works 3	Ravines Calm condition Temperature - inversion Generate haze Factory district steel works power line factory zinc works sulfuric acid plant	Rivers/flatland Calm condition Temperature inversion Generate haze Humidity 90% Densely populated Cold odor smog	Coastal basin Temperature inversions and ocean mist take place almost daily throughout the year. Generate white haze Sudden increase in - population Expansion in number of - cars More consumption of - petrol-based fuels	A large amount of hydrogen sulfide gas was leaked into the town from an operational accident at a gas plant. Temperature - inversion
Damage	10 times of the normal death rate; along with the death of 60 people. occurrence of acute irritated respiratory disease in all ages. main symptoms include coughing and difficulties in breathing, lethal damage to cattle, birds, and vegetation. fatalities showed signs of chronic heart and lung disease	Among a population of 14,000: Serious illness 11% Medium degree 17% Mild illness 15% Of this 18 who suffered symptoms of lung irritation died, others suffered chronic heart and lung disease, most complained of cough, difficulty in breathing, feeling of constriction in the region of the arm	Excess death rate of 4,000 people in 2 weeks, following this 8,000 died within a 2 month period. Sudden increase in frequency of hospitalization of patients of all age, suffering heart and lung disease, severe cases were seen in particular for those over 45 years old, fatalities included those who suffered such symptoms as chronic bronchitis, asthma, dilation of the bronchial tubes, and pulmonary fibrosis	Continued, recurrent irritation of the mucous membrane in the eyes, nose, air way, and lungs. Displeasure in everyday activities (entire population), damage to cattle, vegetation fruits, rubber products and buildings	Of 22,000 people 320 suffered from acute toxic poisoning, 22 died, coughing, difficulties in breathing and irritation of the mucous membrane were some of the major complaints
Causative Substances	Sulfur dioxide (SO <sub>2</sub> ), sulfuric acid (H <sub>2</sub> SO <sub>4</sub> ), and fluorine compounds from factories. Carbon monoxide (CO), fine particles, etc.	Sulfur dioxide (SO <sub>2</sub> ) and mixtures with sulfuric acid fine aerosols emitted from factories.	Sulfur dioxide (SO <sub>2</sub> ) from coal fuel. 60% came from home heating systems, the rest from factories, and power generators Fine aerosols, dust, etc.	SO <sub>2</sub> , SO <sub>3</sub> , NO <sub>2</sub> , aldehyde, ketone, acid, aromatic and olefinic hydrocarbons, acrolein, formaldehyde, ozone, nitroolefin, etc. from petrol-based fuel combustion.	hydrogen sulfide

The word smog, frequently used in describing a type of air pollution, is a compound derivative from the words smoke and fog, and was first used in Glasgow, Scotland. In 1909, an incident causing an excessive death rate of 1,063 people was reported, presumably a result of the mixture of smoke and soot from coal combustion and fog. Since then, the most infamous case of smog occurred in London in 1952.

Prior to this, in Los Angeles, California in 1947, the onset of photochemical smog brought on the implementation of various countermeasures to resolve the dilemma, but a permanent resolution to the problem has yet to be found.

Table 2.1.1 illustrates several air pollution episodes of major significance which have occurred around the world, including those incidents previously mentioned.

### 2.1.2 England <sup>1)</sup>

Following the Industrial Revolution, large amounts of coal began to be used in England, contributing to the progression of air pollution. In London, the number of patients with respiratory disease increased, and by the end of the Second World War an excess death rate of 4,000 people due to smog was reported in December 1952 when the social activity was popular (on average the ordinary death rate was around 300 people per day). This smog continued for a period of five days from December 5 to 9. A death rate several times higher than the average was recorded, citing symptoms such as senility due to advancement in respiratory illnesses. Even after the smog had dissipated the high death rate continued for some time. The cause was the synergism of such substances as sulfur oxides (sulfur dioxide), soot and dust, fine aerosols (particle substances), and dust, released into the atmosphere from coal combustion of such sources as household heaters, factories, and power generators.

It was due to this "London Smog Incident" that England passed laws (Clean Air Act) which went so far as to regulate home heating methods. Fig.2.1.1 illustrates the correlation of London's smog and the mortality rate.

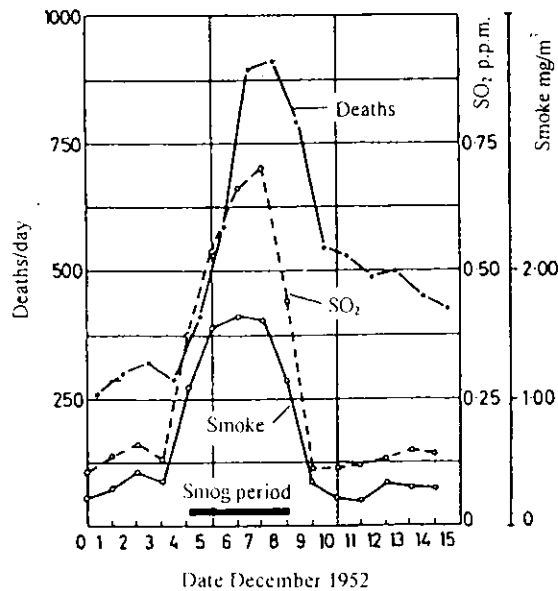


Fig.2.1.1 Correlation between London Smog and Mortality Rate

As seen here, the peak concentration of sulfur dioxide on average was around 0.1 ppm but this rose to 0.7 ppm during the December period. The normal amount of soot and dust was on average about 0.2 mg/m<sup>3</sup> but surpassed 1.7 mg/m<sup>3</sup> during the same period. For either of these cases, pollution concentration was 7 to 9 times that of normal conditions.

### 2.1.3 America

The following are major examples of air pollution in the United States.

#### (1) Donora City, Pennsylvania <sup>2)</sup>

The city of Donora in Pennsylvania, located 30 miles south from Pittsburgh, is an industrial metropolis located on the banks of the Monogahela River. Major industrial facilities which are representative of the area are steel and steel wire factories, also zinc refineries and sulfuric acid factories. In particular, during the period of October 27-31, 1948, a period which saw stable weather conditions, air pollution worsened resulting in a vast portion of the population falling ill and ending in 20 deaths. As in the 1952 London Smog case, a clear cause of death was not confirmed. In the cases of both of these cities, attention was brought about to the abnormally high levels of sulfur compounds (SO<sub>2</sub>, SO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, and inorganic sulfates), contained in the atmosphere.

#### (2) Pittsburgh, Pennsylvania <sup>3)</sup>

In 1948, the nickname "City of Smog" was fit for Pittsburgh. Black soot and smoke darkened the afternoon skies, and newly constructed buildings were blackened and dirtied within a matter of 2-3 months. This became a major social issue, finally leading to the passage of laws regulating soot and smoke. These laws prohibited the use of highly volatile solid fuel (however, in cases where there was sufficient mechanical facilities, these restrictions did not apply). Steam locomotives were transformed to run on diesel, and regulations were constituted to minimize dust incineration. As a result, the environment became clean beyond recognition and the Department of Public Health calculated that the ratio of decline in soot and smoke from 1945 to 1953 was 70%.

#### (3) Los Angeles, California <sup>4)</sup>

The most famous smog in the US is most likely that found in the city of Los Angeles. The area is located in a basin facing the coastline. The regional climate is such that ocean mist and temperature inversion are year-round factors. Meanwhile, the population in the Los Angeles area has grown rapidly in recent years. The population in 1920 was less than 1 million but rose to 2.86 million in 1940, and exceeded 6 million as of January 1958. Accompanying the development of industry and an increase in automobiles in the area was the inconceivable onset of environmental pollution. In 1947, Los Angeles was established as a region aimed to control air pollution, and smoke and soot and sulfur dioxide regulation law was passed. As a result, pollution in the area was slightly curbed. However, eye irritation and damage to vegetable crops increased, visibility gradually grew worse and the amount of peroxides found in the atmosphere increased. It was discovered that this condition was due to a photochemical reaction mainly of hydrocarbon species and nitrogen dioxide caused by the sun's rays. Hydrocarbon

species and nitrogen dioxide are found largely in gas which is exuded from internal combustion engines. There is a huge number of automobiles in Southern California, and this is thought to be the chief reason behind the smog. Currently a method to reduce smog is being devoutly sought.

The characteristics of air pollution in US cities other than those mentioned above were investigated, and the movement to establish laws to prevent air pollution became more active. In 1955, Stern <sup>5)</sup> used calculations based on surveys taken in the state of New York, and came up with the speculation that there were about 1,000 cities in the US suffering from air pollution. Of course, city limits alone do not serve as political boundaries for air pollution. Not just one city, but also its surrounding cities frequently suffer from the same problem.

In 1955, the US government drew up the Air Pollution Control Law. It aided the research of air pollution control in states and regional areas and it cooperated in establishment the environmental standard for the purpose of air pollution control program. In 1967, environmental standard were adopted in 12 of the US's chief metropolitan cities for the purpose of air pollution control program.

#### (4) New Orleans

Around 1953 in New Orleans, the emergency room records at a charity hospital indicated an abnormal increase in the number of patients undergoing medical treatment for asthma. The cause was stated to be the burning of shrubs and weeds, pollution from the city's incinerators, and grain dust from lifts used to move grain. A clear conclusion has not been released.

#### 2.1.4 Air pollution in the Megalopolises around the world <sup>6)</sup>

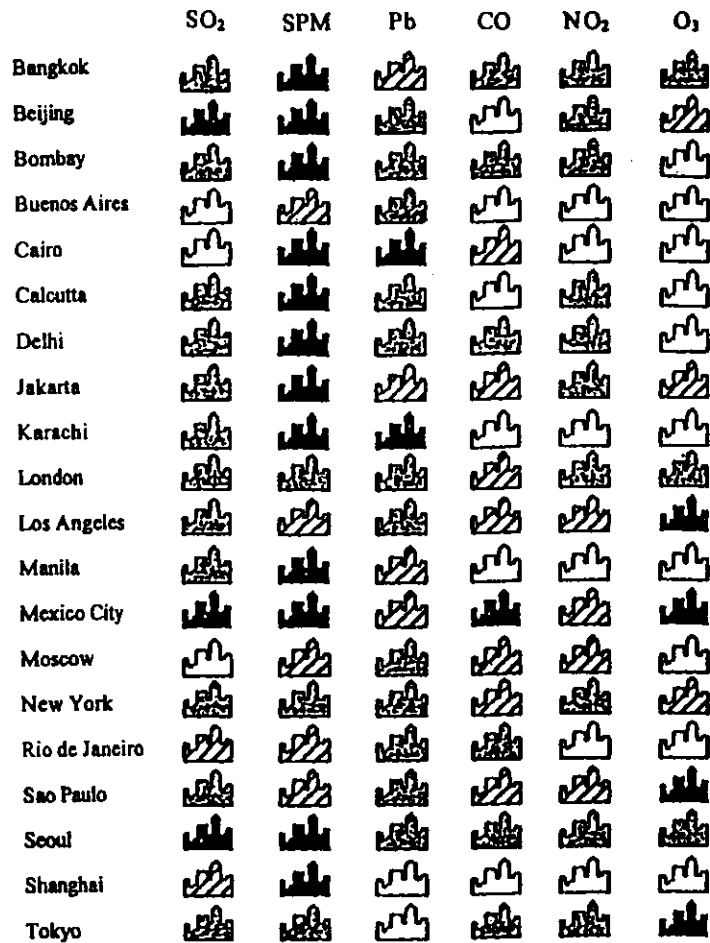
WHO has put out a set of guidelines in order to protect the health of each and every one of us. These guidelines set standards depending on several averaged out timeframes, as there is a difference in the chronic and acute effects caused by air pollution. The WHO guidelines are indicated in Table 2.1.2.

Table 2.1.2 WHO Guidelines

Air Pollution Substances	SO <sub>2</sub> (ppm)	SPM ( $\mu$ g/m <sup>3</sup> )	Pb ( $\mu$ g/m <sup>3</sup> )	CO (ppm)	NO <sub>2</sub> (ppm)	O <sub>3</sub> (ppm)
Average Annual Value	0.017	(60~90)	0.5~1.0			
Monthly Average Value						
24 hours	0.04	70 (150~230)			0.08	
8 hours				9		0.05~0.06
1 hour	0.12			26	0.21	0.075~0.1
30 minutes						
15 minutes				87		
10 minutes	0.17					

Note: ( ) = TSP

Fig. 2.1.2 illustrates a comparison between the air pollution conditions in 20 of the world's megalopolises and the WHO guidelines. This data was taken during the period of 1988-1990.



- High Concentration Pollution.  
Exceeds twice the limit set in WHO guidelines.
- Pollution above Medium Density:  
Less than twice the WHO guideline limits but still exceeding the limit.  
(Exceeds specified point of WHO short-term guidelines)
- Low Density Pollution:  
Nearly meets the WHO guidelines  
(sometimes exceeds short-term guidelines)
- Was not able to evaluate data.

Fig. 2.1.2 Air Pollution Conditions in 20 Megalopolises

## 2.2 Historical Retrospection of Air Pollution in Japan

The following classifies the history of air pollution in Japan into three stages; the first starting from the industrialization of Japan by the Meiji government and lasting up to the end of the Pacific War; the second extends from the industrial restoration to the environmental crisis; and the third stage indicates the time period following thereafter.

### 2.2.1 Air Pollution, the First Stage

#### (1) Development of Industrialization and the Occurrence of Air Pollution <sup>1) 2)</sup>

The industrialization policies of the Meiji government were supported by the potential technology of the Edo period and led to its rapid progression. For example, the popularization of western-style buildings and plans for the additional building of docks for the Yokosuka Iron Works, founded in the last days of the Edo period, led to a forecast of an increase in the demand for cement. This led the government to plan to shift over to domestic production of cement. In May 1875, the former Ministry of Industry's Fukagawa Seisakuryo succeeded in putting into operation a wet method cement kiln. However, it resulted in amassed complaints from citizens due to scattered dust.

After the end of the Edo period, Takatoh Oshima began the operation of a blast furnace which used charcoal in Kamaishi, this became the foundation for Japan's steel manufacturing industry. This also led to the founding of Yawata Steel Mill in 1911. Kamaishi and Yawata became a steel producing cities and eventually began to experience air pollution. At Besshi Coppermine, a refinery was moved to Niihama around 1882 and western refinery methods were adopted. These events also took place at Ashio, leading the historical disputes between the copper industry and farmers, which had erupted due to exhaust gas emitted from the refineries, to repeat themselves. Other industries also began to modernize and prosper, resulting in more emphasis to be placed on air pollution. As in the case of the reclamation done by Soichiro Asano and others along the coastal areas of Tsurumi and Kawasaki in Kanagawa Prefecture in the 1900's, Japan's industrialization was characterized by coastal industrial areas built on reclamations. By the time of the first World War the main stage was set for the initial phase of air pollution in Japan's four major industrial areas, Hanshin, Keihin, Chukyo, and Kita Kyushu.

#### (2) Local Air Pollution Disputes <sup>1) 2)</sup>

Long term disputes over agricultural damage caused by refinery exhaust gas became prominent. Disputes over local pollution caused by new factories became frequent. A typical example of the aforementioned is the case of the Besshi Copper Mine dispute. A private railway was completed to travel between Mount Besshi and Niihama. As the amount of copper production increased, damage to rice paddies materialized in four such as Niihama in 1893. Eventually farmers began to hold demonstration seeking the halt or relocation of the refinery. Negotiations were held between the refinery and farmers but in the following year, 1894, damage to the wheat harvest recurred disputes. In 1905, the refinery was completely moved to an uninhabited island, Shisaka Island, 18 km off the shores of Niihama in the Inland Sea. However, after the refinery began full-scale operations on the island, damages to wheat and rice harvests resulted in agreements which was entered into regulating such items as and the payment of

compensation to farmers the quantity of copper production. And these battles during 47 years come to close due to the production of sulfuric acid from dilute sulfur dioxide in exhaust gas adopting Petersen process in 1929, and the operation of the desulfurization plant for exhaust gas using ammonia in 1937. Also, in the latter case, there were such issues as claims from farmers regarding hydrogen chloride problems caused by the start of the manufacture of monosodium glutamate by the Suzuki Seiyaku company in 1900's Zushi, Kanagawa Prefecture, and the dust issue in Fukagawa caused by the aforementioned cement plant. Because the outbreak of west-south war between political faction in Japan led to tight financial situation, Meiji Government sold the cement factory at Fukagawa operating by Ministry of Industry to Soichiro Asano in 1877. The dust dispersion problem caused by this cement factory operated by Soichiro Asano surfaced as a result of adaptation of rotary kiln imported from U.S.. A relocated petition of factory by citizens of Fukagawa area was approved in 1908. Thus in 1918, Asano Cement has imported the electric dust collector from U.S. investigated by Professor Cottrel and succeeded to retract citizen's petition who recognized the device's effectiveness. However, this did not stop the uprising by citizens opposing plans for the construction of a factory in Daishi Kawahara-village, Tachibana-county, Kanagawa Prefecture. The Asano Cement Kawasaki factory which began operations in 1917 in the village of Tajima in Tachibana-county, also faced repeated long dispute until 1930, immediately following the start of its operations.

### (3) Regional Scale Air Pollution <sup>1)2)</sup>

Industrial cities were posed by the problem of long range air pollution, resulting in the scale of the problem to extend beyond the realms of mere local disputes. Initially, the first time the smoke and soot problem arose in Osaka was during 1883-1884. Osaka Prefecture passed laws cracking down on industrial smoke and soot due to coal combustion. Also, in 1888, smoke and soot emitted by Osaka Electric K.K caused the prefectural government to pass an order "banning the construction of smoke releasing factories within city limits". Osaka prefectural assembly proposed to the governor to control smoke and soot, and in 1910, the prefectural police ordered all public baths to install smoke consuming devices. During the same year, a research group on the prevention of soot and smoke was established with the prefectural governor as its chairman. Osaka City requested funds in 1912 in order to take measures against the smoke and soot emitted from the city-run thermal power plant. Following this, there were continued complaints from mass media were heard against the factory's soot and smoke. However this was to no avail. The prefectural governor sought advice from it's research group on the prevention of soot and smoke and also from the Osaka Chamber of Commerce in order to develop countermeasures, but the research group became defunct in 1917. The smoke and soot problem in Osaka intensified during the First World War. This issue also began to be taken up in Tokyo. In 1924, the Association on Fuel expressed the necessity for smoke prevention for the sake of the city's appearance, the health of it's citizens and in order to prevent squandering fuel. This led the Ministry of Home Affairs to issue an ordinance, in December 1924, to regulate soot and smoke emissions in six major cities. In 1922, the Osaka City Institute of Public Health (currently the Osaka City Institute of Public Health and Environmental Science) began a study on regional scale air pollution. In 1932, the Osaka prefectural government issued an order stating regulations on the prevention of soot and smoke emission. It banned the black smoke emission exceeding a level 3 on the Ringelman's charts over 6 minutes per 1 hour. Similar measures were instituted in 1933 in Kyoto and in 1935 in Hyogo Prefecture. In the Tokyo metropolitan area, local battles related



to factories began early on. In 1927, the Tokyo City Research Laboratory of Public Health began a large-scale study on regional scale air pollution. There are only a few examples of older records concerning the air pollution in the Keihin Industrial Area. Of these, the following reports were found. In 1916, damage to the area's vegetation was observed due to pollutants released from the Hodogaya Soda factory. In 1918, Yokohama (Gyoyu) Fish Oil K.K. was cited for problems concerning soot, smoke, and offensive odor. In 1919, Nihon Hiryo (Japan Fertilizer) K.K. was responsible for releasing offensive odors into the air. In 1922, Yokohama Chemical, Nippon Chemical, and Dai-Nippon Artificial Fertilizer were the cause of damages due to toxic gases. Vegetation in the Kawasaki area was affected in 1929 because of Tokyo Electric K.K. In 1938, NKK Corp.'s operation of a Thomas converter resulted in the emission of iron oxide fumes into the air. In 1935, the Tokyo Prefecture created an outline of guidelines on the prevention of soot and smoke. Beginning in 1937, Kanagawa prefecture implemented policies after deciding on provisions for a "committee on the prevention of soot and smoke". During the same year, the Kanagawa Prefectural Assembly took up the issue of air pollution. In 1940, the Kawasaki City submitted a written opinion to the Governor of Kanagawa and the Minister of Home Affairs, documenting claims by citizens who suffered from the soot and smoke released by the Keihin Industrial Area, in particular symptoms such as adverse effects on the health and hygiene of school children and growth deficiencies in infants. In Kyushu Area, from the latter half of the Meiji era (1868-1911) industries in the area began to develop based on the use of coal as a fuel source. Yawata Steel Mill was the main force behind the prosperity of the Kita-Kyushu Industrial Area. The Omuta Industrial Area, where the problem of air pollution had existed even prior to the Second World War reached its peak of prosperity due to the Miike coalfield.

#### (4) Trends in Air Pollution Survey and Research <sup>1)2)</sup>

Regional scale air pollution surveys instituted early on, were launched in 1922 under the guidance of Kujyuro Fujiwara. These were performed over the long-term by the Osaka City Institute of Public Health and were reported in a summary stating the institute's results.

The following is a famous passage taken from an institute report released in 1927. "The rampant black smoke which swept the skies has not only become the utmost characteristic of our fair city, Osaka, but also has become our biggest dilemma. The enormous damage sustained by the city's citizens from the soot and smoke... it must be said that such matters as the creation of policies for the prevention of soot and smoke are of the utmost urgent mission. At our institute, this was the first step in research to remedy this matter. Beginning a few years ago, we have been performing measurements, taken at various locations throughout the city, for such things as the volume of falling soot and the quantity of airborne soot and dust. However, for those projects which were completed or started after the completion of our report for the previous fiscal year, if we were to provide a general outline it would be somewhat in the following manner..." Yoshiaki Takeda of Osaka Medical College, did research the air pollution in Osaka City from January through November 1928. During this period, he reported that the average monthly concentration of carbon dioxide (CO<sub>2</sub>) in atmosphere was 315-450 ppm, while the average concentration of sulfur dioxide (SO<sub>2</sub>) over a 24 hour period was 0.0075~0.368 ppm. These figures were reported to increase during the winter months.

Full-scale research was launched in the Tokyo metropolitan area when the Tokyo City Research Laboratory of

Public Health began to take measurements for CO<sub>2</sub>, SO<sub>2</sub>, airborne dust, and dust fall. In the Tokyo City Institute of Public Health's first report, Kunitaro Arimoto wrote the following passage. "Accompanying the increase in population and the development of industry, it is a clearly known fact that the city's air is continually becoming polluted. The chimneys which stand like the trees of a thick forest, exude billowing soot and smoke which covers the sky. Of course, while on one hand this is an unmistakable evidence as to how our city is flourishing, when looking at it from the standpoint of the health of the citizens, it must be said that this a matter of great concern... When giving consideration to the origin of the soot and smoke, it is evident that this method is an incomplete combustion of fuel. Included in the soot and smoke that is emitted are large amounts of combustible materials such as soot and tar. It goes without saying that this is a huge loss of fuel...". This report covered surveys which were performed from May 1927 to March 1928, again from May 1934 to April 1935, then again from 1939 to 1941. In addition, Yasuo Miyake made an analysis of the minor quantities of air and rain water he took at the central meteorological observatory and weather station beginning from 1935.

### 2.2.2 Air Pollution, the Second Stage <sup>1) 2)</sup>

#### (1) Facing the Issue of Air Pollution during the period of Industrial Restoration

By the end of 1945 NKK Corp. had halted operations of its blast furnace and open-hearth furnaces in the Keihin Industrial Area. However, right after the end of the Second World War, the company reopened a portion of its pipe manufacturing plants, cokes oven, chemical plant, and steel sheeting factories. In 1946 NKK put into operation 3 of its open-hearth furnaces. In October 1945, Nippon Oil Co., Ltd's Yokohama oil refinery disposed of its paraffin based oil and by May 1946 began to take in crude oil. With this, the restoration of Japan's industry which was destroyed during the war in the Second World War. The main energy source at this time was domestic coal. People once again began to suffer from the dust fall, especially those living in and around industrial areas. In particular, in Ube City, Yamaguchi Prefecture, during the first half of 1940's, it was indicated that the problem of dust fall caused by the combustion of low-grade coal, which was being used to accomplish industrial restoration, needed to be resolved. Furthermore, Japan's steel mills were at blame for the emission of densely red iron oxide fumes from its open-hearth furnaces, steel converters, and electric furnaces adopting oxygen-blowing steel refining technology. This worsened the problem of gray skies for those living in the city. Fallout of dust and soot around the factory, soiled laundry hung out to dry, caused corrosion of daily utensils, and decreased the chances of a clear sight of the area's panorama. The shades of colors in paintings drawn by children living in the cities of Kawasaki and Amasaki were much more dismal in comparison to drawings done by the children living in residential areas. This was attributed to the effects of the soot and smoke. There was noticeable withering of vegetation. Also there was a noted effect on the health of the area's residents. During this period, in Yokohama, the officers and men of the US occupational forces and their family fell ill to a respiratory disease known as "Yokohama asthma". Air pollution was pinpointed as the cause. The fear of the effects of air pollution on human health became a reality.

#### (2) Enactment of Pollution Control Ordinances and Struggles of Local Governments <sup>1) 2) 3)</sup>

Around 1950, the complaints about industrial pollution increased. Civil action against air pollution damage

gained momentum. Local governments were made to face these numerous petitions. In order to deal with this, Tokyo metropolitan area enacted pollution control ordinances in 1949, Osaka prefecture in 1950, and Kanagawa Prefecture 1951. However, during the enactment process for these ordinances, opinions were voiced that if the Department of Health drafted such ordinances, it would be a "harsh blow to industry". In opposition to this was the opinion that should ordinances be drafted by the Department of Economy, they would be "in favor of industry". Also, while opinions were in favor of the general principles backing the ordinances proposed by economic groups such as the Chamber of Commerce, they still supported factories. Some of these issues went so far as to deny the occurrence of environmental pollution, resulting in the rise of a dilemma between social welfare and whether to uphold factories. Local administrations dealing with environmental pollution were constantly handling complaints. The lack of effective measures led to the increased severity of air pollution. In December 1952, about 4,000 excess deaths were reported in London as a result of an air pollution disaster. However, this problem was not widely recognized until much later. In December 1960 the Kawasaki Chamber of Commerce called for the formation of the "Kawasaki Committee on the Prevention of Soot and Smoke". The conversion to heavy oil as an industrial fuel source led to a deterioration in the balance of trade. The worsening international balance of payments led government to reconsider its stance and contemplate a switch back to coal as a fuel source. This gave rise to civil action which sought for industrial circles to rectify air pollution. However, the government found that factories, which had invested heavily in order to make the conversion from coal to heavyoil, were not understanding of its policy. Petitions opposing the policy caused the government to withdraw its plans, instead implementing rulings which placed restrictions on the installation of heavyoil firing boilers. At the time, the conversion to heavyoil, which opposed the government's plans to save the coal industry, was thought also to be valuable to the creation of policy to counter air pollution and led to the Kawasaki Chamber of Commerce to form a committee in conjunction with labor unions, townships, women's clubs, and local assembly. However, this was actually a signal of a switch over from soot and smoke pollution to sulfur dioxide pollution.

### (3) Establishment of Soot and Smoke Emission Regional Law

Full-scale surveys began to take place during the decade from 1945 to 1955. These surveys were performed by local governments which aimed to create legislation which would give them the authority to pass regulations on air pollution. As a result of this, it was found that the main cause of air pollution was fuel combustion performed at factories and enterprises. It proved that pollution was behind the internationally grim occurrence of damage, such as health disorders<sup>1)</sup>. Furthermore, the national government, who had failed to address this issue itself, leaving the responsibility of creating industrial pollution measures up to local governments, became the focus of public inquisition. More opinions were voiced in favor of full participation by the government in the area of environmental pollution. In March 1954, the Ministry of Health and Welfare requested advice from the Japan Public Health Association in regards to permissible allowance of pollutants concentration in exhaust gas and atmosphere. In November of 1955, the association submitted a report to the minister stating that "the emission of black smoke from the source should not exceed 6 minutes every hour" etc.. In August 1955, an outline was created for drafting legislation in regard to the prevention of industrial pollution by Ministry of Health and Welfare. But, the Ministry of International Trade and Industry (MITI) criticized this proposal and in September of the same year

fabricated its own outline for legislation governing the prevention of pollution in line with the activation of an industry. The Ministry of Health and Welfare's proposal emphasized the preservation of the environment and took consideration of specified areas under the jurisdiction of the ministry itself and of that of the local government. In contrast to this, MITI's proposal aimed at a resolution to ensuing disputes and the development of industry but did not take into consideration any specified areas under its jurisdiction. The Ministry of Health and Welfare's proposal was heavily criticized by economic groups. It also faced opposition from the Ministry of Transport and MITI. So, the Ministry of Health and Welfare stopped legislation works. In 1959, MITI began legislation for air pollution control asserting that the legislation has to emphasize not only public health but also development of industry. And MITI started drafting legislation in cooperation with Ministry and Welfare. Modifications were made in combination with MITI and in March 1962, joint legislation was submitted to the 40th Diet.

In May 1962, Soot and Smoke Emission regulation Law was established<sup>4)</sup>. There was ample criticism of the unreliable government who was slow in dealing with the pollution problem due to long deliberations and also of MITI who attempted to conceal the legislative efforts of the Ministry of Health and Welfare. The government, which opted for regulations against soot and smoke as opposed to the creation of a law for the prevention of air pollution, was attacked for its shallow recognition of the issue. After emphasizing the necessity for regulations on toxic gases and automobile exhaust gas, attention was turned to the problem of why electrical and gas companies were exempt from these regulations. There was a request that authority be conferred to large metropolitan cities. There was also forceful dialogue on the need of governmental financial aid in an interpellation at the Diet<sup>5)</sup>. The House of Representatives ① requested that, when needed, in the future, the government should consider making the repayment period for funds, lent to small and medium sized firms to promote their growth, work more in favor of small businesses. Also the government should work to expand its provision of financing and other supplemental aid programs. All of this would be in order to promote the maintenance of such things as soot and smoke treatment facilities for factories and enterprises. ② stated that the government should heavily promote technical research and refer a resolution for the establishment of related measures, in order to deal with environmental pollution such as exhaust gas, noise pollution, vibrations, and offensive odors from cars in the future<sup>4)</sup>. In a summary of the law, there are stipulations for specifying regions by a ministerial ordinance where soot and smoke emissions are to be regulated and facilities subject to said laws. Furthermore, the Minister of Health and Welfare and the minister of International Trade and Industry can define the standards for regulations for each region. They can see that the law is enforced. The governor of the prefecture can establish a reporting system for new or refurbished facilities subject to the said laws as a concrete method for setting up regulations in order to control these set standards. He can also establish measures to deal with accidents or emergencies. Also defined in the law are provisions for out-of-court settlements where the governor acts as mediator in situations where a dispute over air pollution may arise. There are also provisions set for giving financial aid for necessary revisions or corrections to facilities which emit soot and smoke into the atmosphere<sup>4)5)</sup>. In particular, in Article 1 it states that, "By taking such measures as the appropriate treatment of soot and smoke being emitted due to business activities being performed at factories or enterprises, in addition to preventing danger caused by air pollution for the sake of the public's health, it is possible to plan for harmony between the preservation of our living environment and the development of industry..." Article 2 focuses on the following point, "Emission Standard is defined as the allowable limit of soot and smoke content contained in

the exhaust gas which is emitted in atmosphere from soot and smoke emitting facility.” Under this law, exemptions are made for electrical power plant and gas works. This brought about much debate on the issue. However, the most momentous fact regarding this law was the exceptionally tremendous speed at which it was passed, in regards to date of implementation, reporting system, method of measurement, emergency measures, and emissions standards. While this law contained many problems within it, it is deeply significant in the fact that it formed the fundamental framework for our today’s air pollution regulations. It made concrete the “new social order” which was necessary for the environmental pollution measures which were expressed by Naohiko Harada <sup>6)</sup> of University of Tokyo. Also it can be assumed that the formation of this law posed many difficulties for the Ministries of Health and Welfare and International Trade and Industry as it exceeded the concepts of traditional governmental legal theory. When considering the emission standards that were released on July 12, 1963 from a technical standpoint, in regards to soot and dust, it called for the use of high-performance dust collectors on such facilities as pulverized coal firing boilers, steel converters, open-hearth furnaces, coal-continuous burning limestone kiln, cement kiln, and electric furnace etc.. It also allowed for the use of C heavy oil combustion for sulfur oxides (SO<sub>x</sub>). For this reason, this law was effective in the prevention of soot and dust. However, in regards to SO<sub>x</sub> the law was said to be full of loopholes <sup>7)</sup>. When considering the fact that Japan imported crude produced in the Middle East in order to expand industry, it should have been noted that sulfur dioxide (SO<sub>2</sub>) would become a critical problem in the plight of air pollution.

#### (4) Establishment of the Basic Law for Environmental Pollution Control and Air Pollution Control Law

Policies created for the sake of higher economic growth only helped to accelerate the air pollution problem. Japan encountered with the tragedy of watching air pollution spread, maligning an even wide span of the country. In 1967, the government established the Basic Law for Environmental Pollution Control which was a basic stance on environmental pollution. As it regulated environmental quality standards, it invited debate from all around. Furthermore, the government repealed soot and smoke emission regulating law in 1968. In its place, the Air Pollution Control Law enacted and automobile exhaust gas standards were implemented. Emission standard for amounts of sulfur oxides (K-value regulation), for regulating sulfur dioxide (SO<sub>2</sub>) in atmosphere, was put into action. The increase of effective stack height was also put into effect <sup>8)</sup>. Local governments made full use of administrative organizations dealing with environmental pollution. From around 1968, local governments established their institute of environmental research and study. Administrative organizations were set up but the key problem was technology for preventing pollution. Technologies used in dust collectors had developed to some extent. However, the necessary technology for the direct desulfurization of heavy oil was only put into operation as of October 1967, at Idemitsu Kosan Co. Ltd. <sup>9)</sup>. Chiba oil refinery, Professors Keiichi Murakami and Shoichiro Hori from Tohoku University expounded on the idea of using a plentiful quantity of limestone which is an inherent resources for alkaline in solving the problem of sulfur dioxide (SO<sub>2</sub>). They explained the superiority of the limestone gypsum method, which stabilizes sulfur as gypsum and began development of the method in 1955. The two professors succeeded in creating and putting into operation a pilot plant at the Tokyo Industrial Research Institute. After obtaining a patent (Japan Patent Office No. 2152091) <sup>9)</sup>, based on this, in 1960, Mitsubishi Heavy Industries, Ltd. put into operation a 62,500 mn<sup>3</sup>/h prototype unit at NKK Corp.’s Koyasu factory <sup>9)</sup>. However, the Ministry

of International Trade and Industry furthered development of a dry process to compete with the wet limestone gypsum flue gas desulfurization process.

The limestone gypsum method eventually did become the mainstream in flue gas desulfurization. There were doubts about the government's policy which persisted on the use of a dry method. The economy continued to expand its growth, while industrial development was pursued throughout the nation. The characteristic of this era, was the posing problem of air pollution. A typical example is that of Yokkaichi City. This case is hard to leave out when talking about the problem of air pollution which Japan faces, as it tells of a growing number of patients, gale pollution, regulation on total emissions, environmental pollution court cases, and compensation for damage to the health of citizens. Also the development of the industrial area around Negishi Bay gave birth to the "Yokohama type" agreement on environmental pollution control.

#### (5) Post War Air Pollution Research <sup>1)</sup>

In 1956, Sapporo began to focus on its air pollution problems which were being caused by coal heaters. Professor Abe of Hokkaido University was asked to survey the problem. The survey began in December 1955. Samplings of dust fall were taken at 6 locations using a funnel and container and measured. The sulfate ion ( $\text{SO}_4^{2-}$ ) and chloride ion (Cl) in dust and soot have been analyzed.

In Tokyo, from November 1954, Isao Saito and his associates at the Tokyo Metropolitan Research Institute of Public Health used a wide mouthed bottle in order to measure the dust fall at 26 locations throughout Tokyo. This survey measured the amount of solids, ignition loss, ash content, radioactivity for soot fallout, atmospheric concentration of suspended particles, and carbon dioxide. A detailed analysis of the soot and dust was performed once every three month.

Kawasaki City also implemented a similar survey taking samples of dust fall at 16 places starting from June 1956. Yokohama began its survey from August 1956, taking samples at 15 locations.

In May 1957, Kanagawa Prefecture assembled a committee for the establishment of technological measures for dealing with the prevention of air pollution in the Keihin Industrial Area (headed by Professor Tetsuzo Kitagawa of Yokohama National University). The activities of the committee studied the measurement methods, dust fall t, suspended dust and soot, changes in concentration of sulfur dioxide, regional dispersion, meteorological effects, health and hygiene, and techniques for prevention.

Osaka City Research Institute of Public Health, which has the longest history of measuring dust fall, has been continually conducting measurements since before the Second World War in the Osaka area. During the early stages of post-war Japan, air pollution research in Osaka City expanded, and from around 1955 measurements for the volume of dust fall, and concentration of sulfur dioxide and suspended dust and soot were started.

The commencement of research on air pollution in Kobe began at a relatively early stage. From 1958 to 1959, results were already obtained the area distribution of dust fall. The iron content within the dust fall was analyzed, measurements of sulfur dioxide concentration in atmosphere were carried out, and testing was done to determine health effects.

The Mayor of Ube consigned dust and soot survey work to Assistant Professor Yoshikatsu Nose of Yamaguchi Medical College. Measurements of dust fall began in October 1949. This research took into consideration the

consequences on human health. The survey was performed in conjunction with Yamaguchi University Professor Yukata Kamioka and done systematically, with proposals made suggesting the use of technological measures.

In Fukuoka prefecture, in post-war Japan, an air pollution survey party was established between the 5 cities of Kita Kyushu and from June 1959, a survey and research was begun.

Takeo Suzuki and his associates at the National Institute of Public Health created a prototype for the tape air sampler. From December 29, 1954, on the 7th floor of the institute, they began continuous measurement of the concentration of suspended dust and soot in the atmosphere. Likewise, the Japan Society for Occupational Health (current name) made a meteorological study by taking measurements of the sulfur dioxide concentration in the atmosphere at seven locations within the vicinity of Tokyo from January 18-24, 1955. Kiyoshi Kawamura of the Meteorological Research Institute took measurements of the ozone ( $O_3$ ) concentration in the atmosphere in Tokyo from October to November 1950. This research was taken over by Yasuo Miyake and his associates from the same institute, and data obtained from the period of January 1951 to September 1952 was reported on. Furthermore, Miyake and his associates took measurements of the ozone levels ( $O_3$ ), nitrogen dioxide ( $NO_2$ ), and sulfur dioxide ( $SO_2$ ) concentration in atmosphere from December 1957 to March 1959.

Research for the improvement of fuel combustion was encouraged for the aim of creating more efficient uses of fuel and preventing air pollution. Heat management inspections were implemented at specified heat management factories. In addition research was promoted in the fields of flue gas desulfurization technologies and dust collector.

Air pollution gradually became the focus of researchers. Eight of Japan's representative air pollution researchers invited people to become founders for a new organization. About 80 people gave their approval to the project and on December 19, 1959, the Japan Society of Air Pollution (currently the Japan Society for Atmospheric Environment) was established at the National Institute of Public Health.

### 2.2.3 Air Pollution, the Third Stage

#### (1) An Outbreak of an Environmental Crisis

In 1970, the media began to heavily attack the delay in improvement of environmental pollution. On May 21, 1970, a high concentration of lead was found in the blood of those who took a public health examination held near an intersection in Ushigome, Yanagi-cho, Tokyo. This was used as an opportunity to take measures to reduce the amount of lead additive found in gasoline. Also, the occurrence of health injuries due to photochemical smog became more prevalent. This led to the recognition that this was an important topic which needed to be resolved and measures targeting regulation of nitrogen oxides ( $NO_x$ ) and hydrocarbons became the issues at hand. However, the only improvements seen were to the fallout of soot and dust and carbon monoxide ( $CO$ ). There was not even the slightest sign of a decline in the sulfur dioxide ( $SO_2$ ) concentration in the atmosphere. Emergency measures to curb the sulfur dioxide levels in various areas were done recurrently. There were some areas where high levels of sulfur dioxide concentration, 0.7 ppm per hour, were found in the atmosphere. Focus was placed on automobile exhaust gas, and it became a critical issue how to deal with this new problem. It was unsure as to how to deal with this new problem. Pressure from the mass media and the nation's citizens grew stronger, placing the country in a true environmental crisis.

## (2) Establishing the Diet's "Pollution Session" -Dealing with the Environmental Crisis <sup>(10)(11)</sup>

In May 1970, the House of councilors special committee on environmental pollution decided, the "issue of environmental pollution". The proposal called for the national government to "consider the items set forth and plan for the further promotion of environmental pollution measures". The items set forth and emphasized in the proposal were as follows: (1) along with the strengthened administration of the conference on Environmental Pollution Control for the sake of the comprehensive and cooperative promotion of environmental pollution measures, to ensure, without any regret, such matters as the use of functions which would allow for the comprehensive or general adjustment or correction of related matters, and (2) to devise further budgetary, financial, and tax measures, plus other necessary measures for the planning of the Improvement of the maintenance of facilities used in the prevention of environmental pollution. In July of the same year, there was strengthened centralization of the information system for environmental pollution. With the prime minister acting as chief, the headquarters on environmental pollution control was established on July 31, 1970 to provide a comprehensive means for creating and dealing with environmental pollution measures. Following this, a similar groundwork was done on the prefectural level, with prefecture governors heading their own regional headquarters for environmental pollution control. The administration's recognition of the pollution problem spread nationwide. At this point, governmental departments began to endorse such things as, promoting the establishment of environment quality standards, approval of plans for preventing environment pollution, partial revision of Basic Law for Environmental Pollution Control, Air Pollution Control Law, Road Traffic Law, and the Law for the Prevention of Pollution of the Sea by Oil, clarification of responsibilities of the enterprises, reform of laws related to water quality regulation, and the establishment of pollution offense law. It recognized the fact that it was not carrying a problem which would be easily dealt with. On November 25, 1970, the 64th Diet (Pollution Session) was held. Prior to holding of this session, on September 21, 1970, then Prime Minister Eisaku Sato spoke at a public hearing related to the national government, expressing the ideology of "there is no growth without well-being". This remark, along with the Prime Minister Sato's opening remark of his general policy speech at the Diet "...This Diet session, I have submitted a proposal for the revision of the Basic Law for Environmental Pollution Control, from the standpoint of putting the lives of our citizens first...", was very impressive. The 14 proposed laws that were submitted in the draft of the environmental pollution control law that was introduced to the Diet was passed with only a single segment having been revised. The 14 laws included are as follows: legislative bill to revise a portion of the Public Nuisance Counter measures Basic Law, legislative bill to revise a segment of the Road Traffic Law, legislative bill for partial revision of the Noise Control Law, legislative bill for Waste Disposal Law, legislative bill for revision to part of the Sewage Water Law, legislative bill regarding the Law Concerning Entrepreneur's Bearing of the Cost of the Public Pollution Control Works, legislative bill for the Law relating to the Prevention of Marine Pollution and Maritime Disaster, legislative bill relating to penalization for environmental pollution crimes which inflict on the health of individuals, legislative bill revising a portion of the Agricultural Chemicals Regulation Law, legislative bill regarding such matters as the prevention of pollution to soil used for agricultural purposes, legislative bill for Water Pollution Control Law, legislative bill for reform of a segment of the Air Pollution Control Law, legislative bill to revise a section of the Natural Parks Law, and lastly, a legislative bill to revise part of the Poisonous and Deleterious



Substances Control Law. A partial revision to the Air Pollution Control Law led to the removal of the article on reconciliation from the original draft. The idea of "preventive medicine" in areas not inflicted with eminent levels of air pollution led to the abolishment of regulations which designated certain areas due to their heavy pollution levels. Also, hazardous materials and specified toxic substances were added to the list of dust and soot controlled subjects. Other matter such as nitrogen oxides, cadmium, and lead were made controlled subjects. Furthermore, regulations and measures were devised for the dust made during mechanical crushing and transportation. Controls placed on automobile exhaust were widened, and regulations on hydrocarbons and nitrogen oxides were instituted. The emission standards for the dust and soot were strengthened. Stricter prefectural standard for hydrogen fluoride and chloride emission were implemented. Reduction of fuel sulfur due to fuel regulations placed on such things as building heaters in densely populated areas of a city. Regulations were also adopted which helped in strengthening emergency measures. Reforms were made to laws which had allowed for exemptions to electric thermal plants and gas works. Prefectural governors, in fact, became able to control air pollution measures such as those for thermal power plants etc.. This revision provided for the promotion of the enactment of laws for avoiding environmental crises. However, there existed many technical problems. In particular, regarding photochemical smog, there were many issues at hand such as phenomenon analysis, and prevention of the emission of nitrogen oxides and hydrocarbons.

### (3) Establishment of the Environment Agency <sup>10)</sup>

In July of 1971, the Environment Agency was established as a way for the government to centralize its administrative planning of environmental pollution issues. The framework and significance of this was stated in a report on legislative bills relating to environmental pollution, such as the draft for revisions to the Basic Law for Environmental Pollution Control, submitted on November 19, 1970, by the Central Council for Environmental Pollution Control. It stated that "In order to deal with the wide and diversified range of environmental pollution issues, and from a standpoint based on the future further development of economic and social conditions, while planning a mutually harmonious affiliation, it is believed that not only is the maintenance of various institutions necessary, but a broad scope of promotion of all measures is also needed. Therefore, along with the execution of research into the problem of centralizing several administrative organizations, we look to the government to further establish and promote comprehensive policies". And as if referring to this opinion, the Headquarter for Environmental Pollution Control, which had succeeded in building a basic framework for environmental pollution control, held an extraordinary organization in which it recognized the need to centralize the regulatory functions, which were dispersed among several ministries, into one permanent institution, in order to strengthen its control over environmental pollution <sup>10)</sup>.

### (4) Environment Agency and Air Conservation Measures

The establishment and the revision of environmental quality standards, introduction of regulation of total emission for sulfur oxides and nitrogen oxides, will and hesitation on hydrocarbons emission regulation, auto exhaust gas emissions regulations, promotion of improvements to fuel, good relationships with local governments, and promotion of research for the air pollution have been noticed as activities of Environment Agency. This process

led to issues which were filled with dilemmas such as adjustment and discord with other ministries, dealing with the system for Pollution-related Health Damage Compensation and civil action, debate with industrial circles related to regulations and adaptations with the local governments who were at the brunt of civil disputes.

Following the establishment of the Air Quality Bureau, with the assistance of local governments, a system was established to measure total air pollution emissions nationwide. Also, in regard to offensive odors, standards were set and with the aid of the National Institute for Environmental Study, research was performed on the structure of the creation of oxidants. Responsibility for a survey investigating the effects of complex air pollution on the health of human beings done by the Ministry of Health and Welfare and the national air monitoring network were taken over by the agency. Local governments were notified of emergency measures and a system to provide aid for setting up facilities was put into operation. The agency supervised the creation of a regulations manual on total emissions, a manual on air pollution continuous monitoring system, and guidelines governing the methods of measurement for air pollutants. It surveyed technology for controlling air pollution and established air quality standards. Several times, focus was placed on stationary emission sources in order to strengthen both the standards regulating the emissions of nitrogen oxides and automobile exhaust gas emissions regulations. Decisions on plans for preventing environmental pollution control, Environmental Pollution Control Service Corporation, and the Pollution-related Health Damage Compensation system all became important issues in the fight to preserve air quality. Environmental impact assessment and pollution-related damage compensation prevention system were also impressive. In order to deal with the energy crisis which began in 1978, a conversion from liquid fuel to solid fuel was promoted. Focus was placed on only those items which threatened to reverse the trend of accomplishing the preservation of air quality. The Environment Agency played a significant role in avoiding an environmental crisis. During this time, such matters as the future establishment of the Environment Basic Law, global environmental issues, and contributions to the international environment helped to gain further recognition of the agency's necessity.

#### (5) Local Government Response to Environmental Crisis

Prefectures, ordinance-designated cities set up their own administrative organizations. The area monitoring for air pollution and the number of controlled substances targeted for measuring were increased. Plans were made for the strengthening of the functions performed by environmental pollution study and research institutions. With this, the local environmental administrations were completed. The government, who sought for caution in regards to strict standard for sulfur oxides, was ignored. There were those local governments who enforced total emission regulations through ordinances and legal outlines. This trend was also illustrated in the regulations of the total emissions of nitrogen oxides. Regulations of total emissions for sulfur oxides as defined under the Air Pollution Control Law were implemented throughout various regions. Regulations stipulated under the same law for controlling total nitrogen oxide emissions were implemented in Tokyo metropolitan, Kanagawa, and Osaka prefecture. In 1971, in the Kanto region, complaints arose concerning eye and skin irritation due to rain. This raised to the problem of polluted rainwater. Research on acid rain was performed mainly by local government level. Local governments settled on local environmental measurement plan and made efforts to promote environmental monitoring system and to enlarge environmental research organizations. The local governments fulfilled their role

as the source for knowledge on local environmental technology. Its contribution to the elusion of an environmental crisis was remarkable.

#### (6) Response by the Ministry of International Trade and Industry and Industrial Circles

Focus was turned to the training of engineers and development of pollution control facilities by the Ministry of International Trade and Industry (MITI) and Industrial circles and the system for pollution control managers. There is need to make special note of the completion of the dust collection technology for converter in steel mills and thermal electric plants. Desulfurization plants for free gas became popular. Currently there are some 2,000 units in operation around Japan. Japan, along with the US and Germany are one of the world's leading nations which possess Desulfurization plants for free gas. Furthermore the adaptation of desulfurized heavy oil and the conversion to natural gas for fuel, contributed to the dramatic decrease in sulfur dioxide concentrations in atmosphere. After the energy crisis of 1978, energy saving techniques spread and enabled for the planning of streamlined fuel combustion. Industrial circles, in order to deal with the impending regulations on nitrogen oxide emissions, established low NOx combustion and exhaust denitration technologies. Also there were a spread in the use of hydrocarbon recovery technology.

Promotional measures by the car industry in order to deal with auto emissions regulations covering such as problems as waste incinerators and facilities which emitted foul odors, were given positive evaluations. However, large-scale land development and air pollution such as the Negishi Bay development and agreement on environmental pollution control, Project of Oogi-sima, Keiyo industrial area and air quality preservation, and Yokkaichi and the ensuing court litigation, are still unforgettable.

#### (7) Furthering Research on Air Pollution

Monitoring techniques and research institutions on the occurrence of air pollution were advanced and research was also performed on the long transport of air pollutants. There was more activity in diverse research and technology which enabled the prediction of air pollution was developed. These factors contributed to regulation of total emission of sulfur oxides and nitrogen oxides and helped to establish the environmental assessment system. Research into stench and foul odors was also promoted. Also, the development of technology in the area of preventing air pollution was exceptional. Local institutes researching environmental pollution, the National Institute for Environmental Study, National Institute for Resource and Environment, and the Training Institute for Environmental Pollution Control functioned, helping to establish a research system. The environmental research institutes furnish grants to such nations as Thailand, China, Indonesia, and Chile.

### (2.1) Literature cited

- 1) Ministry of Health, "Mortality and Morbidity During the London Fog of December, 1952". Report of Public Health and Medical Subjects No. 95, London, H.M.S.O. (1954)
- 2) Shrench, H.H. and others: "Air Pollution in Donora, Pennsylvania Epidemiology of the Unusual Smog Episode of October, 1968", Public Health Service Bulletin No.306, Washington, D.C., Government Printing Office (1949)
- 3) Mallete, F.S.; "A New Frontier : Air Pollution Control" Proc. Inst. Mechanical Engineers (London) 22 : 589, (1952)
- 4) Halliday, E.C. "A Historical Review of Atmospheric Pollution" in Air Pollution, World Health Organization, Columbia University Press, New York, 7 (1961)
- 5) Stern, A.C. "Administrative Needs and Patterns of Air Pollution Control" in Problems and Control of Air Pollution, F.S. Malletic, Editor, Reinhold, New York (1955)
- 6) WHO and UNEP : Urban Air Pollution in Magacities of the World, Blackwell Publishers (1992)

### (2.2) Literature cited

- 1) Himi, Yasuji, et al.; Association History, *J. of Japan Society of Air Pollution*, 24, 319-338 (1989)
- 2) Himi, Yasuji; History and Development of Air Pollution Research in Metropolitan Areas, *J. Environmental Pollution Control* 20, 314-322, 734-752 (1984)
- 3) Himi, Yasuji; My 43 years together Environmental Policy, (1,2), *Lifestyles and the Environment*, 42, (1) 83-89, (2) 103-107 (1997)
- 4) Kato, Ichiro; Creation and Development of Pollution Laws, pp. 59-98, Iwanami Shoten (1971)
- 5) Government Newsletter Extra No. 40 Diet Session Upper House Minutes No.34 (1962)
- 6) Harada, Naohiko; Pollution and Administrative Laws, pp. 10-101, Kobundo (1980)
- 7) Yamashita, Kiichi; Pollution Problems due to Toxic Gas, *Lifestyles and the Environment*, 9, (7) 73-79 (1964)
- 8) Ogata, Masahiko, et al.; Heavy Oil-Exhaust Gas Desulfurization Techniques, p.7, Nikkan Industrial Daily (1974)
- 9) Kishi, Masahiro, et al.; Limestone-Gypsum Method Desulfurization Device, *Air Pollution*, 1, 167-171 (1965)
- 10) Environment Agency 10th Anniversary Executive Committee; 10 Year History of the Environment Agency, Administration (1982)
- 11) Uematsu, Morio; Introduction to New Pollution Laws, Tokyo Education Information Center (1971)

## Chapter 3 Background to air pollution regulations in Japan

### 3.1 Overview

The air pollution status in a specific area are determined by the amount of pollutants emitted into the atmosphere and the climatic conditions, but the former is dependent on the desire of the local residents to keep the air clean. Quite naturally, a direct indicator of the desire held by the local residents is the air pollutant emission regulations. The background and topics related to these regulations cannot be ignored as a part of their effective implementation. Many events have occurred in the course of the enactment of air pollutant emission regulations as was touched upon in Chapter 2, and this Chapter will focus on the background and topics related to these processes.

### 3.2 Impetuous and background to establishment of regulations

The direct impetus behind air pollution regulations has been the activities of local citizen groups which have complained about the damage caused by air pollution. These citizen groups prompted the surveys and research on air pollution that formed the foundation for the air pollution regulations. As touched upon in Chapter 2, complaints from the public focused on damage from black smoke, falling ash and sulfur dioxide, and at times these pollutants have lead to economic loss and deterioration of health. Thus the pollution control administrations were begun by the local governments confronted with directly these problems based on the enactment of the local ordinances on pollution control. Surveys and research on the air pollution attributes and effects which were launched at the coordination of local governments were behind elucidation of the exact causes of pollution and the damage it caused together with the subsequent enactment into law of air pollution regulations<sup>1)</sup>.

Then with the moving forward of these surveys and research, although electric power plants and gasworkers were to be exempted and a debate arose, the Smoke and Soot Regulation Law was enacted which created the basic framework for today's air pollution regulations. As was touched upon in Chapter 2, Naohiko Harada of the University of Tokyo<sup>2)</sup> has stated that "It is not enough to tackle the current complex pollution based on the principle of individual civic responsibility (such as criminal responsibility, responsibility for illegal activities and police responsibility). A new social system is required that differs from the principles of civic law to eradicate pollution and protect the natural environment." The need for a new social order and the concrete codification of this order into law is a major result of the civic movements which brought suits against air pollution damage and the research into air pollution which took place against this backdrop. If it is considered that the settlement and mediation system which is set down under this law has gone past the private dispute non-intervention rules<sup>3)</sup> based on traditional administrative law theory, then the presence of a major significance can be discerned in the impetus behind the creation of regulations and its background.

### 3.3 Technical aspects behind the implementation of regulations

A wide range of technical issues were involved in not only the survey and research, but also the execution of

the air pollution regulations. The first that will be considered herein are the analytical and measuring methods of air pollution. In other words, it can be said in both the survey and research on the attributes of air pollution and also in the development of countermeasures, and technology that any debate on the environment without data is simply an empty argument thus, objective data which can be used to objectively evaluate these areas is absolutely essential. Analytical and measuring methods of atmospheric pollutants were sorely lacking in Japan at that time. The paucity of appropriate techniques lead to the development of the continuous automated analysis technology which is in use today and serves as the foundation for Japan's analytical chemistry. Furthermore, it goes without saying that these analysis and measurement techniques made a major contribution to the development of countermeasures and technology. The creations of the emission factors required complete an air pollution model for carrying out streamlined regulations and of the methods for evaluation of countermeasure technology are the tabulation of measurement data at the emission sources and fuel components. In addition, climatic data, air pollution dispersion theory and wind tunnel experimental results were critical in the construction of an air pollution model.

Thus engineering knowledge and experience was required in the development of the dust collector, the exhaust gas desulfurization, denitrification equipment and fuel desulfurization technology to achieve the emission standards set down in the air pollution regulations. These were comprehensive technologies which formed the foundation of equipment engineering knowledge such as chemical engineering and mechanical engineering. These technologies together with their maintenance control systems which were fruits of this technology along with the efforts of the engineers who were involved in their development and the engineers who worked in industry should be paid particular attention. In addition, public health and sanitation knowledge related to health effects of air pollution based primarily on immunology was essential for the establishment environmental quality standards that is object of administration on air pollution control. The construction of the air pollution monitoring systems and their maintenance and control which was critical to the air pollution regulations would have been inoperable without the backing of a wide range of technical fields such as analytical chemistry, electronics and meteorology. Furthermore the factory inspection which is a direct regulatory action would be impossible to execute smoothly without not only the technical knowledge required to measure air pollutants in these emission sources, but also a deep technical knowledge of their operation. The same is true of checking of countermeasures against emission sources of air pollution. For such reasons, it should be understand that a wide range of technical achievements are found in the backdrop to Japanese air pollution regulations.

### 3.4 Social aspects behind the implementation of regulations

The Japanese air pollution regulations were initiated based on the enactment of the Smoke and Soot Regulation Law, and this Act was established against the backdrop of a wide range of social considerations. Hideo Nakajima, a member of the Diet stated in a General Session of the 40th Diet during which a draft of the Soot Regulation Act was debated that "it is true that a politician stated that soot and smoke are the soup of the citizens living in industrial areas. Thinking about that today, it can be said that statement was extremely intemperate, and this way of thinking had become generally accepted and taken root, and a dark canopy has come to cover the lives of our urban residents. The rising smoke was a sign of industrial prosperity and it should be welcomed. There are many who think that the

movements that emerged that called for the elimination of this smoke were a dangerous threat to the development of industry. However the development of science has bolstered the performance of equipment for removing and prevent smoke and dust such as dust collector and in particular, the new electrostatic precipitator have advanced to a level where both soot and dust as well as bacteria such as tubercle bacillus and E. coli can be eliminated. Accordingly, the anti-pollution movement has gradually expanded over time with the support of public opinion, and pollution prevention ordinances have been set up by local governments. This movement has worked hard for the benefit of the regional population, and they have provided financial support for pollution prevention equipment in some prefectures. It has been only the government over the years which has been lacking in the zeal to attack this problem. When the Ministry of Health and Welfare drafted the Air Pollution Control Bill in 1955 and 1956, it was opposed by the Ministry of International Trade and Industry, and the bill was tabled and forgotten. Now I have heard that the Air Pollution Control Law has been presented to the Diet, and while what it is the past is past, I have great expectations for the future..."<sup>3)</sup>. Nakajima's statement indicates that a forward looking approach was not taken at that time.

The Air Pollution Control Law was established in 1968, subsequently revised in 1970 and then followed by the implementation of setting down regulation of total emission on sulfur oxides and nitrogen oxides. These regulations were greatly affected by the social circumstances at the time which extended to the historic shift described in Chapter 2 and the severe air pollution health damage and environmental pollution trials as illustrated by the Yokkaichi which is described in Chapter 5 and 12 <sup>4)</sup>. If a corporation cannot verify that there is no cause and effect relationship between damage to health and air pollution, then corporate joint illegal action and responsibility is recognized with the establishment of the cause and effect relationship, and this had an impact on environmental pollution policy.

It goes without saying that local governments were caught in a dilemma at the time of trying to balance the citizen movements which were attacking the corporate intransigence. At the time, the situation may have been overly sensationalized, but the power of the mass media to appeal to public opinion surrounding air pollution could not be discounted. The power of the press cannot be ignored if we look at shocking news that sounded a warning to industrialized society including the criticisms leveled by the mass media which provided the direct impetus for the "environmental crisis" of 1970.

Of course, there is the economic background to the power of Japanese corporation, but with this social background, corporate executives have been spurred on to strive to prevent air pollution in accordance with air pollution regulations.

### **3.5 Administrative structure as an aspect of implementation of regulations**

The smoke and soot emission regulation law unmistakably has created the first step of air pollution control administration in Japan, and while it is regrettable, it was the regulations that were effective in the prevention of air pollution. For that reason, administration of regulations is important.

As was previously stated in <sup>2)</sup>, this law correspond to the demands of a new social order and they exceeded traditional theories of public policy. It was thus for that reason that the establishment and operation of this

administration organization was relatively difficult. It was those in charge of administration that succeeded in overcoming these difficulties. Administrative officials found themselves struggling with science and technology which they knew little or nothing about and despite their lack of experience grappling with legal interpretations, technical officials struggled to get appropriate qualifications because without a wide scope of scientific knowledge, policy could not be moved forward, and researchers strove to obtain essential scientific knowledge through measurement and analysis to promote policies to preserve the atmosphere. All found themselves faced with enormously difficult tasks and groping in fields they were inexperienced.

The researchers who were involved in survey and research on air pollution went through an era in which they encountered conditions that ruined measurement instrument<sup>1)</sup>. In addition, almost no scientific support system was in place, and only a few people in society actually understood what was going on.

In the midst of this, personnel in the local governments were instrumental in supporting and promoting organizations and leaders and the promotion of personnel education activities in the Institute of Public Health in the Ministry of Health and Welfare, National Institute for Resources and Environment, in the Agency of Industrial Science and Technology in the Ministry of International Trade and Industry and the Training Institute for Environmental Pollution Control in the Environment Agency were not just important in policy to preserve the atmosphere but in the development of environmental regulatory policy in Japan.

The activities of academic bodies such as "Japan Society of Air Pollution which was air pollution research nationwide council (present Japan Society for Atmospheric Environment)"<sup>1)</sup> which was established in Japan in 1959 had a major effects on these activities and formed its background.

### 3.6 Background to the era

The effects of regulations, trials and compensation that were advanced against the background of the air pollution problems that arose were major. A summary of the background to the age is summarized in Table 3.1<sup>5-7)</sup>.



Table 3.1.1 (1) Backdrop by Age to Air Pollution Regulations in Japan<sup>5-7)</sup>

Year	Proposed air pollution problems (background)	Measures to cope with the air pollution problems (regulations)
1877	Pollution problems frequently arise in Osaka with the three companies of steel plant, smithy and bath house.	The Manufacturing Industries Regulations are released in Osaka
1884	Problems first arise caused by factory smoke and soot from the burning of coal in Osaka because the city was at the forefront of industrialization.	A city circular was issued by the city of Osaka banning the establishment of factories which gas coke is burned in Shimanouchi ship yard.
1888	A smoke and soot problem arose with the Osaka Light Company.	A city ordinance is issued banning the construction of factors with smoke stacks within the city.
1895	Large scale crop damage occurred in Matsukimura in Tochigi prefecture due to the expansion of refinery at the Ashio copper mine. A number of village residents filed suit to stop work because of the crop damage caused by sulfur dioxide from the Besshi copper mine in 1893. The company announced that its work was unrelated to the damage, several hundred farmers stormed the company, and arrests were made.	An out of court was agreed to between the Ashio mine and the farmers. The headquarters of Sumitomo drafted a plan for transferring the Shikyuhama Refinery to Shisakujima. Construction was commenced in 1896.
1909	Hitachi Mining completed a large scale refinery in 1908, damage caused by sulfur dioxide became severe in 1909.	Compensation was paid based on negotiations.
1916	The Zushi plant of Suzuki Seiyakujo (Ajinomoto Plant) caused hydrogen chloride damage.	Kanagawa Prefectural factory regulation rule was enacted.
	Respiratory organ dysfunction in children caused by sulfur dioxide from Osaka Alkali Co., Ltd. Suit brought by farmers	An order is issued by the city of Osaka to install prevention and removal equipment. The suit is won by the plaintiffs (farmers) in 1919
1919	Pine trees, which are called shingenko hatakakeno matsu, located near the Hinoharu Station on the Chuo Line of the national railroad are withering and dying as a result of smoke from the locomotives.	The owners of the trees sue for damages, and win in a decision by the Supreme Court
1924	The Fuel society of Japan (present the Japan Institute of Energy) formed the Special Committee on Fuel for city use. This society focused its attention on the prevention of smoke in response to concerns related to beautification, health and fluctuations in fuel prices as well as proposing regulations requiring the use of only smoke-free fuel (hard coal) in Tokyo.	The Ministry for Home Affairs issued the smoke and soot regulations in December of the same year in six major cities (Tokyo, Osaka, Kyoto, Nagoya, Kobe and Yokohama).
1932	The Osaka Smoke and Soot Prevent Investigation Committee which was inaugurated in 1927 decided upon smoke and soot damage surveys, the use of smoke-free fuel and electrification, complete combustion and smoke and soot regulations.	The Osaka Smoke and Soot Regulations consisting of a total of 13 Articles were announced, and emissions of black smoke with a Ringelman's smoke chart concentration of 3 or higher were prohibited for six minutes or more per hour. The same measures were adopted in Kyoto in 1933 and in Hyogo Prefecture in 1935.
1935	The clouds of war began to gather on the horizon even in the capital. The move toward war could be seen in the expansion of existing and construction of new factories, and air pollution began to gradually attract attention as a problem.	The Smoke and Soot Prevention Guidelines were set down in Tokyo in 1935, and Kanagawa Prefecture enacted the Smoke and Soot Prevention Committee Regulations in 1937.
1949	Along with the reconstruction of industry in the post-World War II era, the level of pollution generated by factories increased.	Tokyo was the first city in the country to enact regulations in the form of the Tokyo Metropolitan Factory Pollution Prevention Ordinance. These regulations did not contain quantitative standards, but instead focused on areas such as the generation of noise.

Table 3.1.1 (2) Backdrop by Age to Air Pollution Regulations in Japan<sup>5-7)</sup>

1950		Enactment of Osaka Industrial Pollution Prevention Ordinance
1951		Enactment of Kanagawa Prefecture Industrial Pollution Prevention Ordinance
1955	Smog from soot and burning of fuel for building heating systems frequently blankets urban areas, and prevention of such smoke becomes necessary.	Enactment of Tokyo Metropolitan Soot Prevention Ordinance (Regulations covering concentration based on the Ringelman's smoke chart: residential areas 2 and other areas 3)
1962	The amount of fuel used increases along with the advanced economic growth. There is a switch in the energy sources from coal to oil.	Enactment of legislation covering regulation of soot and smoke emissions. (The regulations from Paragraph 7 Article 37 are based on JISZ8808.)
1966	The Ube City Council established a Soot and Dust Countermeasures Committee in 1951 as an inquiry body. This committee was made up of three groups, corporate representatives, city council representatives and academics (nicknamed the Ube approach).	The amount of coal used in 1951 was 85.6 ton/month increased by 79% by 1962. However the amount of soot and dust dropped by one-third from 55.9 t/km <sup>2</sup> /month through voluntary regulations promoted through these activities (close to 40 dust collection units were installed, and the soot and dust was sold as cement admixture for a total of 900 million yen over 10 years).
1967	Tatsuo Maekawa, a member of the city council of Yokkaichi began negotiations with a lawyers' group from 1961, and a plan was drafted to bring suit covering pollution damage in Yokkaichi. Nine individuals who were certified as suffering from pollution related problems brought suit in Yokkaichi court against six companies including first complex in 1967.  Laws were enacted targeting air pollution and deteriorating water quality in the 1950s, but this piecemeal approach was inadequate.	The plaintiffs won the suit in 1972 and 12 plaintiffs paid 88.21823 million yen (the court ruled that it was not necessary to verify a close cause and effect relationship from the standpoint of assisting the victims, and it was confirmed that a joint illegal action causing injury had been committed).  The Basic Law for Environmental Pollution Control was enacted that was designed to promote systematic and integrated pollution policy (Article 9 set down the environmental standard articles, and the Law was revised for sulfur oxides in 1969, carbon monoxide in 1970, suspended particulate matters in 1972, sulfur dioxide, photochemical oxidants and nitrogen dioxide in 1973) (regulations related to the pollution prevention plan were set down in Article 19: (measures to handle the situation were taken for already polluted areas and pre-prevention areas)
1968	Measures against automobile emission became necessary because of the problems that arose related to the regional spread of pollution, diversification of pollutants and the increase in the volume of vehicle traffic.	The Air Pollution Control Law was enacted because of the absence of a tie-up between separate and emergency measures and to cope with the increasingly complex and severe air pollution.
1970	Damage caused by photochemical smog, lead pollution, fluorine compounds, cadmium and chlorine compounds became a problem, and it became necessary to introduce measures to address this problem.	The Pollution Diet which was convened in November of the same year revised and strengthened the Air Pollution Control Law, and five different noxious compounds became regulated.
1971	Many pollution-related problems were arose in 1970, they worsen up until the opening of the Pollution Diet	The Environment Agency was established in May of 1971.

(by Nihei Hisao)

(3.1~3.6) Literature cited

- 1) Himi, Yasuji et al.; The History of Our Society, *Journal of Japan Society of Air Pollution*, 24, 319-338 (1989).
- 2) Harada, Naohiko; Pollution and Administrative Law, pp.10-101, Koubundo (1980)
- 3) Extra Edition of the Official Gazette Proceedings of the 40th Session of the Diet No. 34 (1962).
- 4) International Center for Environmental Technology Transfer (ICETT), Striving to Contribute to the World Environment Through Improvement of Pollution and the Environment in the City of Yokkaichi (1992)
- 5) The Tokyo Metropolitan Research Institute for Environmental Protection ed.; Pollution and Tokyo (1970)
- 6) Kawana, Hideyuki, Document Japan Pollution (1987)
- 7) Earth Environment and Economic Research Center; Japan Pollution Research (1991)

## Chapter 4 Weather Conditions, and the Variation and Distribution of the Concentration of Air Pollutants

### 4.1 Effect of Weather Conditions

#### 4.1.1 Preface

The meteorological factors that most affect the generation and sustenance of the air pollution are the wind and thermal stability of the atmosphere. Thermal stability of the atmosphere is closely related with a vertical variation of the atmospheric temperature. It is very important to have a correct understanding of the structure of lower layers of the atmosphere, since the air pollution occurs at the bottom layer of atmosphere from the ground to the altitude of several kilometers.

#### 4.1.2 Atmospheric Boundary Layer

Because many of the air pollutants are released adjacent to the ground, their behavior in the environmental air is strongly affected by the weather in lower layers of the atmosphere. The Earth absorbs energy from the Sun at the ground surface. Various weather conditions are caused by the release of this energy in the atmosphere. The atmospheric layer that directly receives the effect from the ground surface is, that is, the atmospheric boundary layer. Although the thickness of this layer is dependent on the characteristics of the ground surface and weather conditions, it generally ranges from 1 to 3 km. The atmosphere above this layer is called the free atmosphere. Within the atmospheric boundary layer, the layer from ground surface to a few tens of meters above surface is called the surface boundary layer.

#### 4.1.3 Vertical Variation of Air Temperature

The degree of atmospheric diffusion is greatly affected by the vertical variation of air temperature. In the standard atmosphere, the air temperature decreases by  $0.65^{\circ}\text{C}$  as the altitude increases by 100 m. However, this value changes considerably as the weather condition changes. The situation of temperature variation that affects the diffusion of air pollutants most is the temperature inversion layer. This is a condition where the temperature of the lower layer is below that of the upper layer. As the air pollutants are trapped in the temperature inversion layer and would not diffuse, the concentration of air pollutants is increased. The radiation inversion and the subsidence inversion are known as two major causes of the temperature inversion. Particularly, the radiation inversion which often appears in a clear night of winter season is the major factor for the generation and sustenance of a high concentration of air pollution. On the other hand, in the daytime of a fine weather, the ground is heated by sunlight to generate a thermal convection, and the vertical mixing of air is enhanced. This atmospheric layer is called the mixed layer. The height of mixed layer during daytime is proportional to the square root of the accumulated solar radiation,  $I$  ( $\text{cal}/\text{m}^2$ ). From the observational result in Tokyo during summer, the height of mixed layer  $Z$  ( $t$ ) (m)

up to 12 o'clock is known to be approximately expressed by Eq. (1). After 12 o'clock, the height is proposed to be derived by multiplying a certain factor to this value. Specifically, factors such as 1.127 at 15 o'clock, 0.696 at 18 o'clock, and 0.357 at 21 o'clock are proposed. As those values strongly reflect the regional characteristics, those must be checked with the actual observation at each site.

$$Z(t) = 76.8 I^{0.499} \dots\dots\dots (1)$$

Japan is usually covered with a high pressure air mass centered in the north Pacific during summer, and a fine weather is sustained. Accordingly, there often appears a subsidence inversion layer. As the mixed layer grown during daytime is suppressed by this inversion layer, a high concentration of photochemical smog will be generated in the atmospheric layer from the ground to about 1,000-2,000 m upward.

#### 4.1.4 Vertical Distribution of Temperature and Diffusion

From observations of the behavior of smoke exhausted from chimneys, the relationship between the smoke flow and the vertical distribution of temperature has been investigated. When a dry air mass adiabatically ascends for 100 m in the atmosphere, its temperature would drop by 0.98°C. When the atmospheric condition shows the same value as this temperature drop, this atmosphere is said to be thermally neutral. When the temperature drop rate is larger than this value, the atmosphere is called thermally unstable, whereas when it is smaller, the atmosphere is called thermally stable. Also, when the temperature of the upper layer is higher than that of the lower layer, the layer is said to be in the temperature inversion state, as described in Section 4.1.3.

The behavior of smoke is categorized into several representative types in accordance with the vertical distribution of temperature. When the atmosphere is in a neutral or a rather stable state, the smoke will diffuse in the form of coning type. Such a condition would occur in the cloudy daytime weather or in relatively strong windy weather. In the stable layer or inversion layer often seen in winter nights, the smoke will diffuse in the form of fanning type as the vertical diffusion is suppressed. In a fine weather with low wind as often seen in summer daytime, the lower layer of atmosphere becomes unstable, while the upper layer will be stable. In this case, the smoke would meander to show a loping type of behavior. When there exists a subsidence inversion layer in the upper atmosphere, the smoke will diffuse in the form of trapping type of behavior as its upward diffusion will be suppressed. If there are any unstable state simultaneously in the lower layer, the smoke will diffuse rapidly toward the ground, and brought about a high concentration will be near the ground. Such a condition is called the fumigation.

When the bump of ground, or the surface roughness varies, an internal boundary layer will be formed. For instance, when the wind blows from the sea or lake, which has a small surface roughness, to the land with a large roughness, an internal boundary layer with larger atmospheric disturbances than on the sea or lake will be developed on the land side. When the air pollutants, generated in the factories and power stations in coastal areas, would reach to the internal boundary layer of landside with larger disturbances from the sea side areas where the disturbance is smaller, there might occur a rapid mixing and diffusion and bring about a high concentration of pollutants near the

ground. This phenomenon is called the internal boundary layer fumigation.

#### 4.1.5 Vertical Distribution of Wind

The vertical distribution of wind is also an important weather factor. The wind distribution between the ground to a height of about 100-200 m is empirically represented by Eq. (2) ;

$$\frac{u}{u_1} = \left(\frac{z}{z_1}\right)^p \dots\dots\dots (2)$$

where  $u_1$  is the wind velocity at a height of  $z_1$ . The meaning of exponent  $p$  will be explained as follows. The of wind velocity increases exponentially as the altitude increases, and the amount of increase varies depending on the physical characteristics of the ground surface and the degree of stability. The value of exponent  $p$  is roughly 0.25 in the neutral state, 0.1-0.2 in the unstable state, and about 0.3 in the stable state. Even in one day, it becomes smaller in the daytime and larger in the night. If the surface roughness varies, the exponent  $p$  would increase as the roughness becomes larger. Accordingly,  $p$  is larger in the urban areas than in the suburbs. The value of  $p$  in the urban area is about 0.2-0.4.

#### 4.1.6 Urban Wind and Air pollution

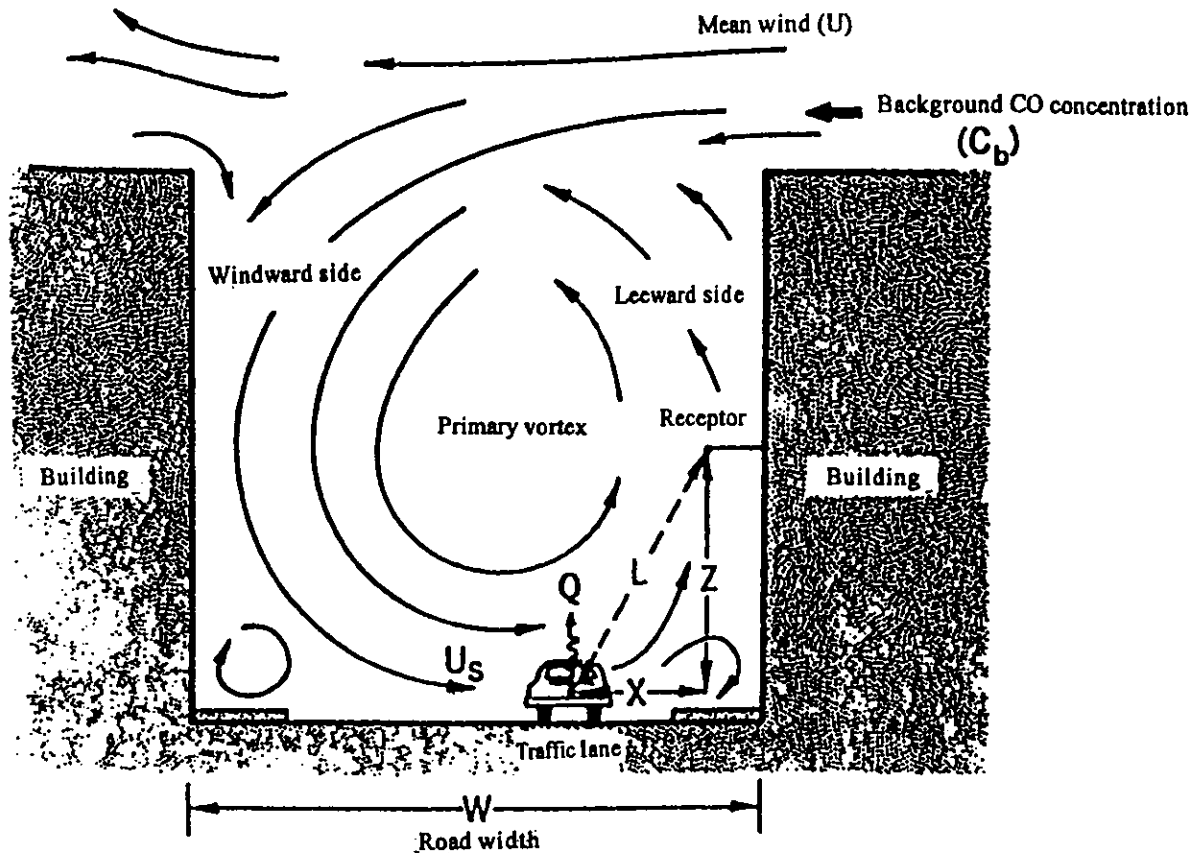
The situation where roads are surrounded by buildings to form an artificial canyon is called the street canyon. In the street canyon, a special type of wind is generated and it gives a significant effect on the air pollution in the adjacent area. The urban wind is closely related with physical conditions such as the height and density of buildings and the width of roads, and also with weather conditions such as the wind direction and velocity and the thermal stability of the atmosphere. Generally, the most typical situation is brought about when the wind blows perpendicularly to the street. The wind distribution in the street canyon in the above situation is illustrated in Fig.4.1.1.

In this situation, the concentration at a receptor,  $C_L$  would be expressed by Eq. (3):

$$C_L = \frac{Q}{k_1 k_2 (U + 0.5) [(x^2 + z^2)^{\frac{1}{2}} + 2]} \dots\dots\dots (3)$$

where  $H$  is the height of building (m),  $W$  is the width of street (m),  $X$  is the horizontal distance from centerline of the street to the receptor (m),  $Z$  is the vertical distance from centerline of the street to the receptor (m),  $U$  is the wind velocity above the street canyon and  $Q$  is the exhaust source strength (mg/m/s).  $k_1 k_2$  is a constant, and is empirically determined as  $K = \frac{1}{k_1 k_2} = 7$ . Although this equation is quite simple, it is useful to generally understand the maximum concentration found around the street. However, the value of  $K$  varies significantly

depending on the situation of the streets and the stability, and therefore appropriate studies such as tracer gas experiments and wind tunnel experiments are necessary to validate the result.



Source: (Cited from Johnson, W.B., Ludwig, F.L., Dabberdt, W.F. and Allen, R.J., An Urban Diffusion Simulation Model for Carbon Monoxide, Journal of the Air Pollution Control Association, Volume 23, No. 6, 490-498 (1973))

Fig.4.1.1 Wind Distribution in Street Canyon

#### 4.1.7 Urban Boundary Layer and Air Pollution

The local weather variation caused by the existence of cities, such as an urban boundary layer, has a very close relationship with the distribution of air pollutants. The urban boundary layer is a type of the internal boundary layer. Generally, the urban area has larger roughness than the suburbs because buildings are tightly concentrated. Also, thermal characteristics of the surface and the amount of exhausted heat are different, and therefore the urban boundary layer is formed by mechanical and thermal factors. The urban boundary layer is known to develop from windward outskirts of the city toward the city center. If the distance from outskirts is denoted by  $X$  and only the balance of sensible heat is considered, the thickness of urban boundary layer,  $Z_h(X)$  at the position  $X$  is expressed by Eq. (4). This equation works primarily for the situation in winter nights. In the equation,  $U$  is the general wind velocity.

$\gamma$  is the temperature gradient in the ground surface inversion layer, and  $\frac{Q_s}{C_p \rho}$  is the of sensible heat flux.

Furthermore, as Eq. (5) holds when the difference between the temperature of suburbs,  $T_r$  and that of the city,  $T_u$  is denoted by  $\Delta T_{u-r}$ , the temperature difference between the point  $X$  in the city center and the suburbs is represented by Eq. (6). This is the situation called the heat island.

$$Z_h(X) = \left( \frac{2}{U \gamma} \int_0^x \frac{Q_s}{C_p \rho} dx \right)^{\frac{1}{2}} \dots \dots \dots (4)$$

$$\Delta T_{u-r} \propto \gamma Z_h(X) \dots \dots \dots (5)$$

$$\Delta T_{u-r} \propto \gamma^{\frac{1}{2}} U^{\frac{1}{2}} \left( \int_0^x \frac{Q_s}{C_p \rho} dx \right)^{\frac{1}{2}} \dots \dots \dots (6)$$

From Eq. (3), it is found that the temperature difference between city and suburbs is proportional to the square root of total sensible heat added through the distance  $X$  from the outskirts, and is inversely proportional to the square root of the general wind velocity in the city. This heat island, where the temperature is higher at the city center than at surrounding areas, causes the stagnation, circulation and sustained and elevated concentration of air pollutants. The air pollutants released in the interior of the city are trapped within this urban boundary layer to cause highly concentrated air pollution. On the other hand, when the pollutants released from facilities located in the windward side are taken into the region of  $Z_h(X)$  in urban area through the urban boundary layer surfaces by being transferred through atmosphere without much diffusion or dilution in the stable layer above, an internal boundary layer fumigation occurs.

#### 4.1.8 Synoptic Meteorology and Local Meteorology

The velocity and direction of wind and the stability of atmosphere are basically controlled by meteorological phenomena of synoptic scale caused by the air flow of global scale and the physical characteristics of ground surface, but the variation of local weather also gives significant effects on the distribution of air pollution.

Among others, the sea-land breeze is particularly important. As the sea and lake have large heat capacity, their diurnal variation of temperature is relatively small, while it is large in the land area. As a result, the temperature on land area becomes higher than that of water surface during the daytime while lower in the night. Thus the temperature difference is caused between airs above the land and the water, which then causes the atmospheric pressure difference. Because of this difference, the air flows from the sea and lake toward the land in the daytime while the direction is reversed in the night. This is the sea-land breeze. The sea-land breeze has a characteristics of the gravity flow, and a conservative wind field is formed at the head of the breeze where a vortex is formed. Thus the air pollutant would be trapped within this field to cause a high concentration. As the sea-land breeze moves with time, the area of high concentration would also move with it. The line of discontinuity of the wind formed, when



the sea wind and the land wind comes in, is called the sea breeze front and the land breeze front, respectively. In mountainous areas or basins, the air temperature at the same altitude is different for the slope and the valley, and the wind blows from the valley to the mountain in the daytime, and in the opposite direction in the night. The air circulation caused by this mountain-valley breeze would give effect to the air pollution.

## 4.2 Formation Mechanism of Air Pollutants

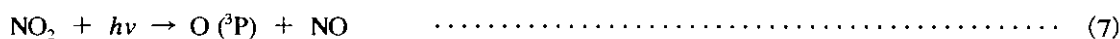
### 4.2.1 Introduction

It is well known that, among air pollutants, there are primary pollutants which are released directly by the human activities, and secondary pollutants which are formed by the reaction of primary pollutants in the atmosphere. In this chapter, the mechanism of formation and transformation of the secondary pollutants will be described, particularly the formation of photochemical ozone, the atmospheric chemical reaction involving hydrocarbons, and the reaction involved in the acid rain.

### 4.2.2 Photochemical Reaction of Ozone Formation

The reactions involved in the formation and transformation of secondary pollutants are mainly the formation reaction of photochemical smog in which solar energy is added to the primary pollutants, i.e. the oxides of nitrogen (NO<sub>x</sub>) and the hydrocarbons (HC). NO<sub>x</sub>, which is composed of NO (nitrogen monoxide) and NO<sub>2</sub> (nitrogen dioxide), has rather broad range of concentration, because its source is rather maldistributed and it has only a short life in the atmosphere. Generally, its concentration is 20-500 ppb (ppb is a thousandth of ppm) in the urban area, 1-10ppb in the rural area, and 0.05-0.2 ppb for the background including on the sea. The reactivity of NO<sub>x</sub> is very high in the atmosphere, so its life is very short. It involves in almost all reactions in the atmosphere both directly and indirectly to have an important role. Particularly, NO<sub>x</sub> has a major role in the formation of ozone in the troposphere, as revealed in the occurrence of photochemical smog in the urban area. The presence of NO<sub>x</sub> is very important, because only NO<sub>2</sub> can be the potential source of ozone in the troposphere.

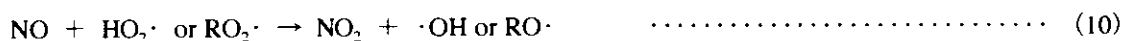
If there is a fractional amount of NO<sub>x</sub> in the air of troposphere, it receives solar radiation and the ozone is formed via the formation of NO, as indicated by Eq. (7) and (8):



where O (<sup>3</sup>P) represents the oxygen atom in the ground state. However, if those are the only reactions, the ozone concentration would attain a steady state and does not reach to any extremely high value, because the reaction (9) will occur in which ozone will be destroyed by NO produced in the reaction (7):

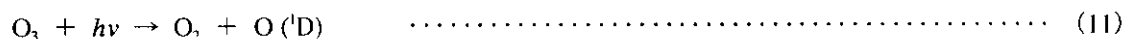


However, if there are any hydrocarbons in the atmosphere, NO will be transformed to NO<sub>2</sub> by peroxy radicals such as HO<sub>2</sub>· and RO<sub>2</sub>·, produced by the reactions of hydrocarbons, without destroying ozone, and the reaction indicated in Eq. (10) will proceed.



NO<sub>2</sub> produced in this reaction is then used to form ozone through reactions of Eq. (7) and (8). In Eq. (10), R is a group derived from hydrocarbons such as alkyls. As a result, the concentration of ozone becomes high under the condition of strong solar radiation.

Chemical reactions in the troposphere are frequently initiated by ·OH radicals. Particularly, alkanes and aromatic hydrocarbons, which do not react with ozone, are destroyed by the reaction with ·OH radicals. ·OH radicals in the troposphere are primarily produced through the reactions represented by Eq. (11) and (12), and are reproduced by radical chain reactions involving NO<sub>x</sub> and hydrocarbons, as shown in Eq. (7), to join in the reaction again:



In the above equations, O (<sup>1</sup>D) is the oxygen atom in the excited state. The oxygen atom in the ground state, O (<sup>3</sup>P) would not react with water.

#### 4.2.3 Reaction of Hydrocarbons

The reaction shown in Fig.4.2.1 is a typical photo-oxidation reaction of a common hydrocarbon, alkane. Through such a chain reaction, the carbon in hydrocarbons is gradually oxidized to carbon dioxide, and the ozone is accumulated through NO→NO<sub>2</sub> conversion, as described previously. For Type (b) products (peroxyl radicals), not only the oxidation of NO indicated in Fig.4.2.1 but also the formation of nitrate ester (RONO<sub>2</sub>) through addition and rearrangement reactions of NO, as shown in Eq. (13), becomes important, when the chain of alkyl becomes longer or larger:



Also, Type (c) products, alkoxy radical (RO·) react mainly with oxygen, but they also produce nitrate esters and nitrite esters through NO<sub>x</sub> addition reaction, as shown in Eq. (14), when enough concentration of NO<sub>x</sub> exists:



The nitrite ester is easily decomposed by visible light, and could start the chain reaction shown in Fig.4.2.1 by producing alkoxy radicals. Thus, the nitrite ester is known to act as an important initiator for the photochemical reaction, if it is accumulated during the night by the reaction of Eq. (14) in conjunction with the ozone-olefin reaction or the reaction between NO<sub>3</sub> radicals and hydrocarbons <sup>1)</sup>.

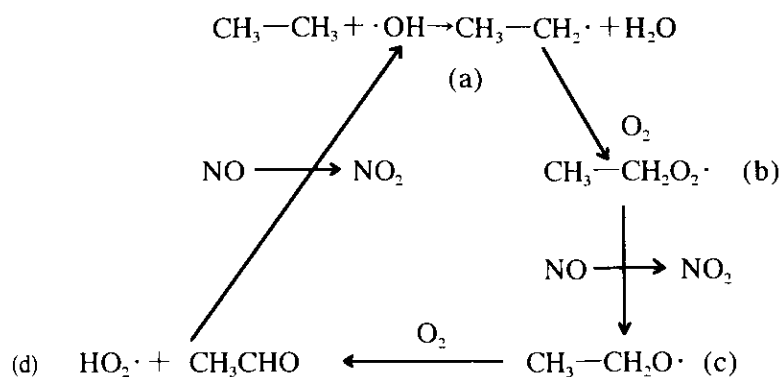
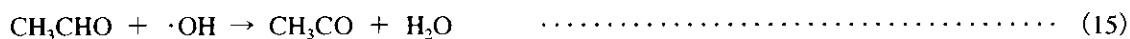


Fig.4.2.1 Photo-oxidation chain of hydrocarbon initiated by OH radical

Another important product that contains NOx is the compound called PAN. PAN was originally the generic name of peroxyacyl nitrate (RC(=O)OONO<sub>2</sub>), but now frequently indicates peroxyacetyl nitrate (CH<sub>3</sub>C(=O)OONO<sub>2</sub>), the most important one among them. Its most important formation process is the one that starts with the reaction between acetaldehyde produced by photochemical reaction of hydrocarbons of molecular weight larger than ethane (or released as primary pollutants) and ·OH, as shown in Eq. (15) to (17).



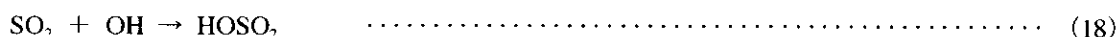
PAN is one of the photochemical oxidant, and a substance with a strong toxicity which gives visible damage to the plant and is also known to give damage to humans. Recently, its importance as a role for a nitrogen reservoir is pointed out<sup>2)</sup>. Since the life of NOx in the atmosphere is very short, NOx would disappear through reactions in the vicinity of its source without traveling long distances (long range transport). However, once PAN is formed and moves to the upper layer where the temperature is low, its life becomes very long. For instance, the life of PAN is only 1.7 hours at the temperature of 20°C (Its process of disappearance is mainly the thermal decomposition expressed by the reverse reaction of Eq.(17).), but it increases to 50 hours at 0°C and 105 days at -20°C<sup>3)</sup>. Accordingly, it is quite probable that nitrogen would be transferred for a long distance in the form of PAN and then NO<sub>2</sub> would be released by its decomposition to involve into the photochemical reaction. Actually, a high concentration of ozone, often seen in early spring in the troposphere of a middle latitude region in northern hemisphere, is postulated to be caused by an ozone source produced by NO<sub>2</sub>, which is produced as PAN has been accumulated and transferred during the winter and decomposed as temperature rises<sup>4)</sup>.

#### 4.2.4 Atmospheric Chemical Reaction Associated With Acidification of Tropospheric Air (Acid Rain)

The acid rain is one of the environmental problems of global scale that is attracting a major concern. Although the acidification of rain itself is a problem, the fundamental problem is the acidification of tropospheric air, and the acid rain should be considered as only the outcome of the latter. Of course, the acidification of tropospheric air is caused by the increase of sulfur oxides and nitrogen oxides released from various human activities. Those compounds are oxidized in the atmosphere to form sulfuric acid and nitric acid, and those would be dissolved into rain water to form the acid rain.

The increase of NO<sub>x</sub> causes the increase of concentration of ozone or various oxidizing radicals ( $\cdot\text{OH}$ ,  $\text{HO}_2\cdot$ ,  $\text{RO}_2\cdot$ , etc.), and thus enhances the oxidizing capability of atmosphere. Together with the increase of precursor for acid substances, those materials are also causing the increase of acid substances in the atmosphere.

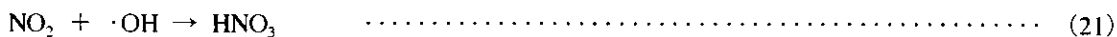
In the formation of acid substances, there are the formations of sulfuric acid through oxidation of SO<sub>2</sub> and of nitric acid through oxidation of NO<sub>x</sub>. In the oxidation of SO<sub>2</sub>, the gas phase homogeneous reaction by OH radical, as shown in Eq. (18) to (20), as well as the reaction of SO<sub>2</sub> in solution after dissolved into water in cloud, fog or raindrops are both important. Hydrogen peroxide is performing a major role in the reaction in solution <sup>5)</sup>.



On the other hand, the formation process of nitric acid through oxidation of NO<sub>x</sub> is relatively simple, and the liquid phase oxidation reaction, seen in the case of SO<sub>2</sub>, hardly occurs because NO<sub>x</sub> is not readily soluble in water.<sup>6)</sup> The oxidation is considered to proceed primarily by three reactions as follows:

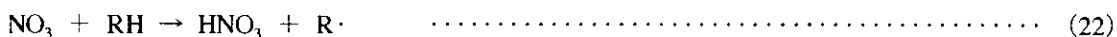
##### (1) Reaction with $\cdot\text{OH}$ radical

The role of  $\cdot\text{OH}$  is important also in the formation of nitric acid, as shown in Eq. (21):



##### (2) Reaction of NO<sub>3</sub> radical

NO<sub>3</sub> radical is a highly reactive radical produced by the reaction between ozone and NO<sub>2</sub>. Particularly, it has a very important role in the reaction of various organic compounds including hydrocarbons during the night time when the light is not available. NO<sub>3</sub> radical is converted to nitric acid by reacting with organic compounds such as hydrocarbons and aldehydes, as shown in Eq. (22):



(3) Reaction of  $N_2O_5$

$N_2O_5$  is formed by the reaction between  $NO_3$  and  $NO_2$ , and is acting an important role as one of the nitrogen containing pollutants in the reactions in the nighttime.  $N_2O_5$  is, in a sense, a nitric acid anhydride, and capable of hydrolysis as shown in Eq. (23). Here, however, the rate constant of direct reaction with water vapor in the gas phase homogeneous system is less than  $2 \times 10^{-21} \text{ cm}^3 \cdot \text{molecule}^{-1} \cdot \text{s}^{-1}$  <sup>7)</sup>, and is not important. However, the extinction process of  $NO_x$  or the formation process of nitric acid would be accelerated with the existence of a surface, and it is important as a formation process of nitric acid in the open air.



### 4.3 Concentration Variation of Air Pollutants

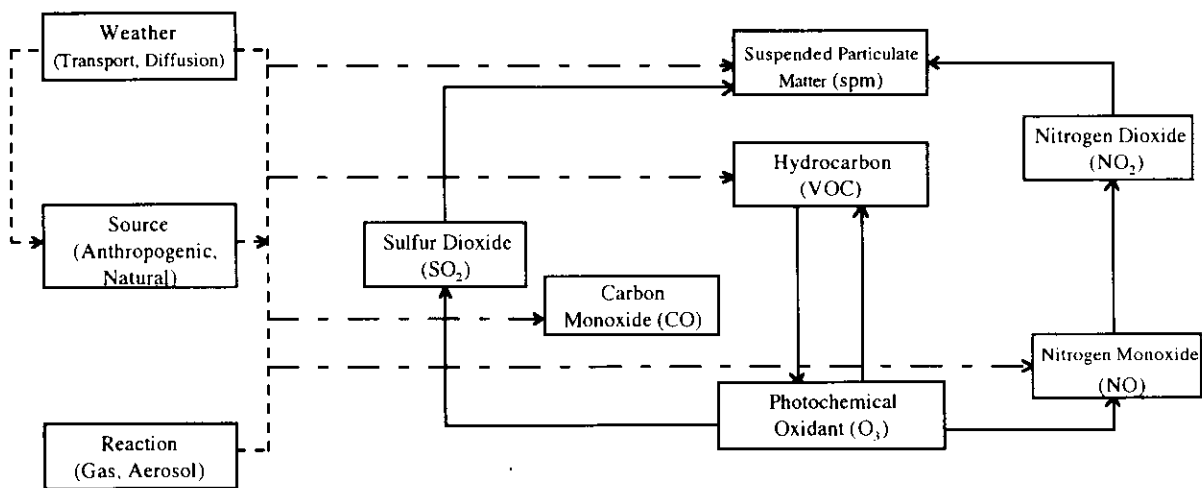
#### 4.3.1 Introduction

The concentration of air pollutants changes as a result of changes in the meteorological factor, chemical reactions in the atmosphere, variation of the amount of air pollutants released, etc. In the variation of concentration, diurnal, seasonal and annual variations should be considered. In this section, the governing factors for the variation of air pollutant concentration in each time scale will be explained.

#### 4.3.2 Diurnal Variation of Air Pollutant Concentration and the Mutual Relationship Among Pollutants

The weather and the chemical reaction give the largest effect on the diurnal variation of air pollutant concentration. Among the weather factors, the wind and stability have quite a large influence. Also, the amount of air pollutants generated has an influence on the diurnal variation. The primary factor of diurnal variation is the operational pattern of factories and the usage pattern of automobiles. It is also related with the life pattern of the people. For instance, in the diurnal variation of automotive traffic, a double-peaked distribution is common with peaks at morning and evening period of commuting hours, and the concentration increases significantly when stable, low wind speed weather condition would fall on those peaks.

Generally, the diurnal variation of pollutant concentration in the winter is different from that in the summer, and also very different by substance. In order to identify specific differences, it is necessary to understand the mutual relationship of air pollutants. The mutual relationship of air pollutants is shown in Fig.4.3.1.



Note: Dashed line indicates the flow of primary air pollution and solid line indicates that of secondarily generated air pollution.

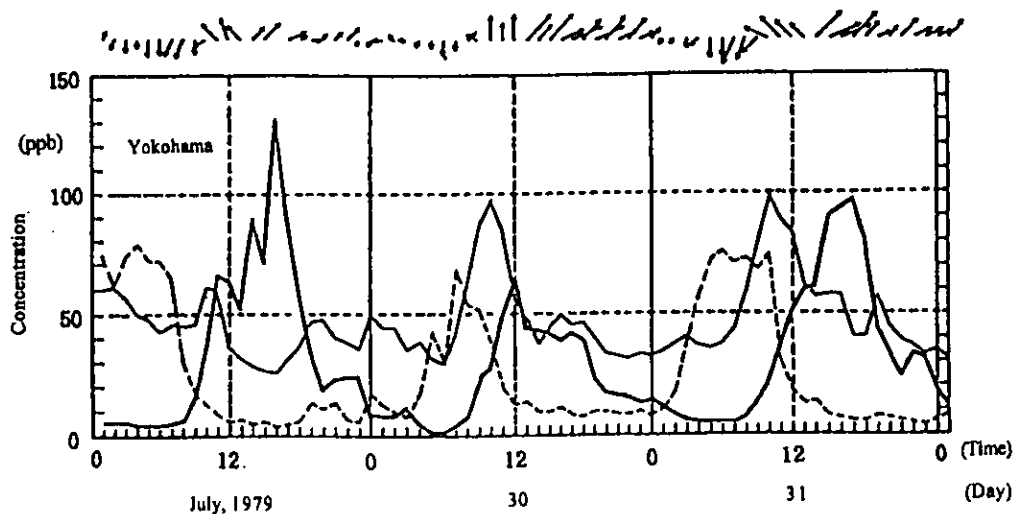
Source: Shinji Wakamatsu, Air Pollution Simulation Model, Air Conditioning and Sanitary Engineering, Vol. 70, (11), p.879-887 (1996)

Fig.4.3.1 Mutual Relationship of Air Pollutants

The dotted line in Fig.4.3.1 indicates the route in which air pollutants are released directly from the source of air pollutants. There are complex relationships among NO (nitrogen monoxide), NO<sub>2</sub> (nitrogen dioxide), VOCs (volatile organic compounds) and ozone. Some hydrocarbon species are included in the volatile organic compounds, but aldehydes are also included to be called generally as VOCs. Details of the reaction mechanism of hydrocarbon species are explained in Section 4.2, and the essential points will be described again below.

The VOCs would accelerate the formation of photochemical air pollution through the following mechanism. The major component of photochemical air pollution is ozone, and the only reaction that produce ozone in the troposphere is the photochemical decomposition of nitrogen dioxide. That is, nitrogen dioxide would be photochemically decomposed to nitrogen monoxide and oxygen, and this oxygen would combine with an oxygen molecule to form ozone. However, since nitrogen monoxide will react with ozone to become nitrogen dioxide again, the ratio of nitrogen monoxide to nitrogen dioxide to ozone becomes constant. This condition is called a photostationary state. When VOCs are not existing, such a stable condition will be established. However, when there are any VOCs in the atmosphere, the substance with very high reactivity called radical would be produced, and it will oxidize nitric oxide to nitrogen dioxide. As this reaction would produce nitrogen dioxide without destroying ozone, ozone would be produced further. Since more than 90% of the nitrogen oxide generated from automobiles and combustion facilities is nitrogen monoxide, the existence of highly reactive VOCs is a key for the increase of ozone.

This radical would also accelerate the production of aerosols (spm). Roughly more than 100 VOC components, which have up to 10 carbon atoms and are volatile, are existing in the atmosphere. Because each of those VOCs would have different rate of radical formation, the amount of each component contained is very important for ozone formation. As well as promoting the photochemical reaction, some of the VOCs such as benzene are hazardous by themselves. A typical diurnal variation of air pollutant concentration is shown in Fig.4.3.2.



Note: Shows the time variation of the concentration of NO (---), NO<sub>2</sub> (-), and Ox (-·-) and the wind direction and velocity at Yokohama in the period of July 29 to 31, 1979. There is a peak in nitrogen monoxide (NO) concentration in the early morning, and, after that the concentration of nitrogen dioxide (NO<sub>2</sub>) increases. As the sea breeze from the Bay of Tokyo, from southeast to south, excels in the late morning to midday, and then as that from the Bay of Sagami, from southwest, excels in the afternoon, the concentration of oxidant (Ox) increases.

Fig.4.3.2 Typical Diurnal Variation of Air Pollutant Concentration



### 4.3.3 Seasonal Variation of Air Pollutant Concentration

The concentration of ground level ozone of stratospheric origin sometimes increases to more than 80 ppb in the northern hemisphere at latitude of 30 degrees in the spring. In Japan, the concentration of nitrogen dioxide increases in Kansai region in the spring. Those phenomena are caused by the facts that the ozone of stratospheric origin would give influence on the ground in the spring, and that ozone is produced by photochemical air pollution.

In the summer, the photochemical air pollution would be generated as the intensity of ultraviolet radiation and the temperature are high. As a result, the amount of secondarily produced particulate matters would also increase. The ozone concentration in the oceanic air mass in the summer is relatively low with around 20 ppb.

In the winter, the air pollutant concentration becomes higher because the radiation inversion is often formed and stable atmospheric layers are formed during the night and early morning hours. However, as most people in Japan takes holidays in the new year season and the automobile traffic decreases, the air pollutant concentration decreases. Although the nitrogen oxides by background ozone or photochemical ozone would occur even in the winter time, its degree is lower than in the summer and the ratio of NO within NO<sub>x</sub> increases.

### 4.3.4 Annual Variation of Air Pollutant Concentration and Its Statistical Analysis

In order to evaluate the countermeasures for air pollution and to understand the structural changes of environmental loads, the information of the annual variation of air pollutant concentration is quite useful. It is necessary to remove the effect of weather conditions to correctly understand the characteristics of annual variation in relation to pollution sources.

As the environmental standard is usually determined as annual average values and hourly values, it is important to understand the relationship between average values and high concentration values to evaluate the environmental concentration of air pollutions. For these purposes, the analysis of the type of cumulative frequency distribution of hourly values and the relationship between percentile values and annual average values is performed. The evaluation by hourly values would respond to acute effects, while the analysis by annual average values would respond to chronic effects. Also, the evaluation by a cumulated concentration (dosage) in excess of certain threshold value is used in analysis of the damage to plants or the risk.

## 4.4 Distribution of Air Pollutants

### 4.4.1 Introduction

Both reaction and weather condition give effects to the distribution of air pollutants. Therefore, the movement of concentration peaks associated with the intrusion of air flow is frequently observed when the long range transport of air pollutant occurs. In this section, the characteristics of distribution of the air pollutant, particularly that produced by photochemical reactions, will be explained.

### 4.4.2 Flow of Air Pollution from City to Suburbs

The chemical reaction and the weather condition have compound influence on the formation of air pollution. In the Japanese metropolitan areas of Tokyo and Osaka, the stationary sources of air pollution are concentrated on coastal areas. Also, another source of pollution, the automobile, is distributed throughout the city area. Furthermore, the form of appearance of the air pollution differs by urban and rural areas as it is strongly affected by local air flows typically represented by the sea-land breeze. The schematic diagram for spatial distribution of the air pollution in urban area is shown in Fig.4.4.1.

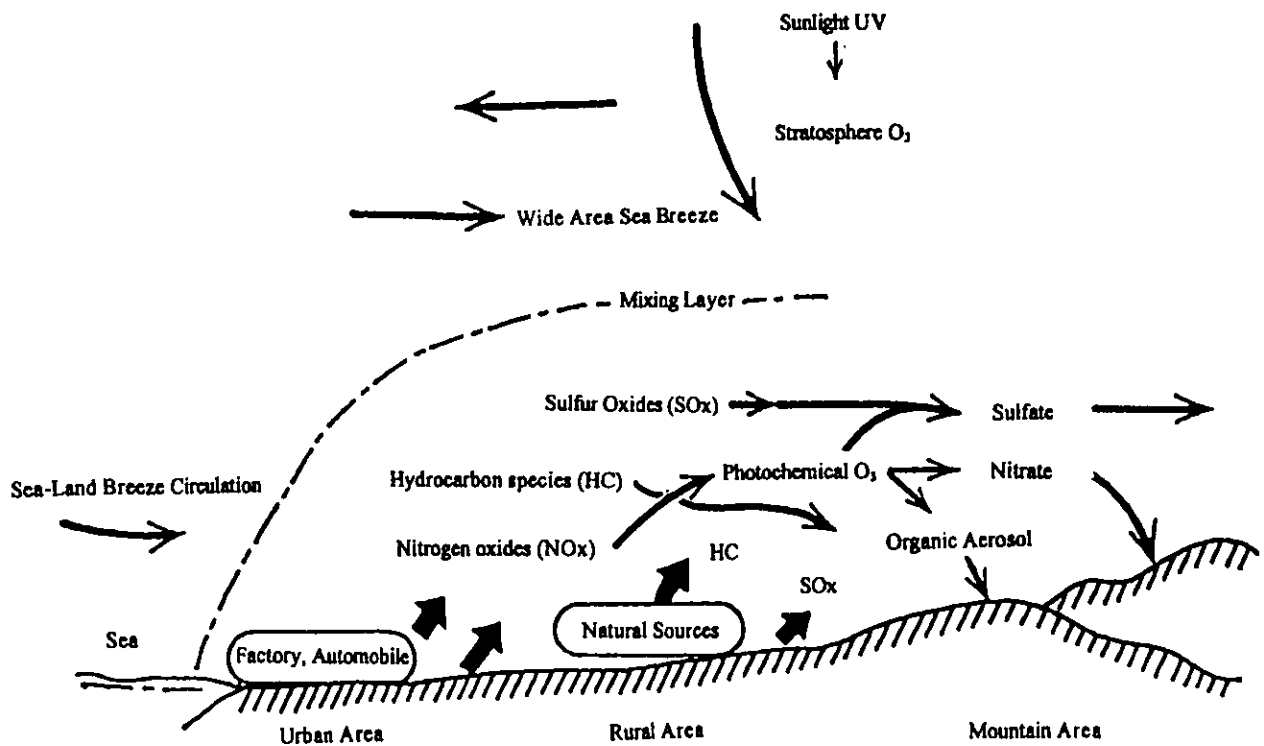
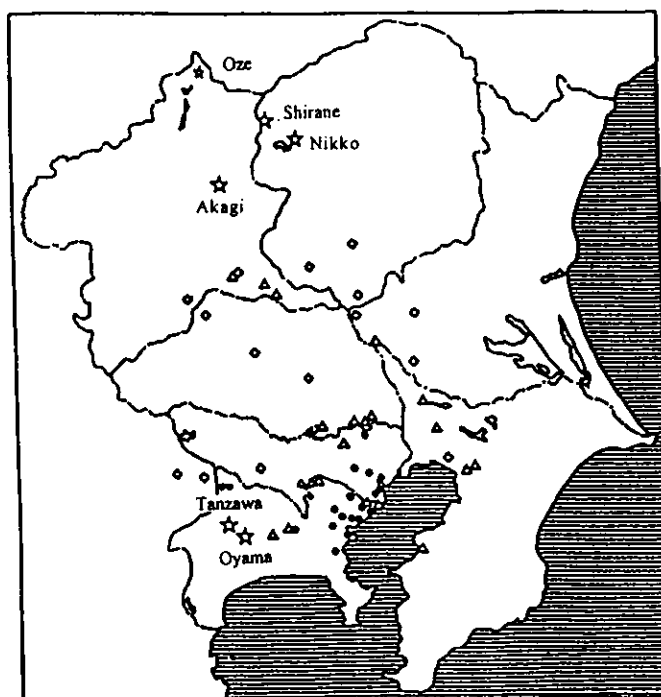


Fig.4.4.1 Schematic Diagram of Spatial Distribution of Air Pollution  
 —Transport and Chemical Change of Air pollutant from Urban to Rural Areas—

Generally, the concentration of nitrogen monoxide would first become high, and next that of nitrogen dioxide become high in urban area, and after that the concentration of ozone and aerosol would become high in rural areas.

Such characteristics are found in the time variation and area distribution of secondary products. Both chemical reactions and weather conditions are influencing the formation of such distribution. When there is a long range transport of air pollutants, it is frequently observed that the peak of concentration moves with an intrusion of air flow. An example of analysis of the distribution of air pollution in Kanto area is shown in Fig.4.4.2. The situation of transport and chemical change of the air pollutant from urban to rural areas are seen in this figure.



- : NO<sub>2</sub> Top ranked stations for annual average values (ranked among national worst 20)
- △: SPM Top ranked stations for annual average values (ranked among national worst 20)
- ◇: OX Stations with many measurement days when hourly value in excess of 0.12 ppm was observed (ranked among national worst 20)
- ☆: Sites where effects to plant were recently reported in the newspaper, etc.

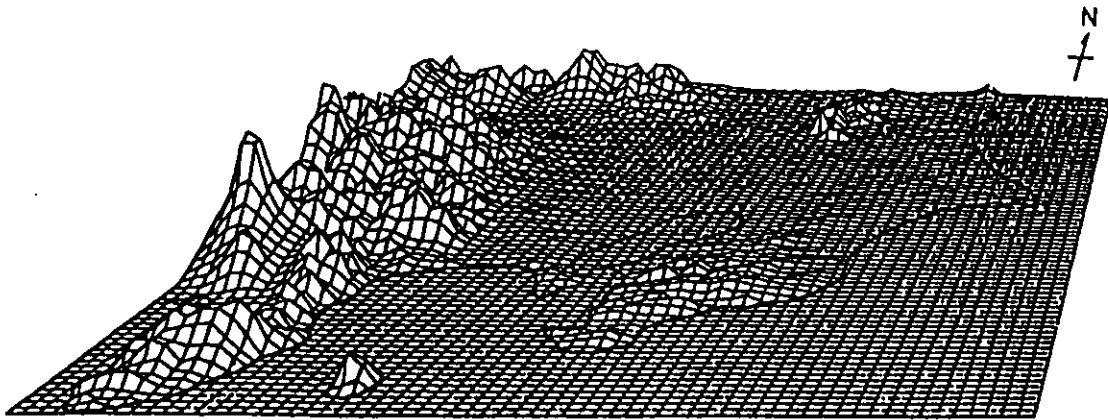
Note: All data are taken from the FY1992 General Atmospheric Measurement Station Data.  
 Source: Yagishita, S., Matsugu, H., and Ohi, M. Consideration on the Development of Total Strategy toward Measures for Suspended Particulate matters, J. of Atmospheric Environment Society, Vol. 31, No. 2, P. A26 (1966)

Fig.4.4.2 Example of Analysis of the Distribution of Air Pollution in Kanto Area

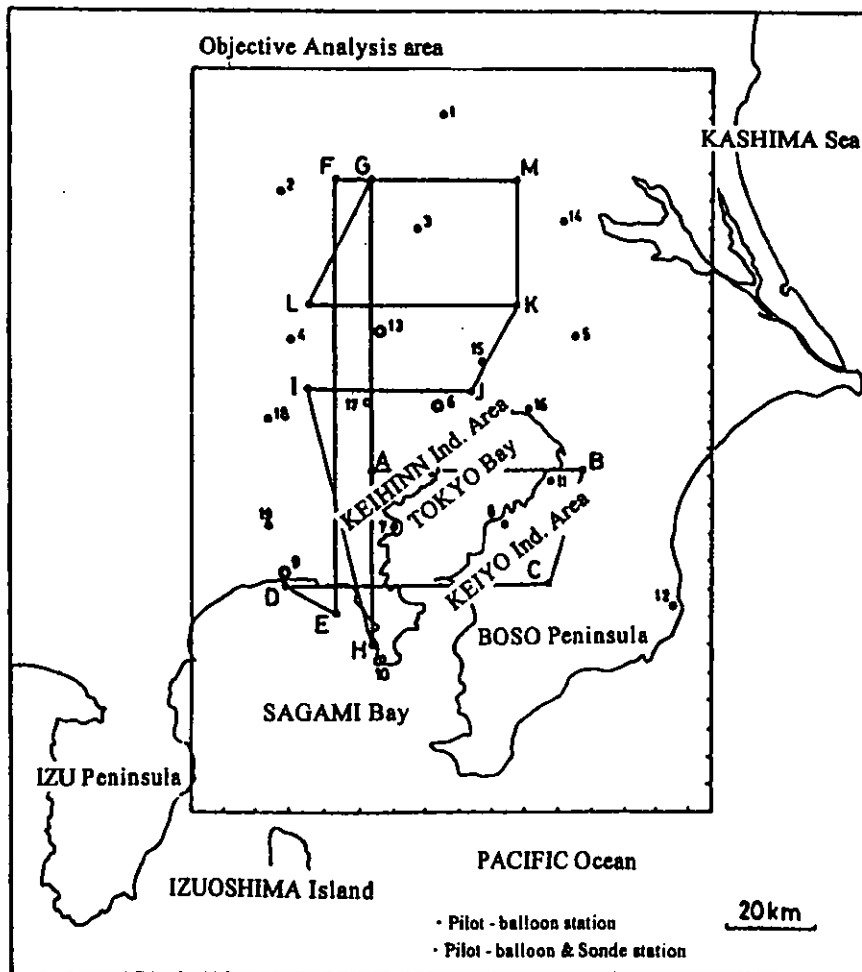
#### 4.4.3 3D Distribution of Air Pollution

The 3D distribution (the three dimensional distribution) of air pollution depends on the height of source and the pollutant. The 3D distribution of single stack smoke from the stationary source such as the boiler is described in the predictive model of atmospheric diffusion in Chapter 10. In this section, the characteristics of 3D distribution of the air pollution of urban scale will be described. A typical vertical cross sectional distribution when the photochemical air pollution occurs is shown in Fig.4.4.3. It could be seen that, in the result of aerial observation

from July 31, 1979 to August 1, there is a close relationship between the distribution of  $O_3$  and  $NO_2$  and the sea-land breeze circulation or the vertical distribution of temperature. The height of mixing layer on that day was about 1500 m, but the vertical distribution of highly concentrated ozone differed greatly by in and out of the mixing layer, and the time variation coincides well with changes in the sea-land breeze. Also, a highly concentrated photochemical ozone in excess of 120 ppb, formed at the altitude from 500 to 1200 m in the previous day, was stagnant in the night through early morning. When the general wind is weak and the local sea-land breeze circulation would continue for several days, the accumulation of pollution would occur as the above described situation repeatedly occurs.

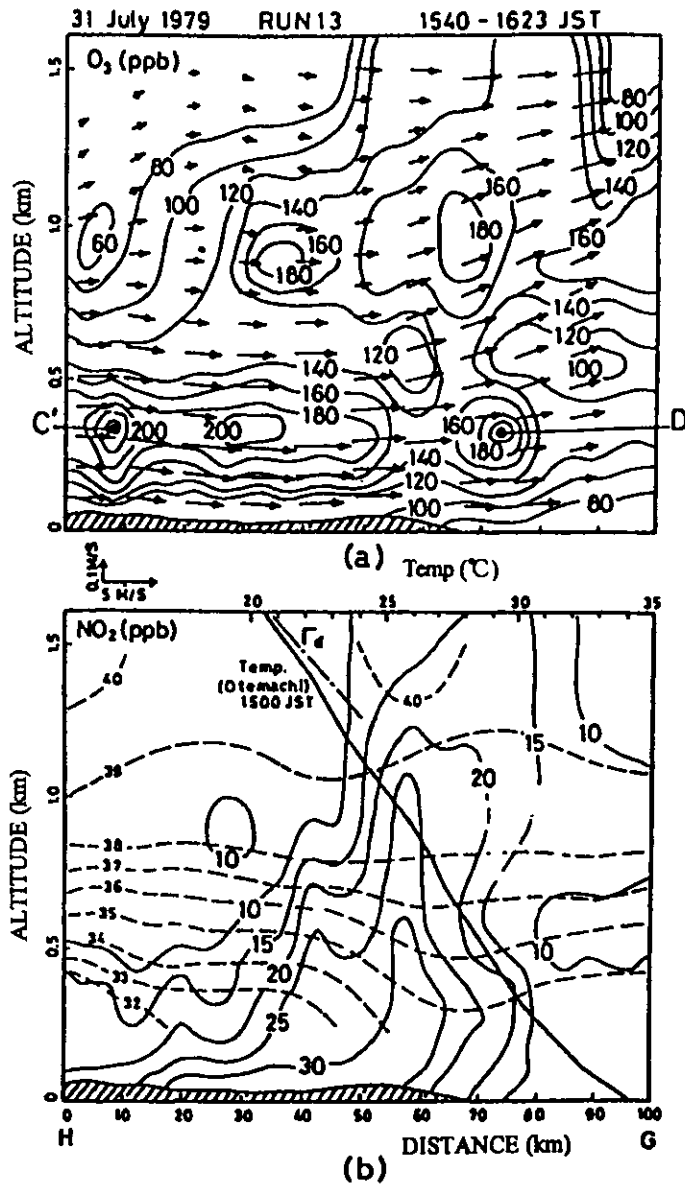


Topography of the Tokyo Metropolitan Area viewed from the south-east direction.  
Horizontal grid size is about 3 km square.



Map of Kanto District. flight paths and objective analysis area of wind field.

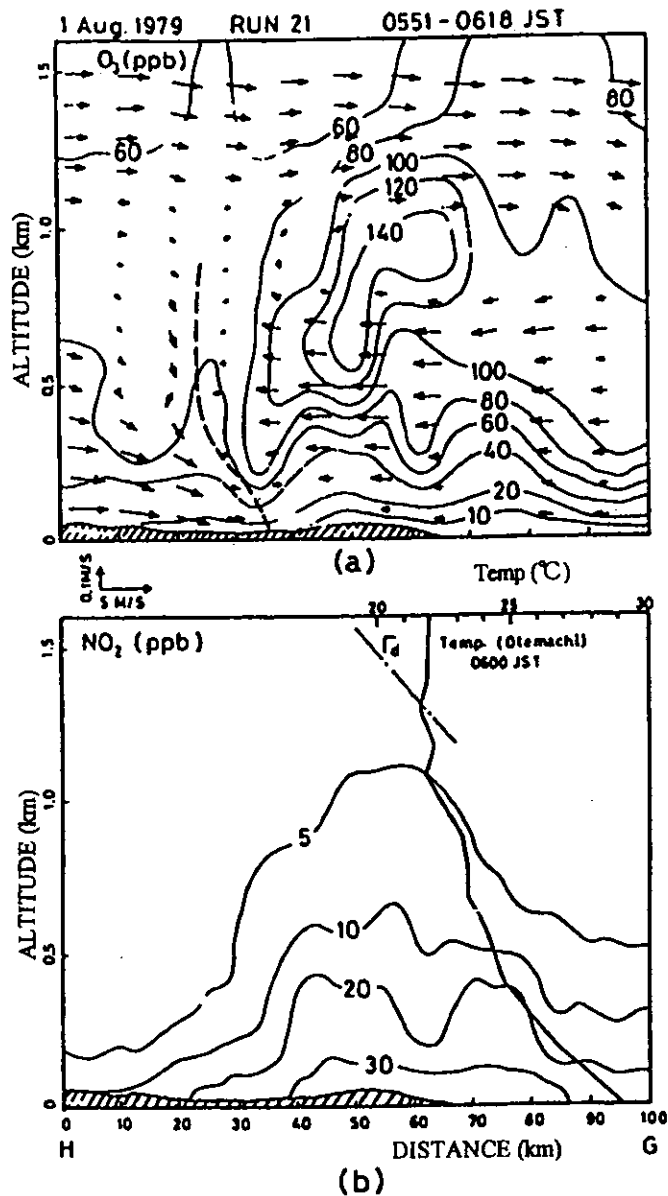
Pilot-balloon and sonde observation points are as follows: 1. Oyama; 2. Kumagaya; 3. Satte; 4. Iruma; 5. Inzai; 6. Otemachi; 7. Yokohama; 8. Sodegaura; 9. Chigasaki; 10. Miura; 11. Ichihara; 12. Ohara; 13. Urawa; 14. Tsukuba; 15. Nagareyama; 16. Tsudanuma; 17. Musashino; 18. Hachioji; 19. Atsugi.



Vertical distribution of pollutants in the afternoon of 31 July 1979 (RUN 13: 1450-1630JST. Cross-section H-G). (a)  $O_3$  distribution and vertical wind profile ( $v$  and  $w$  components) calculated from the modified MATHEW method. Vertical wind component  $w$  is emphasized (see wind scale). The label 'C' and 'D' in (a) indicate the marked air masses of Trajectories C and D in Fig. 10. (b)  $NO_2$  distribution and potential temperature (dashed line) and temperature profile at Otemachi (1500JST).

Source: Uno, I., Wakamatsu, S., Suzuki, M. and Ogawa, Y., Three-dimensional behavior of photochemical pollutant over the Tokyo metropolitan area, Atmospheric Environment, 18, 751-756 (1984)

Fig.4.4.3.1 An Example of 3D Distribution of Air Pollution in Kanto Area



Vertical distribution of pollutants in the early morning of 1 August 1979 (RUN 21: 0445-0620 JST, cross-section H-G). (a)  $O_3$  distribution and vertical wind profile (v and w components) calculated from the modified MATHEW method. The hard dashed line indicated the discontinuity line of wind and in the near of this line, the small downdraft wind zone are detected. (b)  $NO_2$  distribution and temperature profile at Otemachi (0600 JST).

Source: Uno, I., Wakamatsu, S., Suzuki, M. and Ogawa, Y., Three-dimensional behavior of photochemical pollutant over the Tokyo metropolitan area, Atmospheric Environment, 18, 751-756 (1984)

Fig.4.4.3.2 An Example of 3D Distribution of Air Pollution in Kanto Area

#### 4.4.4 Distribution of Wide Area Air Pollution

The weather condition has an important role in determining the regional distribution and time variation of photochemical ozone. Large cities in Japan such as Tokyo, Osaka and Nagoya receive strong influence from the

sea-land breeze because they are located in the coastal area. The air pollutants are transported toward inland by a sea breeze, undergoing chemical reactions on the way. As nitrogen monoxide reacts readily with ozone or  $\text{RO}_2$  radicals in the atmosphere to form nitrogen dioxide, a concentration peak of nitrogen dioxide appears usually near the central urban area. Furthermore, in the process of its inland transport, the concentration of photochemical oxidant and secondary aerosol increases. When highly reactive hydrocarbon species components are discharged in large amount, the production rate of secondary air pollutant such as photochemical oxidant and aerosols is increased and a high concentration appears nearer to the center of the city. Also, when the wind velocity is high, the air pollutants are transferred faster to the inland. In this way, the chemical reaction and weather would determine the time variation and area distribution of air pollutant concentration, giving influence to each other. Depending on the weather condition, the air pollutants in and around the urban area are transported further to the inland to bring about a wide area air pollution.

The contents of air pollution is changing as the structure of environmental loads and the social system changes. The problems in atmospheric environment in Japan are becoming more and more widespread and diverse to have very close relationships with our life style. The primary source of today's urban air pollution is the automobile. As a result of the increase in number of automobiles and the expansion of road network, the amount of air pollutant including nitrogen oxides is increasing and becoming widespread. The increase in total amount of air pollutant is altering the problem of atmospheric environment qualitatively. Because of these changes, the air pollution is spreading from central city areas toward surrounding areas, in both Kanto area centered around Tokyo and Kansai area centered around Osaka. Associated with these changes, the frequency of appearance of relatively high concentration of photochemical air pollution and secondary aerosols is increasing in suburban areas rather than in urban areas. Also, as the pollutants become to contain not only a single substance but several substances, it becomes necessary to consider the mutual relationship of pollutants and other hazardous chemical species. On the other hand, the air quality problems spreading across the boarder should be seriously considered. Concerning to the aerosol and acid deposition, the contribution of transport across the sea caused by a rapid increase in the amount of air pollution from Asian countries, particularly China and Korea, can not be ignored, and the monitoring and data analysis for this problem is now very important.

#### 4.4.5 Measurement of Distribution and Monitoring of Air Pollution

Against the air pollutants for which environmental standard and guidelines are determined, a constant monitoring based on the measurement of hourly average values is performed. Across the country, 1728 Air pollution monitoring stations and 395 automobile exhaust gas monitoring stations are in operation as of FY1994. In Japan, the wet method using absorbent solution has been used as a measuring method for sulfur dioxide, nitrogen dioxide, and photochemical oxidant, but measurements with the dry method are also admitted as equivalent. It is necessary to establish a method of calibration for the measuring equipment to proceed the measurement. Most of the presently existing measuring station for air pollution are concentrated in the urban area because those have been primarily established for monitoring pollution of higher concentration based on the historical background of taking measures against public hazards. As those stations are managed by each administrative district, the consideration for



the formation mechanism of air pollution is not always taken into account in determining their location. As the distribution and life time of pollution source are different by each pollutant, it would be reasonable to consider the density and measuring area of the station independently for each pollutant. It will be necessary to perform a timely review of the measurement methods and items and the density of measurement in accordance with the progress in measuring technologies and changes in social necessities while assuring the consistency of measurement, as well as to clarify the purpose of air pollution measurement. It is also important to know the environmental concentration of VOC for predicting its component. Presently, however, the measurement of VOC components is not performed at the monitoring station in this country at all, and only NMHCs (non-methane hydrocarbons) are measured by counting total number of carbon by the FID (flame ionization detector). It will be necessary in the future to analyze the major substances of VOC components automatically, and to evaluate the measured values.

#### (4.1) References

- 1) Chiaki, E., Moriguchi, M., and Ogawa, H.; "Environmental Pollution and Weather", Asakura Shoten (1990)
- 2) Environment Agency of Japan; "Report on Atmospheric Environment Study of the South Kanto Area" (1977)
- 3) Johnson, W.B., Ludwig, F.L., Dabbert, W.F., and Allen, R.J.; An Urban Diffusion Simulation Model for Carbon Monoxide, *J. of the Air Pollution Control Association*, 23, (6) 490-498 (1973)

#### (4.2) Literature cited

- 1) Finlayson-Pitts, B.J., and Pitts, Jr. J.N.; "Atmospheric Chemistry", p.429, John Wiley & Sons, Inc., New York (1986)
- 2) Singh, H.B., and Hanst, P.L.; *Geophys. Res. Lett.*, 8, 941 (1981)
- 3) Finlayson-Pitts, B.J., and Pitts, Jr. J.N.; "Atmospheric Chemistry", p.550, John Wiley & Sons, Inc., New York (1986)
- 4) Penkett, S.A. and Brice, K.A.; *Nature (London)*, 319, 655 (1986)
- 5) Calvert, J.G. ed.; "SO<sub>2</sub>, NO and NO<sub>2</sub> Oxidation Mechanism: Atmospheric Considerations", Butterworth, Boston (1984)
- 6) Pandis, S.N. and Seinfeld, J.H.; *J. Geophys. Res.*, 94, 1105 (1989)
- 7) De More, W.B., Sander, S.P., Golden, D.M., Molina, M.J., Hampson, R.F., Kurylo, M.J., Howare, C.J., and Ravishankara, A.R.; "Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling, Evaluation Number 9", p.217 JPL Publication 90-1 (1990)

#### (4.3) References

- 1) Wakamatsu, S.; Simulation of Air Pollution, *Air Conditioning and Hygiene Engineering*, 70, (11) 879-887 (1996)
- 2) FY1981 Consignment Study Report for Environment Agency of Japan, Literature Review of Photochemical Oxidant, Science and Technical Information Center of Japan (1982)
- 3) Committee on Tropospheric Ozone Formation and Measurement: Rethinking the Ozone Problem in Urban and Regional Air Pollution, National Academy Press (1991)

#### (4.4) References

- 1) Matsugu, H., Ohi, M., and Yagishita, S.; Consideration on the Development of Total Strategy toward Measures for Suspended Particulate Matters, *J. Atmospheric Environment Society*, 31, (2) A23-A33 (1966)
- 2) Ohara, T., Uno, I., and Wakamatsu, S.; Recent Trend in Precursor Concentrations and Oxidant Distribution in the Tokyo and Osaka Areas, *Atmospheric Environment*, 30, 715-721 (1996)
- 3) Ogawa, M., Suzuki, Y., Uno, I., and Wakamatsu, S.; Three-Dimensional Behavior of Photochemical Pollutant over the Tokyo Metropolitan Area, *Atmospheric Environment*, 18, 751-761 (1984)
- 4) Kurita, H., Muroga, H., Sasaki, K., Ueda, H. and Wakamatsu, S.; Long-Range Transport of Air Pollution under Light Gradient Wind Conditions, *J. Climate and Applied Meteor.*, 24, 425-434 (1985)

## Chapter 5 Effect of Air Pollution

### 5.1 Health effects on human

#### 5.1.1 Introduction

In this section, effects of typical air pollutants upon our health will be described. Chronic damage to respiratory organs is important in many cases. In some cases where the ambient air is frightfully polluted, however, eye and/or airway irritations (involving tears, cough, sore eyes and/or throat, etc.) become additional problems.

#### 5.1.2 Respiratory system

The first target organs attacked by air pollutants are respiratory system. Therefore, let us gain a general understanding of them in advance. Fig. 5.1.1 outlines the respiratory system of human. From the nasal cavity to near the bronchi, which constitute the passage of air, mucus covers the mucous epithelium. The airway of trachea and the bronchi are provided with cilia to eliminate foreign substances. Also there are alveolar macrophages for phagocyte in the alveola of the lungs exchanging carbonic dioxide for oxygen.

Of the air pollutants inhaled, the larger particulate matters are caught in the nasal cavity. However, so-called suspended particulate matters, measuring  $10 \mu\text{m}$  or less in diameter pass through the nasal cavity to reach the trachea and/or the alveola. Of the gaseous substances, sulfur dioxide, which is soluble in water, is absorbed mainly in the upper airway, causing chronic bronchitis or asthma. On the other hand, ozone, nitrogen oxides and other insoluble gaseous substances advance deep in the lungs, causing asthma or chronic bronchitis or possibly pulmonary emphysema. Also, carbon monoxide, when coming into contact with hemoglobin contained in the blood in alveola, disturbs transportation of oxygen by the blood because the substance combines with hemoglobin more easily than oxygen.

#### 5.1.3 What respiratory disease are induced by air pollutants?

In the case where the air is severely polluted, aged persons and patients with certain chronic base diseases in particular are in danger of excess death by suffering from acute bronchitis or so (refer to the section on the history of air pollution). In many cases, however, air pollution causes chronic respiratory disease, especially asthma, chronic bronchitis and lung emphysema. These respiratory diseases are known generally as chronic obstructive pulmonary diseases (COPD). Characteristics of these diseases as revealed by the pulmonary function check include smaller values for the forced expiratory volume % in one second (what percentage of their whole expiration the patients can put out in one second when they breathe out as fast as they can) and lower values for the maximum peak flow, while the vital lung capacities of the patients remain normal.

Definitions of these diseases are given in Table 5.1.1. In the case of chronic bronchitis, patients' illness is characteristically diagnosed based on their subjective symptoms such as continuous coughing and phlegm. Asthma

has come to be defined differently, now being considered to be inflammatory injury to the airway based on recent studies. Incidentally, causes of these diseases are not limited to air pollution.

Other diseases that can be caused by air pollution include lung cancer. It can be caused by asbestos, as is well known. Concerning smoke and soot or exhaust gas from diesel engines, because carcinogens have been found sticking to some particles contained in it, there is a suggested possibility of it causing lung cancer. However, a sequence of cause and effect between them is yet to be established.

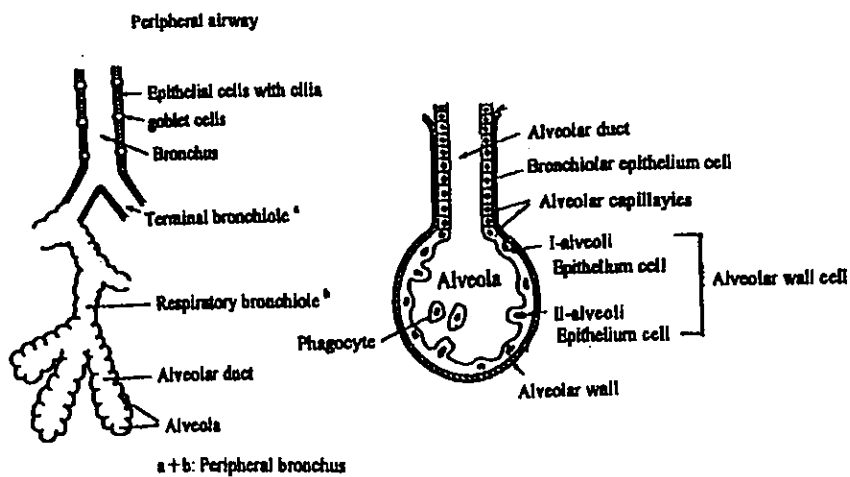
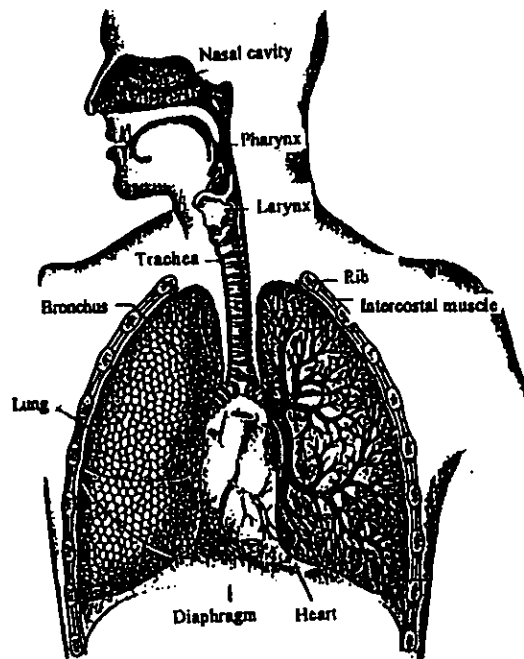


Fig.5.1.1 Respiratory System

Table 5.1.1 Definitions of Chronic Obstructive Lung Diseases

Names of diseases:	Definition
1) Chronic bronchitis	The clinical image of this disease is that excessive mucus is secreted in the bronchus and that phlegm is cough out chronically or repeatedly. In most cases, these symptoms continue almost every day for at least three months each year for at least two consecutive years. (ATS)
2) Asthma	The main characteristic of this disease is exacerbation of reaction to various stimuli of the trachea and the bronchus and its most important symptom is stenosis of extensive parts of the airway, whose intensity change spontaneously or can be changed through treatment. (ATS) The most recent definition of the disease, which recognizes that "the existing definition of asthma is unsatisfactory because it is impossible to fully understand the nature of the disease from its viewpoint of obstructed airway and airway hyperreactivity," is: "chronic inflammatory injury to the airway in which many cells such as mast cells and eosinophils are implicated."
3) Pulmonary emphysema	In this disease, an anatomical change to a morbid state is presented: this disease is mainly characterized by enlargement of air chambers from the ends of the tracheae to the farthest ends of the whole respiratory organ, which is accompanied by destruction of alveolar walls. Only few cases are peculiar clinically. However, the X-ray photos of some cases do not reveal the disease. In many cases, the disease is accompanied by usual coughing, which in turn is accompanied by wheeze. Its patients find it difficult to breathe and feel very much fatigued. (ATS)

#### 5.1.4 Health effects of typical air pollutants

##### (1) Sulfur dioxide

Main air pollutants generated when coal is burnt are sulfur dioxide and smoke and soot. In the famous episode of excess deaths due to air pollution in London, many aged people with respiratory or cardiovascular diseases died. Their common symptom was bronchial stenosis accompanied by dyspnea<sup>1)</sup>. At that time, the peak concentration of sulfur dioxide was about 1.4 ppm. It is now considered that these acute symptoms occur furiously when particulate matters such as smoke and soot exist together rather than when only sulfur dioxide exists. Later as coal was replaced by oil as fuel, smoke and soot decreased. But concentration of sulfur dioxide has increased inversely, with air pollution having come to occur in more extensive areas.

In Japan, the incidence of "Yokkaichi asthma" is famous. It was characterized by very high peak concentrations of sulfur dioxide, which often reached 1 to 2 ppm, measured in Isozu area, where the maximum amount of air pollutants landed from the chimneys in and around Yokkaichi City. In Isozu area, the rate of consultations with physicians about any of the four diseases related to the air pollution, i.e. common cold syndrome, bronchial asthma and pharyngo-laryngitis (including tonsillitis and angina) and symptoms of eyes (including conjunctivitis, keratitis, trachoma and eye irritation) was obviously higher than in any other area in or around the city<sup>2)</sup>. It is considered that the incidence of common cold syndrome included many cases of chronic bronchitis because in those days physicians had not in general made it a practice to use the term, "chronic bronchitis". Fig.5.1.2 shows the trends of the new incidences (three-year moving average) of three diseases of bronchial asthma, chronic bronchitis

and asthmatic bronchitis (children) and frequency rate of the peak concentration (more than 0.2 ppm) of sulfur dioxide in Isozu area from about 1960 through 1970. We can see that, after the start of full-scale operation of the industrial complex in and around Yokkaichi, first the incidence of bronchial asthma, next that of chronic bronchitis and then that of children's asthmatic bronchitis increased and that, since around 1965 when concentration of sulfur dioxide started to be decreased through the adoption of various counter measures, these incidences also started to decline<sup>3)</sup>.

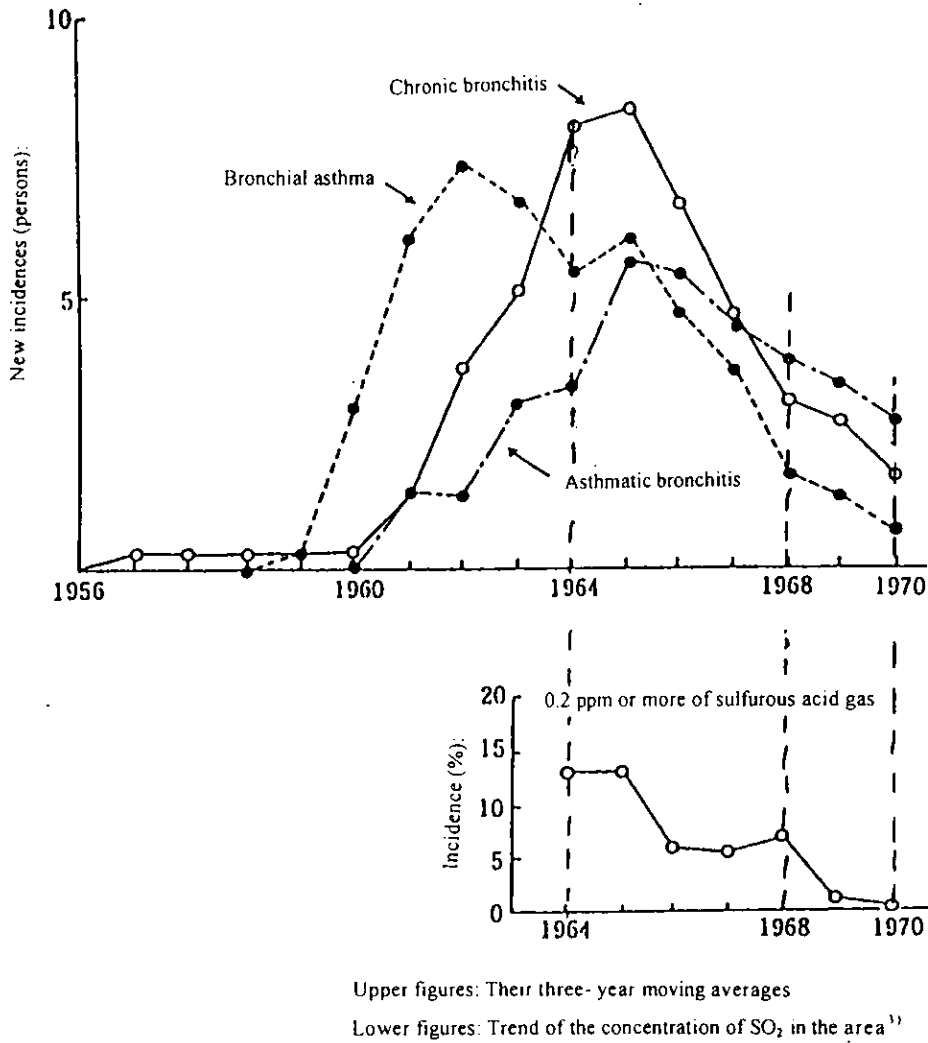


Fig.5.1.2 Trends of the New Incidences of Air Pollution-Related Diseases in Isozu Area of Yokkaichi City

## (2) Particulate matters

Smoke and soot generated when coal or oil does not attain a complete combustion and diesel exhaust particles whose main component is carbon. Because many of these particles measure less than 2.5  $\mu$  m in diameter and can go deep into alveola of the lungs, they may potentially damage to health. On these particles, lots of hazardous substances generated at the same time are stuck. Particularly when these particles exist together with sulfur dioxide, they cause much severe respiratory injury, as described above. Asbestos fibers inhaled induce pneumoconiosis, lung cancer or malignant mesothelioma. In countries where leaded gasoline is consumed, lead in exhaust gas from automobiles has posed problems, causing chronic toxicoses such as anemia, coloring of the gingivae and

neurological diseases. It has been established recently that the concentration of particulated matter has a relationship to the mortality and the number of inpatients with respiratory diseases <sup>4)</sup> and that has adjuvant effects on produce of antibodies that take part in allergic diseases.

### (3) Photochemical oxidants

The main component of photochemical oxidants is ozone, which accounts for almost all part of the substance. Effects of photochemical oxidants upon human health, therefore, are largely induced by ozone. It is considered, however, eye irritation, of which people often complain as an acute symptom, is not caused by ozone alone but is caused when peroxyacetylnitrate (PAN), formaldehyde and so on are also active. This eye irritation, of which people complain most often, is considered to be the most sensitive indicator of effects of photochemical oxidants. It can rightly be considered that the threshold value of photochemical oxidants for eye irritation is 0.1 (0.08-0.15) ppm <sup>5)</sup>. When the hourly average value of concentration of oxidants reaches 0.2 ppm approximately, the respiratory function generally starts to reduce (the forced expiratory volume % in one second decreases, etc.). It has been reported, however, that, even when the value is 0.12 ppm or less, people taking outdoor exercise can present the same symptom <sup>7)</sup>. Concerning long-term health effects of photochemical oxidants upon human, there is a suggested possibility of the substances affecting the incidence of respiratory diseases. However, their contribution rates are not yet to be established.

### (4) Nitrogen oxides

The current Japanese environmental standards were adopted in 1972 report based on results of the following researches including the respiratory function test also mentioned below: epidemiological survey that established that the incidence of continuous coughing and phlegm is in direct proportion to concentration of nitrogen oxides and the respiratory function test made at the same time that revealed that, as concentration of nitrogen oxides increases, the forced expiratory volume % in one second and the peak flow described above decrease <sup>8)</sup>. In addition, researches conducted recently have discovered that in areas where the annual average value of the concentration of nitrogen oxides exceeds 0.03 ppm (cf. 0.06 ppm as the environmental standard), the new incidence of asthmatic symptoms among children tends to be higher <sup>9)</sup>. We must consider that other factors may also work to bring about these research results. However, results of most experiments with animals also support the research results as facts.

As described above, when air pollutants effect our health, many factors are combined to bring about each symptom. Therefore, it should be necessary to grasp which air pollutant affects health most in each area concerned.

## 5.2 Effect on Vegetation

### 5.2.1 Introduction

In terms of the damage to plants caused by air pollution, it was observed that forests were damaged and agricultural areas showed worsened growth and yield. This was caused by the sulfur dioxide (SO<sub>2</sub>) and hydrogen fluoride (HF) from nearby stationary sources. Plant damage was also observed resulting from mobile sources including automobiles which were spurred by the crowded of populations in cities. This plant damage was caused by photochemical oxidants, originating from nitrogen dioxide and hydrocarbon species, etc.

### 5.2.2 Expressions of visible plant damage

Plant damage resulting from air pollution largely entails the visible damage on leaf surfaces, etc. Table 5.2.1 lists each type of pollutant that causes visible damage. It also presents the threshold concentration of these gases which induce the visible damage. Fig.5.2.1 also divides characteristics of the visible damage into four types and different levels caused by pollutant gases are expressed by+figures. Photographs of examples of damage are also presented.

Table 5.2.1 Apparent symptoms of acute damage of plants caused by air pollutants and threshold concentration at which damage developed

Air pollutants	Thresholds value at which acute symptoms developed	Main symptoms
Sulfur Dioxide (SO <sub>2</sub> )	0.1 - 1.5 ppm	Random spots between leaf veins, brownish-yellow spots on the marginal areas of the leaf (broad leaves), browning on tip or mid-section (coniferous leaves), general spotting
Nitrogen Dioxide (NO <sub>2</sub> )	10 - 50 ppm	Similar to effects of sulfur dioxide
Ozone (O <sub>3</sub> )	0.05 - 0.2 ppm	White or brown spots spread evenly over leaf surface, necrosis, leaves falling prematurely, leaf curvature
PAN	0.05 ppm	Large silver or bronze spots on leaf surface (forming a horizontal band across the entire leaf surface)
Ethylene (CH <sub>2</sub> =CH <sub>2</sub> )	0.05 - 1.0 ppm	Epinasty of leaf blade, abnormal blooming (pistil degeneration, change from male to female flower, etc.), premature fruit falling, premature defoliation, enhanced yellowing
Hydrogen Fluoride (HF)	5 - 10 ppb	Chlorosis at leaf tip and leaf margin, necrosis
Chlorine (Cl <sub>2</sub> )	0.1 - 0.3 ppm	Chlorotic spots between leaf veins, yellowing of leaf tip

The dose response relation, the relationship between concentration of gases and period of exposure in regards to the visible damage stated above, is presented in inverse proportions. One factor which decides the plant's resistance to visible damage is the plant's absorbency. Fig.5.2.2 lists SO<sub>2</sub> absorption capacity on the abscissa and the corresponding level of apparent damage on the vertical axle. The graph reveals that higher absorbency corresponds to a plant being more easily damaged.







Symptoms of Damage				
	( Yellowing to browning of leaf tip and leaf margin )	( Spots between leaf veins )	( Small spots on leaf surface )	( Shininess of abaxial leaf surface change in color: silver gray~bronzing )
Pollutant type				
Hydrogen Fluoride	++	+		
Chlorine	++	+	+	
Ozone		+	++	
PAN		+		++
Sulfur Dioxide		++	+	
Sulfuric Acid Mist	+	+	++	( ++ observed frequently ) ( + observed sometimes )
Nitrogen Dioxide		++	+	

Fig. 5.2.1 Characteristics of symptoms of visible damage plant leaves by air pollutants (Yamazoe, 1975)

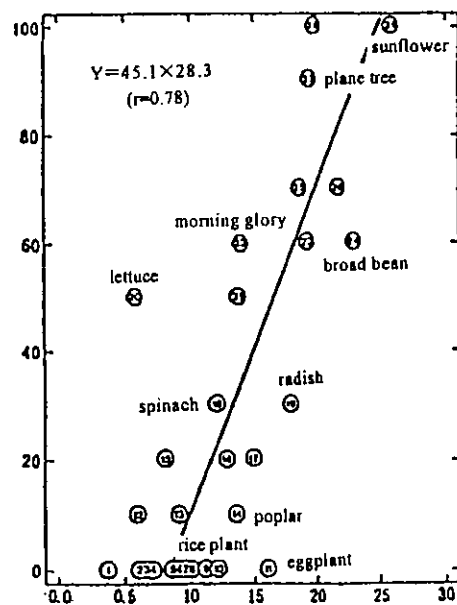
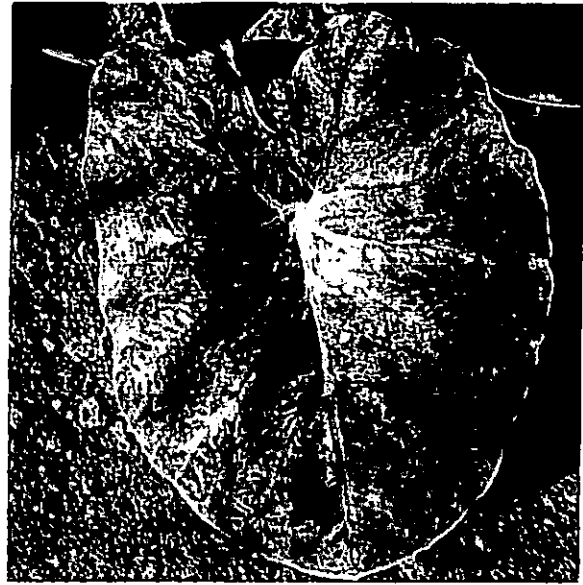


Fig. 5.2.2 The relationship between amount of SO<sub>2</sub> absorbed on leaf surface by SO<sub>2</sub> exposure and level of visible damage on leaf surface (Furukawa et al, 1980)



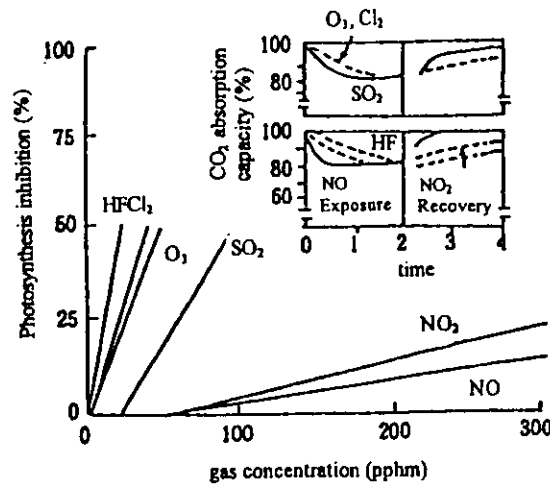
Foliar injury by ozone in morning glory.  
Upper; control, Lower; injured leaf.

Foliar injury by ozone in taro (upper) and spinach (lower).

Photo.5.1 Apparent symptoms of leaf damage caused by Ozon

### 5.2.3 The effect on plant's physiological function and yield

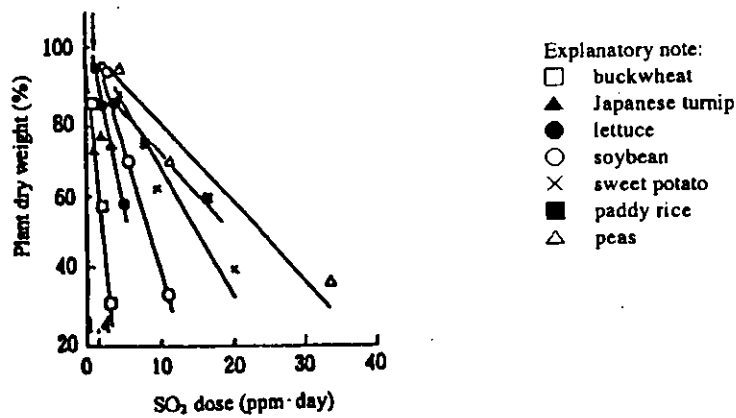
Although visible damage usually appears after short term exposure to high concentrations of pollutant gases, plant's fundamental physiological functions including photosynthesis, respiration, transpiration, etc., are affected and plant development stunted by long term exposure to low concentrations of pollutant gases as well. Photosynthesis is particularly sensitive to pollutant gases and is affected even by extremely low concentrations. Results of the effects of various gases on photosynthesis are presented in Fig.5.2.3. Level of photosynthesis inhibition is presented as a relative value (%) to the control on the ordinate. In the Figure lines with the greater slope show more sensitive, revealing the strength of the poisonous effects of these gases. Hydrogen fluoride (HF) is most toxic followed by gaseous chlorine ( $\text{Cl}_2$ ), ozone ( $\text{O}_3$ ), sulfur dioxide ( $\text{SO}_2$ ), and nitrogen dioxide ( $\text{NO}_2$ ). is most toxic followed by gaseous chlorine ( $\text{Cl}_2$ ), ozone ( $\text{O}_3$ ), sulfur dioxide ( $\text{SO}_2$ ), and nitrogen dioxide ( $\text{NO}_2$ ).



Note: 2 hour exposure for each pollutant gas concentration

Fig. 5.2.3 Effect of pollutant gas on the canopy net photosynthesis of barley and oats (Bennett and Hill, 1974)

When plant's photosynthesis is inhibited, the dry weight growth is affected adversely thereafter. Fig.5.2.4 presents the effect of sulfur dioxide exposure on dry weight growth of several crop plants (concentration ranging from 0.065 to 0.26 ppm, refer to Table.5.2.2). The abscissa plots the gas concentration times number of days of gas exposure (this is referred to as dose). Also, the ordinate presents the relative dry weight of plants as compared to control values. The more the figure line slopes the higher the plant's sensitivity. Also, the smaller the dose at which relative dry weight growth begins to be affected the greater the sensitivity of the plant. In the figures, buckwheat plants has the highest sensitivity. In Fig.5.2.4, line drawn in parallel with the 100% dry weight of the plant the  $\text{SO}_2$  dose at the point of intersection with each related line is  $1.4 \text{ ppm} \times \text{days}$ . Therefore, this value presents the dose limit at which these plants are affected. For instance, if the concentration of  $\text{SO}_2$  in the air is 0.02 ppm, the number of days of exposure at which there would be no effect would be  $1.4 \div 0.02 = 70$  days. Thus, crop plants harvested within around 70 days would not be affected by the concentrations of 0.02 ppm. Adversely, threshold  $\text{SO}_2$  concentration for plants requiring 100 days of growth periods would be 0.014 ppm. Table 5.2.3 presents data for pollutant gases other than  $\text{SO}_2$ .



Source: after Fujiwara (1975). (Adapted by Totsuka, 1979)

Fig. 5.2.4 The relationship between  $\text{SO}_2$  dose on several crops and the dry weight growth of plant weight (% to a control value).

Table 5.2.2  $\text{SO}_2$  exposure conditions for the crops applied in Fig. 5.2.4.

Species name	exposure time (days)	$\text{SO}_2$ concentration (ppm)	plant material measured
paddy rice	66	0.065-0.26	dry weight of whole plant
sweet potato	77	0.065-0.26	dry weight of whole plant
soybean	42	0.065-0.26	dry weight of aerial parts
peas	112	0.05-0.3	dry weight of aerial parts
Japanese turnip	14	0.065-0.26	dry weight of whole plant
lettuce	20	0.065-0.26	dry weight of whole plant
buckwheat	13	0.065-0.26	dry weight of whole plant

Source: Totsuka (1979), after Fujiwara (1975)

Table 5.2.3 Concentration threshold value for each gas pollutant causing growth damage

Threshold value concentration of NO <sub>2</sub>	Exposure period	Damage	Plant name	Reference
0.068 ppm	56~140 days	Smaller leaf area and decrease of plant dry weight	Kentucky blue grass	Ashenden (1979)
0.15~0.26 ppm	10~22 days	Reduced leaf area and decrease of plant dry weight	Tomato	Taylor and Eaton (1966)

Threshold value concentration of O <sub>3</sub>	Exposure time	Rate of inhibition	Plant	Reference
0.04 ppm	During entire growth period	Approximately 15% reduction in weight of rough rice	Paddy rice	Plant Environmental Research Institute (1981)
0.05 ppm	During entire growth period	0.05 ppm day/0.025 ppm night 23% less in yield	Paddy rice (Shinsenbon, medium rice)	(Chugoku Agric Inst. Soil Nutn. Lab.) (1981)
0.05 ppm	8hrs./day × 22 days	Growth inhibition (growth in middle)	Tomato(eiju)	Nat. Inst. (1981)
0.05ppm	8hrs./day × 26 days	Growth inhibition (leaf and root) thinning of leaf	Cucumber (tokiwa No.3 P)	

Threshold value concentration of PAN	Exposure time	Damage	Plant	Reference
0.003 ppm	16 months	Root growth, defoliation, concentration estimated by Matusoka	Navel orange	Thompson and Kats

Threshold value concentration of HF	Exposure time	Damage	Plant	Reference
2~3 ppb	13~26 months	Reduction of trunk diameter, tree height, width of branch, trunk volume and leaf surface, decrease of amount of fruit and particularly, 25%~35% reduction in leaf surface.	Navel orange	Brewer et al. (1960)
2.5 ppb	Until harvest	Reduced amount of fruit and seeds	4 types of kidney bean	PACK (1971)
0.7 ppb	43 days	25% reduction in wet weight of bean	Kidney bean	Maclean et al. (1969)

## 5.3 The effects of air pollution on materials and cultural properties

### 5.3.1 Introduction

The effects of air pollution are not only on peoples' health and living things such as plants, but also extend to man-made items such as materials like metals and cultural properties. To recognize and investigate the effects of air pollution on materials and cultural properties is to evaluate the economic loss from the air pollution and at the same time improve the safe maintenance of public assets, starting with municipal facilities. Furthermore, we must identify the long term effect on cultural properties, which are assets of all mankind but cannot have an economic value placed on them, and improve measures to preserve them. Because of this, each region and each individual country must investigate and identify their effects on the materials and cultural properties, and it is very important to apply these measures

### 5.3.2 The effects of air pollution relating to raw materials

The degree of the effect of air pollutants materials on materials is shown on Table 5.3.1<sup>1)</sup>. Hydrogen sulfide greatly affects silver and copper, and sulfuric acid has a large effect on iron, steel and stone. Also, ozone has a large effect on high molecule organic substances. In this way, depending on the material and its type, the degree of the effect of air pollutants will be different. Accordingly, knowing the characteristics of a region's air pollution is the foundation for knowing what materials will be affected to what degree by that region's air pollution.

Table 5.3.1 Damage sensitivity of materials to air pollutants

Materials \ Pollutants	Silver	Copper	Bronze	Brass	Iron	Steel	Tin	Lead	Pewter	Nickel	Aluminum	Stone	Polymers	Lacquer	Paper	Photographs	Textile
Water	L	L	L	L	H	H	L	L		N	L	L	N	N	H	L	N
Carbon dioxide	L	L	L		M	M		M		L							
Ammonia	M	M	L	L	M	M	L	L	N	N	L						
Nitrogen oxides	N								M	M	M	L			L	M	
Hydrogen sulfide	H	H	M	L	N	M	N	L	H	N	L			L		M	
Carbonyl sulfide	H	H															
Sulfur oxides	L	L	L	L	H	H	N	L	H	M	L	H		M	M	M	M
Hydrogen chloride	L	L	L	L	H	H	L	L		L	L	L					
Formaldehyde		L			L	N					N						
Organic acid	N	M	M	L	H	L	L	H									L
Ozone	M	M								M		M	H	M		L	L
Hydrogen peroxide					L						L					L	

H = High, M = Medium, L = Low, N = None, Blank = No data

The results of the Land Steel Preservation Research Council's Japan-wide investigation of SO<sub>2</sub>, which was a large air pollution problem from the late 1950's until the early 1970's, are shown in Fig.5.3.1<sup>2)</sup>. The rate of steel

corrosion in Kawasaki, an area with very heavy air pollution, was especially high, approximately ten times that of inland Takayama. By looking at this data we can see how much air pollution accelerates corrosion on materials.

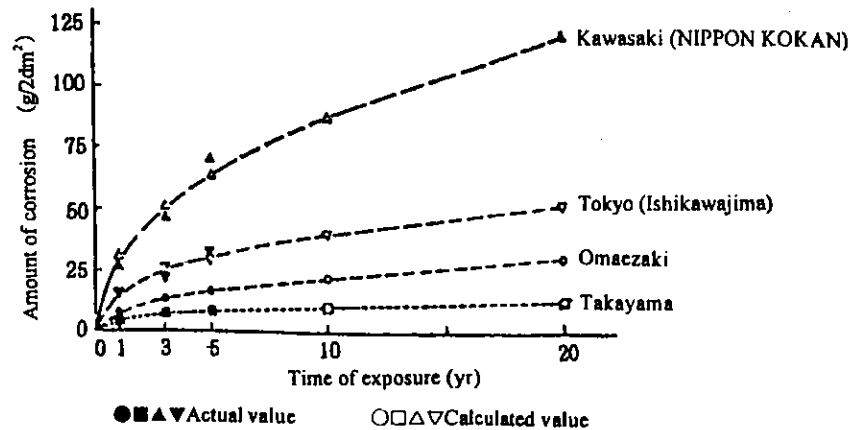


Fig.5.3.1 Amount of corrosion by calculated price of *kildo* metal

Using the example of a study of the relationship between air pollution and copper corrosion, the results of an investigation of the appearance of patina on copper outdoors in the eastern United States<sup>3)</sup> are shown in Fig.5.3.2. We can confirm from Fig.5.3.2 that as air pollution progresses copper corrosion accelerates.

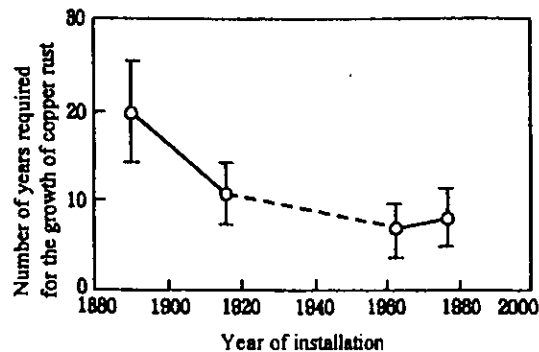


Fig.5.3.2 Number of years required for the growth of copper rust on copper surfaces installed each year in the eastern United States

Conversely, there is also the example of ranking the degree of pollution by the speed of corrosion on materials. The ranking by ISO material categories is shown in Table 5.3.2~5.3.4<sup>4)</sup>. Table 5.3.2 gives the categories of the major environmental factors which affect corrosion. Table 5.3.3 shows the speed of corrosion for different materials, but if the corrosiveness categories are all ranked the same, the speed of corrosion on steel is the highest, followed by zinc and copper, and aluminum is the lowest. The current corrosion rate of steel throughout Japan is in the lowest corrosion category. Table 5.3.4 shows the relationship between environmental factors and speed of corrosion by category of material. For instance, we can understand that aluminum has a rather high corrosion speed as  $S_3$ .

Table 5.3.2 Categorization of principal environmental factors

Time of wetness		SO <sub>2</sub> amount applied			Amount of sea salt particles	
No.	h/y	No.	mg/(m <sup>2</sup> ·d)	μ g/m <sup>3</sup>	No.	Cl <sup>-</sup> ·mg/(m <sup>2</sup> ·d)
τ <sub>1</sub>	≤ 10	P <sub>0</sub>	≤ 10	≤ 12	S <sub>0</sub>	≤ 3 (5)
τ <sub>2</sub>	> 10~250	P <sub>1</sub>	>10~35	>12~40	S <sub>1</sub>	> 3~60 (99)
τ <sub>3</sub>	> 250~2,500	P <sub>2</sub>	>35~80	>40~90	S <sub>2</sub>	>60~300 (495)
τ <sub>4</sub>	>2,500~5,500	P <sub>3</sub>	>80~200	>90~250	S <sub>3</sub>	>300~1,500 (2473)
τ <sub>5</sub>	>5,500					

Note: Numbers in parentheses are NaCl·mg/m<sup>2</sup>·d

Table 5.3.3 Atmospheric exposure, First year rate of corrosion (r<sub>cor</sub>)

Category number of corrosiveness	Rate of corrosion of metal (r <sub>cor</sub> )				
	Unit	Carbon steel	Zn	Cu	Al
C <sub>1</sub>	g/(m <sup>2</sup> ·y)	≤ 10	≤0.7	≤0.9	Negligible
	μ m/y	≤ 1.3	≤0.1	≤0.1	-
C <sub>2</sub>	g/(m <sup>2</sup> ·y)	> 10~ 200	>0.7~ 5	>0.9~ 5	≤0.6
	μ m/y	> 1.3~ 25	>0.1~0.7	>0.1~0.6	-
C <sub>3</sub>	g/(m <sup>2</sup> ·y)	>200~ 400	> 5~ 15	> 5~ 12	>0.6~2
	μ m/y	> 25~ 50	>0.7~2.1	>0.6~1.3	-
C <sub>4</sub>	g/(m <sup>2</sup> ·y)	>400~ 650	>15~ 30	>12~ 25	> 2~5
	μ m/y	> 50~ 80	>2.1~4.2	>1.3~2.8	-
C <sub>5</sub>	g/(m <sup>2</sup> ·y)	>650~1,500	>30~60	>25~ 50	>5~10
	μ m/y	> 80~ 200	>4.2~8.4	>2.8~5.6	-

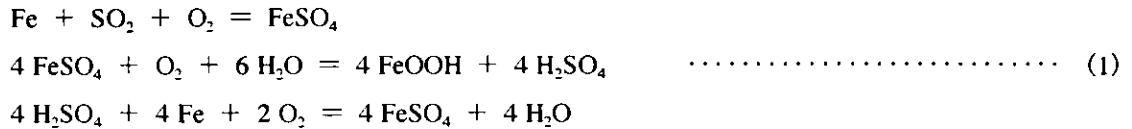
Table 5.3.4 Chart of categories of environmental corrosiveness by level of environmental factors

Metal	Carbon steel			Zn and Cu			Al		
	P <sub>0</sub> -P <sub>1</sub>	P <sub>2</sub>	P <sub>3</sub>	P <sub>0</sub> -P <sub>1</sub>	P <sub>2</sub>	P <sub>3</sub>	P <sub>0</sub> -P <sub>1</sub>	P <sub>2</sub>	P <sub>3</sub>
τ <sub>1</sub>	S <sub>0</sub> -S <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub>
	S <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>2</sub> or C <sub>3</sub>
	S <sub>3</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>2</sub>	C <sub>2</sub> or C <sub>3</sub>
τ <sub>2</sub>	S <sub>0</sub> -S <sub>1</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>1</sub>	C <sub>1</sub> or C <sub>2</sub>
	S <sub>2</sub>	C <sub>2</sub>	C <sub>2</sub> or C <sub>3</sub>	C <sub>3</sub>	C <sub>1</sub> or C <sub>2</sub>	C <sub>2</sub>	C <sub>3</sub>	C <sub>2</sub> or C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>
	S <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub>	C <sub>3</sub>	C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub>	C <sub>4</sub>
τ <sub>3</sub>	S <sub>0</sub> -S <sub>1</sub>	C <sub>2</sub> or C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub>	C <sub>3</sub>	C <sub>3</sub>	C <sub>3</sub>	C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>
	S <sub>2</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub> or C <sub>5</sub>	C <sub>5</sub> or C <sub>4</sub>	C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub> or C <sub>5</sub>
	S <sub>3</sub>	C <sub>4</sub>	C <sub>4</sub> or C <sub>5</sub>	C <sub>5</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub>	C <sub>4</sub>	C <sub>4</sub>	C <sub>4</sub> or C <sub>5</sub>
τ <sub>4</sub>	S <sub>0</sub> -S <sub>1</sub>	C <sub>3</sub>	C <sub>4</sub>	C <sub>5</sub>	C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub> or C <sub>5</sub>	C <sub>3</sub>	C <sub>3</sub> or C <sub>4</sub>
	S <sub>2</sub>	C <sub>4</sub>	C <sub>4</sub>	C <sub>5</sub>	C <sub>4</sub>	C <sub>4</sub>	C <sub>5</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>4</sub>
	S <sub>3</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>
τ <sub>5</sub>	S <sub>0</sub> -S <sub>1</sub>	C <sub>3</sub> or C <sub>4</sub>	C <sub>5</sub>	C <sub>5</sub> or C <sub>4</sub>	C <sub>3</sub> or C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>4</sub> or C <sub>5</sub>	C <sub>5</sub>
	S <sub>2</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>
	S <sub>3</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>	C <sub>5</sub>



### 5.3.3 Mechanisms for the effects of atmospheric pollution

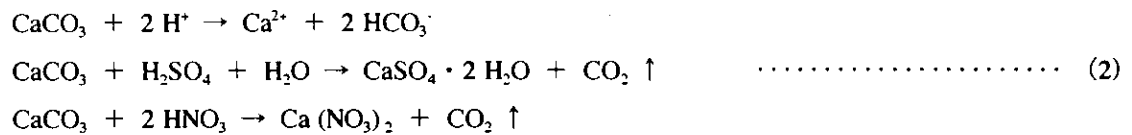
The Schikorr formula<sup>5)</sup> measures the chemical effect of SO<sub>2</sub> contribution to steel erosion and is a representative example of how air pollution contributes to the breaking down of materials. The following is example (1) of the Schikorr formula.



This structure is called auto-catalytic, the reaction repeatedly produces FeOOH and promotes corrosion.

With marble, acid substances in the atmosphere promote deterioration by the following process.

Atmospheric SO<sub>2</sub> and NO<sub>2</sub> oxidized in the atmosphere and become SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> which makes rain become acidic causing acid rain. Acid rain acts on marble as shown in Eq. (2) and promotes deterioration.



Also, air pollution promotes the deterioration of other metallic materials.

### 5.3.4 Conclusion

It is difficult to measure air pollution's effect on cultural properties and materials, and the method must include a wide variety of investigations, making it difficult to standardize, but if the conditions necessary to standardize investigations are clarified, it is important to investigate utilizing the composite examination results to create measures addressing air pollution.

## 5.4 Visibility degradation

### 5.4.1 Introduction

When visibility is degraded, the surrounding scenery becomes difficult to see clearly compared with normal conditions. People's first experience of air pollution is normally visibility degradation. Visibility degradation can also be caused by meteorological conditions such as fog, but this paper addresses the problem of visibility degradation due to air pollution.

### 5.4.2 Causes of visibility degradation

The light from the sun deteriorates through its being absorbed and scattered due to aerosols, absorption by air pollutants and water vapor, scattering by airborne particles, and so forth. The main cause of visibility degradation due to air pollution are aerosol and gasses in the atmosphere, but the visibility conditions can differ greatly due to atmospheric conditions such as humidity, the optical characteristics of the target matter, and the strength and distribution of the light at the time in question. When air pollution is severe, the atmosphere appears to be colored, but the color can vary depending on the type of pollution. Air pollution appears black when it is due to soot from the burning of fossil fuels, but has a whitish hue when it is due to photochemical air pollution.

#### (1) Extinction coefficient ( $b_{ext}$ )

If ( $I$ ) is the strength of the light when it reaches the eyes of a person at distance ( $x$ ) from the light ( $I_0$ ) of the visibility target, then Eq. (1) is established according to Lambert's rule:

$$I = I_0 [Exp(-b_{ext})]^x \quad \dots\dots\dots (1)$$

This is called  $b_{ext}$  extinction coefficient, and Eq. (2) can be obtained. The unit is  $m^{-1}$ .

$$b_{ext} = \frac{\ln I_0}{x} \quad \dots\dots\dots (2)$$

Because the primary cause of visibility degradation due to air pollution is the absorption and scattering of light, the extinction coefficient  $b_{ext}$  can be analyzed in four parts, as per Eq. (3) below. Namely, scattering coefficient of light due to gases:  $b_{sg}$  (this is normally called the Raleigh scattering coefficient), scattering coefficient of light due to particles:  $b_{sp}$  (scattering coefficient of light due to menute particles is normally called the Mie scattering coefficient), absorption coefficient of light due to gases:  $b_{ag}$ , and absorption coefficient of light due to particles:  $b_{ap}$ .

$$b_{ext} = b_{sg} + b_{sp} + b_{ag} + b_{ap} \quad \dots\dots\dots (3)$$

Of these four, scattering coefficient of light due to particles:  $b_p$  is vital for visibility degradation. The optical features of airborne particulate matter vary greatly depending on the diameter of the particles concerned. Light is refracted when the diameter of the particles is 10 or more times greater than the wavelength of the light. Rainbows caused by water droplets in the air are a phenomenon representative of this. In contrast, when both the wavelength of the light and the diameter of the particles are approximately the same size, scattering becomes the main effect. Because the mean wavelength of solar light is approximately  $0.52 \mu\text{m}$ , Mie scattering due to minute particles with an approximate diameter of between  $0.1$  and  $10 \mu\text{m}$  has the greatest effect on visibility deterioration.

(2) Threshold contrast value: According to results obtained in the laboratory under daytime conditions, the threshold contrast value which can be detected by humans is  $I/I_0 = 0.018$  to  $0.03$ .  $X$  is substituted by  $0.02$ , the mean value, in Eq. (2), Eq. (4) is obtained. This is called the Koschmeider formula.

$$b_{ext} = \frac{3.912}{x} \dots\dots\dots (4)$$

When the visibility ( $x$ ) is  $10 \text{ km}$ , the extinction coefficient  $b_{ext}$  becomes  $3.91 \times 10^{-4} (\text{m}^{-1})$ , and when visibility is  $1 \text{ km}$ , the extinction coefficient  $b_{ext}$  becomes  $3.91 \times 10^{-3} (\text{m}^{-1})$ . On the other hand, an atmosphere with no atmospheric pollutants is called ideal atmospheric conditions, and only the Raleigh scattering due to air molecules has any effect. The extinction coefficient when the elevation above sea-level is  $0 \text{ m}$  is approximately  $13.2 \times 10^{-6} (\text{m}^{-1})$ , and the threshold contrast distance becomes  $296 \text{ km}$ . It becomes approximately  $403 \text{ km}$  when the evaluation is  $3 \text{ km}$  aloft.

There has been little recent research into the relationship between visibility and the concentration of air pollution, but as an approximate measure, when the concentration of floating particulate matter in the atmosphere exceeds approximately  $0.12 \text{ mg per } 1 \text{ m}^3$ , visibility is reduced to  $10 \text{ km}$  or less, when the concentration exceeds approximately  $0.24 \text{ mg}$ , visibility is reduced to  $5 \text{ km}$  or less, and when the concentration exceeds approximately  $1.2 \text{ mg}$ , visibility is reduced to  $1 \text{ km}$  or less. However, these values differ according to the composition of the particles and meteorological conditions.

### (3) Visibility degradation and atmospheric pollutants

Air pollutants that affect visibility degradation differ greatly depending on both the season and the characteristics of their sources in the particular region in question. Visibility is also very closely connected to meteorological conditions. When ground inversion occurs and the winds are weak, such as during the early morning of winter, the air pollutant concentration rises due to a reduction in atmospheric diffusion capability, and visibility consequently worsens. During the day, there are many occasions when visibility is restored alongside the eradication of the inversion layer. On the other hand, in the case of photochemical air pollution during the summer, visibility degrades from noon onwards and throughout the afternoon due to generation of secondary generation particulate matter alongside the escalating reaction. This polluted air mass is carried inland by the local sea-land breezes, and the phenomenon that regions where visibility is poor shift in response to the movement of the air mass

is often observed. According to results measured in Los Angeles, when a degradation in visibility can be seen during the summer, a striking increase in particulate matter with a diameter of 0.1 to 1.0  $\mu$  m can clearly be seen when compared with days on which there is no visibility degradation.

Seinfeld (1986) collated the results of various measurements taken in America that relate to visibility, and has expressed the following opinions in their regard.

1. Visibility degradation of 60 to 95% is caused by the scattering of light due to particles.
2. Sulfate is always the matter most responsible for the scattering of light. The next most important are organic carbon particles. Nitrate, is important in some regions.
3. Light absorption due to particles of soot are the cause of a visibility degradation of 5.40%.
4. Except for smoke emissions from thermal power stations, carbon dioxide does not cause very much visibility degradation.
5. Of all the airborne particulate matter, the ability of soot particles in units of cubic content to degrade visibility is three times that of sulfate, nitrate, and organic carbon particles.

#### 5.4.3 Measuring visibility

Visibility measurement is generally conducted by eye. Specifically, one decides in advance on a marker such as a building or a mountain for which the distance from the measuring site is known in each direction, and this is then viewed by eye and the visibility is measured. When setting the marker, it is hoped for that the background is as whitish a sky as possible, and the marker itself is not white in color, and is of an appropriate size. The distance at which these markers can be barely differentiated is the level of visibility. Visibility measurement at night is difficult, but light that has not been condensed and whose intensity is known at a site whose distance is also already known is taken to be the index. Further, the outline of a mountain or the like against the background of the night sky can also be used as a marker. Depending on the direction of the measurement from the same terrestrial point, values such as maximum visibility, minimum visibility, mean visibility, and prevailing visibility will differ. The value of the maximum horizontal visibility from the measuring point for a total range of 180 degrees or more from all the horizontal directions is called the prevailing visibility. At this time, the ranges do not necessarily need to be contiguous. Specifically, the angles for each segment are totaled in sequence from the segment with high visibility, and when the total angle exceeds 180 degrees, the visibility value for the last segment is applied as the prevailing visibility. Given above is the method used for measuring by sight, but in airports, where visibility is an especially important safety factor, a transmissometer is used. This is a device that projects light towards the horizon from a height of 3 m above ground level, and then collects the light at a remote distance of approximately 150 m, by means of which, it can determine the visibility between both points. At present, devices are being constructed that can also measure within ranges of from 10 m to 100 km.

#### 5.4.4 Effects of visibility degradation

Visibility degradation not only worsens the quality of the living environment through the deterioration of the

scenery, but also reduces the number of tourists in tourist locations because the beautiful scenery has been damaged, thereby inflicting harm upon the local economy. Further, when visibility is degraded, because no visual flights can either take off or land, greater economic damage is incurred. This has become a major problem particularly in the U.S, where there are many privately-owned airliners. Airline flight rules are comprised of both the IFR (Instrument Flight Rule) and the VFR (Visual Flight Rule). Generally, if ground visibility is 5 km or better, and the clouds are a minimum of 300 m high, this is known as VMC (VFR Meteorological Conditions), and the VFR is considered possible. However, any other circumstances are known as IMC (IFR Meteorological Conditions), and the VFR is not permitted. In regions where visibility is poor, special instruments and special proof from the pilot are required for the VFR to be applicable, and this cannot help but add to the social costs. In addition, when visibility is extremely poor, the number of traffic accidents also increases. It is said that on land, when visibility is less than approximately 100 m, both traffic accidents and airplane accidents occur very easily, and when visibility at sea is less than approximately 1 km, ships very easily meet with disaster.

## 5.5 Effects on senses (sense of smell)

### 5.5.1 Sense of smell

Humans use their so-called five senses of sight, hearing, smell, touch, and taste as a means of acquiring information from the outside world. Of these five, smell works to identify the nature of odors and, along with taste, may be called a chemical sense. Further, smell possesses functions that only respond to a limited number of chemical substances (those substances with odors). Humans mainly rely on their senses of sight and hearing to live, for which reason, although sight and hearing are well developed, smell is, by comparison, a somewhat primitive sense. Odors such as that of rotting food and of burned substances aim to provide advance warnings of impending danger. On the other hand, on the basis of helping us to lead our individual lifestyles, such fragrances as can be seen in perfume, cosmetics, flavors, and so forth are part of a sense essential for us to lead fulfilled lives, and if we were to consider life without the sense of smell, we immediately begin to realize just how important a sense it is.

Airborne odors enter the nasal cavity along with inhaled air, and arrive at the olfactory membrane in the roof of the cavity by passing along the nasal airway, where they dissolve into the mucous membrane. The olfactory membrane contains olfactory glands (Bowman's gland) centered on olfactory cells, which are sense receptor cells. The olfactory glands hairs extend from the length of the olfactory cells through the mucous, and the tip of the cells (the olfactory small vesicle) also protrudes into the mucous membrane. Odor molecules that enter the mucous collide with both the olfactory glands hairs and the olfactory small vesicle, and excite the cell membrane, thereby transmitting an impulse (an electrical signal) to the olfactory cells. The olfactory nerve, which protrudes from the cells, enters the olfactory tube, which is the primary axis of the direct sense of smell. Moreover, a stimulus is transmitted to the cerebral cortex, where the nature of the odor is identified <sup>1)</sup> (see Fig.5.5.1).

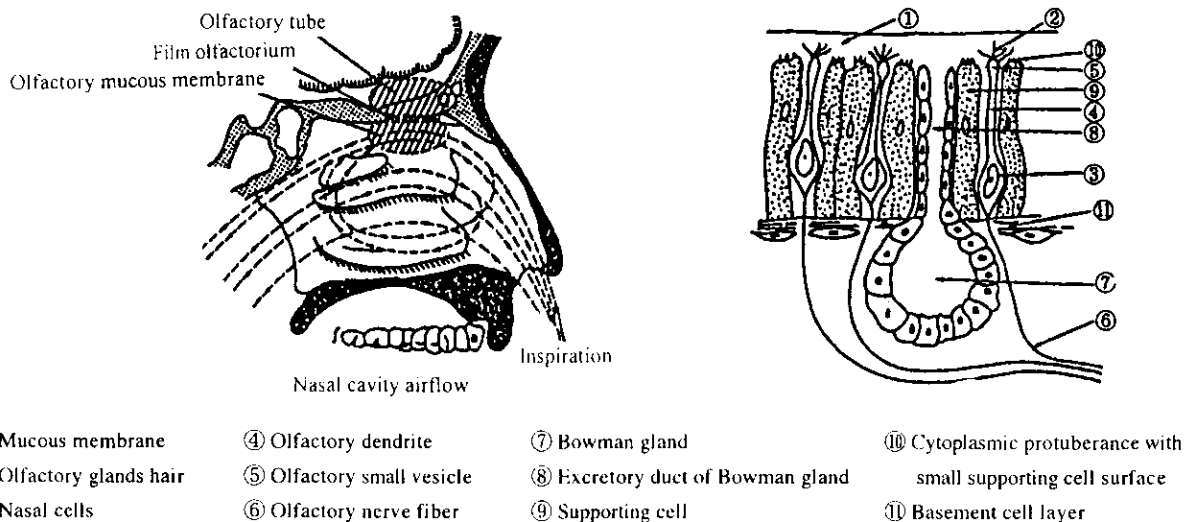


Fig.5.5.1 Micro structure of nasal cavity airway and olfactory mucous membrane <sup>1)</sup>

## 5.5.2 Features of the sense of smell

The features of the sense of smell can be cited as being an acute sense. The human sense of smell is considerably inferior when compared with that of a dog, but such detection limit (threshold value) indicates an excellent degree of sensitivity to a multitude of odorous substances, which far exceed the detection limit of modern analytical equipment (see Table 5.5.1<sup>2)</sup>). The difference between the concentration of an odorous substances and the strength of olfactory sense stimulation is known as the Weber-Fechner's law (Weber was a German psycho-physicist, and Fechner was a German psychologist), and is shown in Eq. (1). It shows that the strength of the sense is logarithmically proportional to the strength of the stimulus (concentration of the odorous substances.)

$$R = k \log S \quad (R: \text{strength of the sense, } k: \text{constant, } S: \text{strength of the stimulus}) \quad \dots\dots\dots (1)$$

In other words, even if the strength of the stimulus were multiplied tenfold, the weight of human senses is revealed to be only doubled in sensitivity. For specifically offensive odorous substances, which are regulated in Japan by the Offensive Odor Control Law, a relationship is reported between the concentration of the substances (strength of stimulus) and six-points odor intensity scale (strength of the sense) (see Table 5.5.2).

When the same odor is smelled for several minutes, the strength of its sense depreciates remarkably, until at last no odor can be detected. The stronger an olfactory stimulus grows, or else the longer that it lasts, the sense of smell becomes extremely fatigued, and the extent of this again depends on the quality of the odor. With such substances as acrolein, the fatigue of the sense of smell is great and, conversely, with such materials as methyl mercaptan, trimethylamine, and isovaleric acid, the sense of smell is not easily fatigued.

Just as with colors, there are color blindness and color weakness, so with odors, both odor blindness and odor weakness can be seen. The phenomenon whereby, while there is a normal sense of smell with regard to other odors, there is no sense with regard to one specific odor, or else the degree of sensitivity is extremely low, is called specific anosmia. For example, 18.2% of Caucasian males, and 4.5% of Caucasian females, cannot sense the odor from hydrogen cyanide.

The function of the sense of smell can be influenced by a variety of illnesses and diseases. The most obvious ones to affect the sense of smell are diseases of the nose such as chronic sinusitis (maxillary empyema), allergic rhinitis, and nasal polyps, and in addition, the sense of smell may also be affected by an external head injury, the side effects of drugs, viral infection, and so forth. A complete loss of the sense of smell is called ageusia, and a reduced capability when compared with the norm is called hyposmia. Such impairments of the sense of smell are caused by damage to the olfactory organs.

Table 5.5.1 Threshold sensitivity values for odorous substances <sup>2)</sup>

(Number of materials: 223) Units: ppm

No	Material	Threshold	No	Material	Threshold	No	Material	Threshold	No	Material	Threshold
Sulfur compounds			Ketones			39	n-propyl isovalerate	0.000056	16	2,4-dimethyl pentane	0.94
1	sulfur dioxide	0.87	1	Acetone	42	40	n-butyl isovalerate	0.012	17	n-octane	1.7
2	carbonyl sulfide	0.055	2	methyl ethyl ketone	0.44	41	isobutyl isovalerate	0.0052	18	isooctane (2MHep)	0.11
3	hydrogen sulfide	0.00041	3	methyl n-propyl ketone	0.028	43	ethyl acrylate	0.00026	19	3-methyl heptane	1.5
4	methyl sulfide	0.0030	4	methyl isopropyl ketone	0.50	44	n-butyl Acrylate	0.00055	20	4-methyl heptane	1.7
5	methyl allyl sulfid	0.00014	5	methyl n-butyl ketone	0.024	45	isobutyl Acrylate	0.00090	21	2,2,4-trimethyl heptane	0.67
6	ethyl sulfide	0.000033	6	methyl isobutyl ketone	0.17	46	metacrylate	0.21	22	n-nonane	2.2
7	acrylic sulfide	0.00022	7	methyl sec-butyl ketone	0.024	Phenol cresol			23	2,2,5-trimethyl hexane	0.90
8	carbon dioxide	0.21	8	methyl tert-butyl ketone	0.043	1	phenol	0.0058	24	n-decane	0.87
9	methyl disulfide	0.0022	9	methyl n-amyl ketone	0.0068	2	o-cresol	0.00028	25	n-undecane	0.62
10	ethyl disulfide	0.0020	10	methyl isoamyl ketone	0.0021	3	m-cresol	0.00010	26	n-dodecane	0.11
11	allyl disulfide	0.00022	11	diacetyl	0.000050	4	p-cresol	0.000054	Unsaturated normal hydrocarbons		
12	methyl mercaptane	0.000070	Fatty acids			Other oxides			1	propylene	13
13	ethyl mercaptane	0.0000087	1	acetic acid	0.0060	1	2-ethoxyethanol	0.58	2	1-butene	0.36
14	n-propyl mercaptane	0.000013	2	propionic acid	0.0057	2	2-n-butoxyethanol	0.043	3	isobutene	10
15	isopropyl mercaptane	0.0000060	3	n-butyric acid	0.00019	3	1-butoxy 2-propanol	0.16	4	1-pentane	0.10
16	n-butyl mercaptane	0.0000028	4	isobutyric acid	0.0015	4	2-ethoxyethyl acetate	0.049	5	1-hexane	0.14
17	isobutyl mercaptane	0.0000068	5	n-valeric acid	0.000037	5	dioximine	0.0000065	6	1-heptane	0.37
18	sec-butyl mercaptane	0.000030	6	isovaleric acid	0.000078	6	Ozone	0.0032	7	1-octene	0.0010
19	tert butyl mercaptan	0.000030	7	n-caproic acid	0.00060	7	uran	9.9	8	1-nonene	0.00054
20	n-amyl mercaptane	0.00000078	8	isocaproic acid	0.00040	8	2,5-dihydrofuran	0.093	9	1,3-butadiene	0.23
21	isoamyl mercaptane	0.00000077	Esters			Amines			10	isoprene	0.048
22	n-hexyl mercaptane	0.000015	1	methyl formate	1.30	1	Methylamine	0.035	Aromatic hydrocarbons		
23	thiophene	0.00056	2	ethyl formate	2.7	2	ethylamine	0.046	1	benzene	2.7
24	tetrahydrothiophene	0.00062	3	propyl formate	0.96	3	n-propylamine	0.061	2	toluene	0.33
Alcohols			4	isopropyl formate	0.29	4	isopropylamine	0.025	3	styrene	0.035
1	methyl alcohol	33	5	n-butyl formate	0.087	5	n-butylamine	0.17	4	ethyl benzene	0.17
2	ethyl alcohol	0.52	6	isobutyl formate	0.49	6	isobutylamine	0.0015	5	o-xylene	0.38
3	n-propyl alcohol	0.094	7	methyl acetate	1.7	7	sec-butylamine	0.17	6	m-xylene	0.041
4	isopropyl alcohol	26	8	ethyl acetate	0.87	8	tert-butylamine	0.17	7	p-xylene	0.058
5	n-butyl alcohol	0.038	9	n-propyl acetate	0.24	9	dimethylamine	0.033	8	n-propyl benzene	0.0038
6	isobutyl alcohol	0.011	10	isopropyl acetate	0.16	10	diethylamine	0.048	9	isopropyl benzene	0.0084
7	sec-butyl alcohol	0.22	11	n-butyl acetate	0.016	11	trimethylamine	0.000032	10	1,2,4-trimethyl benzene	0.12
8	tert-butyl alcohol	4.5	12	isobutyl acetate	0.0080	12	triethylamine	0.0054	11	1,3,5-trimethyl benzene	0.17
9	n-amyl alcohol	0.10	13	sec-butyl acetate	0.0024	Other nitrogen oxides			12	o-ethyl toluene	0.074
10	isoamyl alcohol	0.0017	14	ter-butyl acetate	0.071	13	nitrogen dioxide	0.12	13	m-ethyl toluene	0.018
11	sec-amyl alcohol	0.29	15	n-hexyl acetate	0.0018	14	ammonia	1.5	14	p-ethyl toluene	0.0083
12	tert-amyl alcohol	0.088	16	methyl propionate	0.098	15	acetonitrile	13	15	o-diethyl benzene	0.0094
13	n-hexyl alcohol	0.0060	17	ethyl propionate	0.0070	16	acrylonitrile	8.8	16	m-diethyl benzene	0.070
14	n-heptyl alcohol	0.0048	18	n-propyl propionate	0.058	17	meta-acrylonitrile	3.0	17	p-diethyl benzene	0.00039
15	n-octyl alcohol	0.0027	19	isopropyl propionate	0.0041	18	pyridine	0.063	18	n-butyl benzene	0.0085
16	isooctyl alcohol	0.0093	20	n-butyl propionate	0.036	19	Indole	0.00030	19	1,2,3,4-tetramethyl benzene	0.011
17	n-nonyl alcohol	0.00090	21	isobutyl propionate	0.020	20	skatole	0.0000056	20	1,2,3,4-tetrahydronaphthalene	0.0093
18	n-decyl alcohol	0.00077	22	methyl n-butylate	0.0071	21	ethyl-o-totuidine	0.026	Saturated normal hydrocarbons		
Aldehydes			23	methyl isobutylate	0.0019	Saturated normal hydrocarbons			Monoterpene		
1	formaldehyde	0.50	24	ethyl n-butylate	0.000040	1	propane	1500	1	α-pinene	0.018
2	acetaldehyde	0.0015	25	ethyl isobutylate	0.000022	2	n-butane	1200	2	β-pinene	0.033
3	propionaldehyde	0.0010	26	n-propyl n-butylate	0.011	3	n-pentane	1.4	3	limonene	0.038
4	n-butylaldehyde	0.00067	27	isopropyl n-butylate	0.0062	4	isopentane	1.3	Alicyclic hydrocarbons		
5	isobutylaldehyde	0.00035	28	n-propyl isobutylate	0.0020	5	n-hexane	1.5	1	methylcyclopentane	1.7
6	n-valeraldehyde	0.00041	29	isopropyl isobutylate	0.035	6	isohexane (2-MPen)	7.0	2	cyclohexane	2.5
7	isovaleraldehyde	0.00010	30	n-butyl n-butylate	0.0048	7	3-methylpentane	8.9	3	methyl cyclohexane	0.15
8	n-Hexylaldehyde	0.00028	31	isobutyl n-butylate	0.0016	8	2,2-dimethyl butane	20	Chlorine and chlorides		
9	n-heptylaldehyde	0.00018	32	n-butyl isobutylate	0.022	9	2,3-dimethyl butane	0.42	1	chlorine	0.049
10	n-octylaldehyde	0.000010	33	isobutyl isobutylate	0.075	10	n-heptane	0.67	2	dichloromethane	160
11	n-nonylaldehyde	0.00034	34	methyl n-valerate	0.0022	11	isooheptane (2-MHex)	0.42	3	chloroform	3.8
12	n-decylaldehyde	0.00040	35	methyl isovalerate	0.0022	12	3-methyl hexane	0.84	4	trichloroethylene	3.9
13	acrolein	0.0036	36	methyl n-valerate	0.00011	13	3-ethyl pentane	0.37	5	carbon tetrachloride	4.6
14	metaacrolein	0.0085	37	ethyl isovalerate	0.000013	14	2,2 dimethyl pentane	38	6	tetrachloroethylene	0.77
15	crotonaldehyde	0.023	38	n-propyl n-valerate	0.0033	15	2,3 dimethyl pentane	4.5			



Table 5.5.2 Relation between concentration of offensive odorous substances and strength of odor

Odorous substance	1	2	2.5	3	3.5	4	5
ammonia	0.1	0.6	1	2	5	10	40
methylmercaptane	0.0001	0.0007	0.002	0.004	0.01	0.03	0.2
hydrogen sulfide	0.0005	0.006	0.02	0.06	0.2	0.7	8
methyl sulfide	0.0001	0.002	0.01	0.05	0.2	0.8	2
methyl disulfide	0.0003	0.003	0.009	0.03	0.1	0.3	3
trimethyl amine	0.0001	0.001	0.005	0.02	0.07	0.2	3
styrene	0.03	0.2	0.4	0.8	2	4	20
propionic acid	0.002	0.01	0.03	0.07	0.2	0.4	2
n-butyric acid	0.00007	0.0004	0.001	0.002	0.006	0.02	0.09
n-valeric acid	0.0001	0.0005	0.0009	0.002	0.004	0.008	0.04
isovaleric acid	0.00005	0.0004	0.001	0.004	0.01	0.03	0.3
tolene	0.9	5	10	30	60	$1 \times 10^2$	$7 \times 10^2$
xylene	0.1	0.5	1	2	5	10	50
ethyl acetate	0.3	1	3	7	20	40	$2 \times 10^2$
methyl isobutyl ketone	0.2	0.7	1	3	6	10	50
isobutanol	0.01	0.2	0.9	4	20	70	$1 \times 10^3$
acetaldehyde	0.002	0.01	0.05	0.1	0.5	1	10
propionaldehyde	0.002	0.02	0.05	0.1	0.5	1	10
n-butylaldehyde	0.0003	0.003	0.009	0.03	0.08	0.3	2
isobutylaldehyde	0.0009	0.008	0.02	0.07	0.2	0.6	5
n-valeraldehyde	0.0007	0.004	0.009	0.02	0.05	0.1	0.6
isovaleraldehyde	0.0002	0.001	0.003	0.006	0.01	0.03	0.2

### 5.5.3 Effects on the physiological functions of odors <sup>1) 3)</sup>

The mutual relationship between the sense of smell and psychology is considerably large, and varies greatly from person to person. Many animals besides humans also react to take evasive action to avoid offensive odors they experience. This is an instinctive danger warning system, and is a means of avoiding danger. As a defensive posture against danger, humans have developed autonomic sympathetic nerve functions subconsciously. Although we experience considerable alarm when encountering offensive odors, including stimulant odors, when we continue to receive unpleasant odors, a state similar to the following is created. Namely, we feel deeply disturbed when we sense offensive odors. This sense of unpleasantness leads subconsciously to a tension in the sympathetic nerves. This state of tension in the sympathetic nerves has a variety of effects on various organs throughout the body, such as an increased pulse rate, and a rise in blood pressure. If the stimulus due to the offensive odor lasts only a short period of time, the state of tension in the sympathetic nerves returns to normal along with the disappearance of the stimulus, but if the duration of the stimulus is a long one, or else of the duration is short but reoccurs frequently, the state of tension in the sympathetic nerves is sustained, and ataxia of the autonomic nerves results.

When a pleasant odor is scented, the respiration deepens, but when an even stronger odor is scented, the

respiration reacts by stopping. When the concentration of an odor gradually grows from weak to stronger, the breath count initially increases, but if the concentration rises even further, it is then detected as if it were an offensive odor, and both the breath count and the depth of respiration are reduced. These changes in respiration are due to the use of information pivotal in respiration. When a pleasant odor is detected and the respiration deepens, there are simultaneously changes in the blood pressure, and we feel relaxed. It has also been reported that in addition to both apple and spicy floral odors, nutmeg, mace, neroli, and Valerian root are useful in reducing stress. Further, in animal experiments, with stimulant odors such as ammonia and chloroform, although blood pressure drops initially with a weak dose, a phenomenon whereby it subsequently rises can be seen.

In the case of eating food, appetite increases for food which gives off good odors, there is no appetite for foods with bad odors, and on occasion, we feel nauseous and also vomit. In animal experiments, feed with pleasing smells increase the quantity of feed consumed remarkably. That is to say, the quantity of feed consumed is increased due to a heightening in digestive capabilities through the adding of odors to the foodstuff.

In addition, an effect can also be seen on the reproductive system of animals. The mating season for animals is greatly influenced by odors. Other effects of odors are thought to be effects on sleep and on psychology. It has been reported that unpleasant odors can cause prevention of sleep.

#### 5.5.4 Effects of odors and their uses

Since olden times, people have pacified the spirit through the scent of blooming flowers, soothed their emotions with herbal baths, and have experienced improvements in their mood through odors. Humans have used odors since prehistoric times, and in ancient Egypt, scented materials were being used in the creation of mummies and, in addition, the mixing of medicinal scents is recorded in the Old Testament. Further, the traditional incense-smelling games of "kaori awase" (fragrance identification) and "kado" (fragrance arrangement) were devised in Japan, and also, the customs of impregnating clothing with incense, and burning incense within a room when greeting a guest arose.

At present, fragrances are used in a wide variety of fields such as foodstuffs, cosmetics, animal feed, household goods and, besides these, industrial applications, where their functions are used as anti-bacterials, antioxidants, air fresheners and for masking unpleasant smells, as well as in both physiological and psychological uses. Further, recently, such products as scented handkerchiefs, scented neckties, and other perfumed goods have gone on sale, besides which, the tendency to perfume one's living environment has manifested itself, and involvement in both domestic air fresheners and air conditioning systems has also appeared. A concept known as aromachology is behind this introduction of scents into the living environment.

Experiments to evaluate the usefulness the various physiological and psychological effects possessed by fragrances in both daily living and the treatment of disease have long been conducted in Europe, and the French comparative pathologist Gattefosse has suggested aromatherapy using refined oils that contain both scented medicines and herbs. This is not the traditional treatment methods of taking medicine internally, ointments, injections, or so forth, but a psychological treatment through scents using treatment by inhalation and direct atomization into the nasal cavity. Not only a pharmaceutical reaction incited by olfactory stimuli, but also

psychological effects such as feelings of pleasantness, unpleasantness, excitement, calm, and so forth, are expected, leading to the recognition that it is an effective treatment for both mental and physical ailments.

On the other hand, the previously-cited word “aromachology” is employed when using odors to stimulate not physiological, but psychological changes, employing odors and scents in the daily lives of ordinary, healthy people, and in research aiming to create a more pleasant lifestyle <sup>4)</sup>.

### (5.1) Literature cited

- 1) London County Council; Report of the County Medical Officer of Health and School Medical Officer for the year 1952, 157, The County Hall, Westminster Bridge, S.E.1., (1953)
- 2) Yoshida, Katsumi; Air pollution in Yokkaichi City and "Yokkaichi-asthma", *Science of Labor*, 19, 15 (1964)
- 3) Yoshida, Katsumi; Pollution in Yokkaichi City-concerning Yokkaichi Decision, *Science of Labor*, 28, 4-7 (1972)
- 4) J. Schwartz; Air pollution and hospital admissions for respiratory disease. *Epidemiology*, 7, 20-28 (1996)
- 5) D.L.V. Hammer et al.; Los Angeles student nurse study, Daily Symptom reporting and photochemical oxidants, *Arch. Environ. Health* 28, 255-260 (1974)
- 6) Makino, K. et al.; A study on the threshold of acute effects caused by photochemical oxidants. Proceeding of 4th Inter. Clean Air Congress, 27-32 (1977)
- 7) M.D. Lebowitz, et al.; The effects of air pollution and weather on lung function in exercising children and adolescents. *Am. Rev. Respir. Dis.* 109, 262-273 (1974)
- 8) Central Council for Control of Environmental Pollution Expert Committee; Report from the Expert Committee on environmental quality standards concerning nitrogen oxides, *Journal of Japan Society of Air Pollution*, 7, 151-155 (1972)
- 9) Environment Agency's Air Quality Bureau; Report on results of continuous observation and research on effects of air pollution upon health (1991)

### (5.2) References

- 1) Yamazoe, F.; Environmental Pollution and Agriculture, Types, Affect, Examination, Countermeasures. Tokyo, Hakuyusha (1975)
- 2) Furukawa, A., et al.; Interspecific difference in resistance to sulfur dioxide. *Res. Rep. Natl. Inst. Environ. Stud.*, No. 11, 113-126 (1980)
- 3) Bennet, J.H., and Hill, A.C. "Acute inhibition of apparent photosynthesis by phytotoxic air pollutants." *Am. Chem. Soc. symposium Ser. 3, Air Pollution Effects on Plant Growth.*, 115-127 (1974)
- 4) Fujiwara, T.; Appearance of visible damage in plants exposed to low concentrations of sulfur dioxide and research related to diagnosis. *Denchuken Noden Ken Report*, research report 74401 (1974)
- 5) Totsuka, T.; "Effects of sulfur dioxide on plant growth." *Natl. Inst. Environ. Stud.*, No. 10, 317-332 (1979)
- 6) Ashenden, T.W., "The effects of long-term exposures to SO<sub>2</sub> and NO<sub>2</sub> pollution on the growth of *dactyls glomerata* L. and *Poa pratensis* L." *Environ. Pollut* 18, 249-258 (1979)
- 7) Taylor, O.C., and Eaton, F.A., "suppression of plant growth by nitrogen dioxide" *Plant Physiol.* 41 (1), 132-135 (1966)
- 8) Plant Environment Research Institute, "Research on measurements of photochemical oxidant damage of crop plants exposed to air pollutants." 1980 Research Report, *Denchuken. Environment Research Institute of Biol.*, 1-8 (1981)
- 9) Thompson, C.R., and Kats, G., "Effects of ambient concentrations of peroxyacetyl nitrate on navel orange trees." *Environ. Sci. Technol.* 9, 35-38 (1975)

- 10) Brewer, R.F., et al. "The effects of hydrogen fluoride gas on seven citrus varieties." *Proc. Am. Soc. Hort. Sci.* 75, 236-243 (1960)
- 11) Pack, M.R., "Effects of HF on production and organic reserves of bean seed." *Environ. Sci. Tech.* 5, 1128-1132 (1971)
- 12) MacLean, D.C., et al. "Accumulation of fluoride by forage crops." *Contrib. Boyce Thompson Inst.* 24, 165-166 (1969)

#### (5.3) Literature cited

- 1) T.E Graedel, and R.d McGill; Degradation of materials in the atmosphere. *EnvironSci. Technol.*, 20, 1093-1100 (1986)
- 2) Horikawa, Kazuo, Shuichiro Takiguchi, Hidetsugu Okubo, Yoshio Ishizu, Motoke Kanezashi.; Study Relating to Atmospheric Corrosion on Metallic Materials and Rustproofing (No. 5), *Corrosion Engineering*, 16, 153-158 (1967)
- 3) T. E Graedel.; Capper patinas formed in the atmosphere -1. Introduction, *Corrosion Science*, 27, 639-657 (1987)
- 4) Sotogawa, Yasuhito; Regarding the International Cooperative Exposure Test for a Classification System of Atmospheric Corrosives (ISOCORRAG), *Rust Prevention Management*, 37, 55-66 (1993)
- 5) Hisamatsu, Yoshihisa; Rusted Steel's Atmospheric Corrosion, *Corrosion Engineering*, 20, 207-212 (1971)

#### (5.4) References

- 1) John H. Seinfeld; Atmospheric Chemistry and Physics of Air Pollution, John Wiley & Sons, ISBN 0-471-82857-2 (1986)
- 2) Chalson, R. J. et al.; The direct measurement of atmospheric light scattering coefficient for studies of visibility and pollution. *Atmospheric Environment*, 1, 469-478 (1967)

#### (5.5) Literature cited

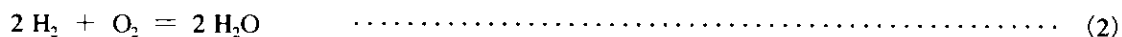
- 1) Offensive odor law research society; Offensive odor control law new revised handbook, Gyosei (1996)
- 2) Nagata, Yoshio, Takeuchi, Oshifumi; Results of threshold value measurements of odorous substances using a three-point comparative odor bag method, Japan Environmental Sanitation Center report, 17, 77-89 (1990)
- 3) Takaki, Sadayuki, Shibutani, Tatsuaki; The Chemistry of Smell, Asakura Shoten (1996)
- 4) Kawasaki, Michiaki; Home and Odor (2), *Odor research*, 25, 227-233 (1994)

## Chapter 6 Overview of Sources of the Air Pollutants

### 6.1 Stationary Combustion Facilities

#### 6.1.1 Introduction

The combustion is the rapid oxidation reaction accompanied with light and heat. The combustibles of which the heat generated could be utilized economically is called fuel. It is classified into solid fuels such as coal and coke, liquid fuels such as heavy oil and light oil, and gas fuels such as natural gas and oil gas. The stationary combustion facilities are the major source of air pollutants besides automobiles. When 12 kg of carbon burns,  $22.4 \text{ m}_N^3$  of carbon dioxide will be produced as shown in Eq. (1), and 4 kg of hydrogen will generate  $2 \times 22.4 \text{ m}_N^3$  of water vapor as shown in Eq. (2). In this way, there are theoretical relationships between the amount of element composing fuels and the amount of air required for its combustion or the amount of exhaust gas produced from combustion. The amount of air theoretically required for the complete combustion is called the theoretical amount of air, and that of exhaust gas the theoretical amount of combustion gas.



As an example, the theoretical amount of air and the theoretical amount of combustion gas will be calculated when 1 kg of heavy oil composed of 87% carbon, 12% hydrogen and 1% sulfur is burnt. Oxygen required to burn 12 kg of carbon is  $22.4 \text{ m}_N^3$ , and the same volume of exhaust gas is produced by this reaction. Therefore, the volume of oxygen required to burn 0.87 kg of carbon contained in 1 kg of heavy oil is  $22.4 \times 0.87/12 = 1.624 \text{ m}_N^3$  and the same volume of the exhaust gas, that is carbon dioxide, will be produced. Also, 0.12 kg of hydrogen is contained in the heavy oil and the volume of oxygen required for its combustion is  $22.4 \times 0.12/4 = 0.672 \text{ m}_N^3$  and twice that volume,  $1.344 \text{ m}_N^3$ , of water vapor is produced. As the molecular weight of sulfur is 32, 32 kg of sulfur will react with  $22.4 \text{ m}_N^3$  of oxygen, and the same volume of sulfur dioxide is generated. Accordingly, the volume of oxygen required to burn 0.01 kg of sulfur contained in the heavy oil is  $22.4 \times 0.01/32 = 0.007 \text{ m}_N^3$  and the same volume of sulfur dioxide will be produced. Thus, the volume of oxygen required for the combustion of heavy oil becomes to  $1.624 + 0.672 + 0.007 = 2.303 \text{ m}_N^3$ . If we convert it to the amount of air, the theoretical amount of air of  $2.303 \times 1/0.21 = 10.97 = 11 \text{ m}_N^3$  is obtained by assuming the content of oxygen in the air as 21%. Also, in addition to the volume of reaction product gases, that is  $1.624 + 1.344 + 0.007 = 2.975 \text{ m}_N^3$ , the volume of nitrogen,  $2.303 \times 0.79/0.21 = 8.66 \text{ m}_N^3$ , which will not participate in the combustion reaction, should be added to obtain the volume of the theoretical burnt gas, and it is calculated as  $2.975 + 8.66 = 11.635 \text{ m}_N^3$ . This is the wet theoretical amount of combustion gas containing water vapor, and the volume of dry theoretical amount of combustion gas is obtained by subtracting the amount of water vapor from the last value as  $11.635 - 1.344 = 10.291 \text{ m}_N^3$ . Generally, the combustion requires a larger amount of air than the theoretical amount of air, and the ratio between actually used air

and the theoretical air amount of is called the air fuel ratio. The combustion will be more ideal as this ratio is as near as 1. Incidentally, the theoretical amount of air is, for instance, about  $10-13 \text{ m}_N^3/\text{kg}$  for fuel oil, and the combustion of 1 dl of fuel oil requires about  $1-1.3 \text{ m}_N^3$  of air.

The calorific value of fuel is different for each fuel, and it is 4,500-8,000 Kcal/Kg for coal and 11,000-11,500 Kcal/Kg for kerosene or gasoline. In order to realize combustion, there must be a combustion field where fuels and air are supplied and mixed with each other and the combustion temperature is maintained. However, the combustion would not proceed with those conditions alone. It will stop unless the heat of combustion and the burnt gas are removed from the combustion field. Thus the combustion furnace has been used, and it utilizes heat and releases exhaust gas through stacks to the atmosphere to cause air pollution problems.

### 6.1.2 Formation of Major Air Pollutants

#### (1) Sulfur Oxides <sup>2)</sup>

Sulfur existing in crude oil as organic polymer compounds of sulfur is largely transferred to heavy oil during the oil refinery process and desulfurized by hydrogenation technique. However, some sulfur will remain in heavy oil and oxidized to sulfur dioxide in the flame of combustion facilities. A part of it will oxide to anhydrous sulfuric acid, and both enter into the exhaust gas and will be released in the atmosphere. Also, sulfur exists in coal as sulfides of iron, organic sulfur compounds, or gypsum, and when they burn, the former two will be transformed to sulfur oxides and some will remain in the coal ash.

#### (2) Soot and Dust <sup>2)</sup>

Inorganic compounds are contained in the coal and transformed to ash by combustion, and a part of it will enter in the burnt gas. Particularly, fine flyash generated by the combustion of pulverized coal has been attracting much attention. Small amount of inorganic compounds is also existing in crude oil and transformed to ash when burned. Also, soot and dust will be formed in the process of combustion which causes black smoke which has been attracting people's attention. The smoke includes the soot, which is formed when the combustible gas component generated by the combustion process burns in gas phase, and the carbon remained cenosphere, which is a carbonaceous material formed by condensation polymerization reaction of thermally decomposed fuel droplets in the furnace. The soot is spherical in shape with diameter of  $0.02-0.05 \mu \text{ m}$  and very fine, and the cenosphere is also almost spherical in shape with  $10-300 \mu \text{ m}$  diameter, but has bumpy surfaces. The chemical composition of soot and dust is generally considered to be carbonaceous material, and actually it is an aggregate of hydrocarbon of mostly around  $\text{C}_{40}\text{H}_{16}$ , and contains many condensed multi-cyclhydro carbons. The mechanism of soot and dust formation is very complex, and they are considered to be formed through processes such as the dehydration of hydrocarbons, thermal decomposition, polymerization, unsaturated bonds, and formation of aromatic rings in case of gas combustion. In the case of liquid spray combustion, tar is formed in the oil droplet by thermal decomposition. Then high order hydrocarbons are formed which finally forms the carbon remained dust by releasing gases.

### (3) Carbon Monoxide

Carbon monoxide is generated by incomplete combustion of fuels, and could be possibly released from industries such as gas work, steel mill and carbonyl compounds manufacturing.

### (4) Nitrogen compounds <sup>2)</sup>

Air is a gas mixture of one volume of oxygen and four volumes of nitrogen. In the combustion reaction, those two elements react mainly in the flame to form nitrogen oxides which are attracting attention as the air pollutant. Also, nitrogen containing compounds such as aniline and quinoline are existing in the fuel, and become as a cause of the generation of nitrogen oxides. Incidentally, the formation rate of nitrogen oxides is faster for the latter reaction (fuel NO<sub>x</sub>) than for the former reaction (thermal NO<sub>x</sub>).

Furthermore, nitrous oxide (N<sub>2</sub>O) which, like fluorocarbon, will destroy ozone layer in the stratosphere is generated from facilities such as fluidized bed coal boilers and sludge incinerator where fuels with high nitrogen content is burned at low temperature.

### (5) Dioxins

The formation mechanism of dioxins in the combustion process is complex, and is not understood fully. The release of dioxins from incinerator of domestic and industrial wastes is becoming a big problem.

### (6) Other Pollutants

The kind of air pollutants and their volume formed in the combustion process are different with different fuels, facilities, or combustion equipment. In addition to pollutants described above, various air pollutants including heavy metals and multi-cyclic aromatic hydrocarbons can be released from the combustion furnaces such as coal or incinerator and glass furnace, which introduce industrial materials into the field of combustion.

## 6.1.3 Thermal Power Station <sup>3)</sup>

### (1) Overview

The thermal power station is the important facility to support our civilization, but is also causing air pollution. The technology of thermal power generation includes the steam power station which makes electricity by rotating the electrical generator via steam turbines that are driven by high pressure steam generated by the boiler, the electric generation which uses internal combustion engines, and that which combines the former two technologies. Fig.6.1.1 shows an example of the system of steam power station, which is composed of internal combustion engine, combustion system, steam system, water supply system, and electrical system <sup>3)</sup>. The sources of air pollutant are the former two systems which use coal, oil and natural gas as a fuel.



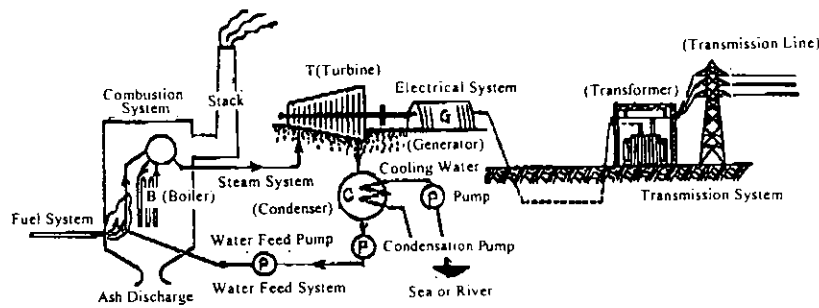


Fig.6.1.1 An Example System of Steam Power Station

## (2) Steam Power Station

### Combustion Equipment and Formation Mechanisms of Air Pollutants

The stoker-type combustion equipment is used which delivers coal to the furnace core by moving grates and burns it there. In the stoker combustion, the coal is heated and its combustible components are evaporated and ignited to heat it further, and the coal burns as it gets softer. Therefore, the surface in contact with air is small and much soot is generated, and also the combustion efficiency is low. As a result, the pulverized coal combustion is devised to make the contact area of fuel and air larger and to increase the combustion efficiency by enhancing their mixing. In this method, 70-80% of the coal is crushed to a fine powder of about 200 mesh pass, and then transferred by air to the burner. It is applied in the recent power stations which burn only coal. Incidentally, the type of pulverized coal burner includes the injection burner which injects the mixture of pulverized coal and air in the center and introduces the secondary air in parallel around it, and the swirl burner which mixes and burns the pulverized coal, mixed by central rotation blades with primary air, with secondary air introduced around it. In the pulverized coal combustion facility, a part of ash enters into an ash collector as slag. Inorganic materials contained in coal are melted and evaporated in the flame, and then are solidified when they exit from the flame as the temperature drops. In this process, they are transformed into very fine flyash as surface areas tend to be maximized and uniform pressure is exerted. The major component of flyash is silica, and the particle size of this fine glassy powder is 0.1-100  $\mu\text{m}$  with an average diameter of about 15  $\mu\text{m}$ . Its content in the exhaust gas is about 15  $\text{g}/\text{m}_N^3$ . Presently, more than about 99% of flyash is recovered by the electrostatic precipitator. In the combustion of heavy oil, it is warmed to the temperature of 85-105°C from about 35°C in the storage tank to reduce viscosity, so that the spraying power required for delivering it to the burner could be reduced. The burner is designed to completely mix air and fuel, which makes it into a spray of heavy oil allowing a larger area of fuel and air to come into contact and to obtain complete combustion. The type of heavy oil burner includes; the pressure injected burner, in which either a spray is formed by pressurizing the heavy oil and injecting it through small holes to give high speed swirling motion, or the heavy oil is injected and burned as swirling motion is applied; the steam spray burner in which heavy oil is sprayed by a jet of steam; the air injection burner in which compressed air, instead of steam, is used for injecting heavy oil through a spiral passage to make a rotating spray; and the injection rotation burner in which a thin layer of heavy oil is made by injecting it into a cylindrical cup rotated at high speed and fine particles of heavy oil are made. Among those, the pressure injected burner and the steam injection burner are commonly used in large capacity boilers. The type of thermal power station includes the heavy oil burning station that burns only heavy oil, the station that burns

kerosene, naphtha, etc. to reduce air pollution, the one that burns the mixture of heavy oil and pulverized coal, and the one that burns either heavy oil or pulverized oil or COM (Coal Oil Mixture), which is a suspension of pulverized coal, mixed at equal caloric value, in the heavy oil. Also, the station that burns imported natural gas is increasing since the technology of sea transportation of liquefied natural gas by tanker has been developed. The number of natural gas thermal power station is increasing because the amount of air pollutants contained in its burnt gas is smaller. Natural gas is burned in the gas burner, and its combustion efficiency is high because its mixing with air is better.

### Boiler <sup>3)</sup>

The boiler is a facility to generate high pressure steam for driving the turbine, and is an important facility in the steam power station along with the generator and the turbine. Fig.6.1.2 is a schematic of the boiler, and it has an overhead drum that contains water delivered from the water supply system. Many water tubes, drawn from this drum, are arranged on the front and rear surface and a part of the side surface of the fire furnace. These tubes absorb thermal radiation generated by the combustion. The feed water to the boiler is heated by the combustion burnt gas in the economizer, and enters into the boiler drum. From here, it flows down through the descending water tube to absorb heat, and returns to the drum, as its temperature rises, through the water tubes in a state of mixed steam and saturated water. Then, the water is removed by a steam separator in the drum, and only steam is delivered to the turbine after heated by a super heater.

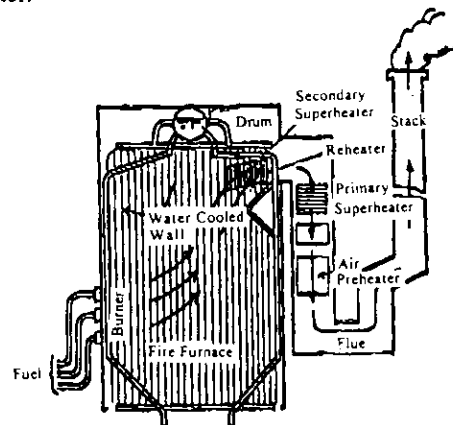


Fig.6.1.2 Schematic Diagram of Boiler

### Steam Turbine <sup>3)</sup>

The steam turbine corresponds to a hydraulic turbine in the hydraulic power plant. High pressure, high temperature steam generated in the boiler is introduced in the turbine blade to obtain power by its high speed rotation. Depending on the working mechanism, it is classified into an impulse turbine and a reaction turbine. Generally, the former is suitable for high pressure steam and the latter for low pressure steam, and therefore they are used in combination with each other. Furthermore, when classified by the use of steam, it is divided into a condensation turbine, a back pressure turbine, a extraction turbine, etc. In the condensation turbine, the steam discharged through turbine is entirely introduced into a condenser to reduce the pressure to near vacuum state and to make the pressure head large so that the heat of steam could be used efficiently only in the turbine system. In the back pressure turbine, the discharged steam is used for process steam without reducing the pressure so much. In the extraction turbine, the

steam for factory process is taken out from the middle of the steam path.

#### Generation Status of Air Pollutants <sup>4)</sup>

The concentration of air pollutants exhaust from the stack of steam power station is dependent on the type of fuel, and the amount discharged depends on the amount of fuel consumed or power generated.

Most of the sulfur oxides (more than 95%) is released in the atmosphere as sulfur dioxide, and its amount can be calculated from the amount of fuel used and the content of sulfur in the fuel. Further, its concentration in the exhaust gas can be calculated from the composition of fuel and the condition of combustion, or air fuel ratio (excess air ratio). Also, in the case of pulverized coal combustion, the concentration of solid particles, such as soot and dust, corresponds to that of flyash, and is about  $15 \text{ g/m}_N^3$  as mentioned previously, although it is quite variant. In the heavy oil combustion, the solid particle generated is mostly soot, and there are data that its concentration is 0.02-0.038  $\text{g/m}_N^3$  for the combustion of Class C heavy oil <sup>4)</sup>.

Accordingly, in the case of mixed combustion of coal and heavy oil, the concentration of solid particle increases roughly in proportion to the mixture ratio of coal. In the case of natural gas combustion, solid particles are scarcely found in the exhaust gas. As for the nitrogen oxides, they are generated and discharged mostly as a form of nitrogen monoxide, but some nitrogen dioxide is also discharged. Therefore, it is a common practice to obtain their combined amount and to convert it into the amount of nitrogen dioxide, with expressing the amount in mass and the concentration in volumetric ratio (ppm).

Considering the formation mechanism of those pollutants, the discharged amount is dependent on the fuel and facility. Particularly, more fuel NO<sub>x</sub>, which originates from organic nitrogen compounds contained in the fuel, is generated in the coal combustion. Furthermore, the concentration of thermal NO<sub>x</sub>, which is formed by the oxidation of nitrogen contained in the air, increases as the flame temperature increases. Accordingly, the concentration of thermal NO<sub>x</sub> increases in the steam power station in which the air for combustion is heated by the air heater. As a result, the concentration of nitrogen oxides tends to be higher in the coal combustion, and then in the oil combustion, while the lowest value is found in the gas combustion. For example, there are data that, in the steam power station, its concentration is 545 ppm for the coal combustion when the oxygen in exhaust gas is converted to 0%, similarly 122-372 ppm for the oil combustion, and 173-672 ppm for the Class C heavy oil combustion <sup>4)</sup>.

#### (3) Other Thermal Power Station

Other types of thermal power station now in operation include, the station operated by diesel engine or gas turbine and the one that operated by the combined system which drives the generator by a gas turbine and a steam turbine, which uses the exhaust of the former as the boiler's heat source, placed in tandem. Particularly, the combined power system is getting much attention as its thermal efficiency reaches to more than 60% even if the energy is used mostly for electric power generation.

Also, the MHD power generation, which generates electricity by flowing high temperature combustion gas plasma through magnetic field, is promising as a large scale direct power generation technology of high thermal efficiency, and its development is attracting much attention.

## 6.2 Source of Dust

### 6.2.1 Introduction

The pulverization or crushing which fragments a solid material by mechanical force and makes fine particles is the oldest mechanical operation of humankind. By this operation, the handling of solid becomes easier and the rate of reaction and dissolution and the catalyst reactivity are improved because the surface area of solid is increased<sup>1)</sup>. These effects are readily seen in such processes as coal pulverization in the pulverized coal combustion facility and ore processing in the puddling. Also, the polishing is an operation to smooth out the surface of solid materials<sup>2)</sup>. The particulate material that is generated and scattered during smashing or other mechanical operations, or associated with the accumulation of the materials obtained through these processes, is called the dust and is attracting attention as a source of air pollution.

The major source of dust includes the pulverizing equipment, polishing machine, sieve, particulate transport facility, pile of ore or soil, and cokes oven. Generally, the size of dust generated by the mechanical operation such as pulverization or crashing is relatively large and is about a few  $\mu$  m and its range is rather broad, unlike the fine fume generated by the chemical reaction.

### 6.2.2 Pulverization or Crushing Process

#### (1) Pulverization or crushing Process

There are various types of pulverization process including the mining of materials of cement, limestone, coal, etc., crushing of clinker in the cement factory, production of pulverized coal in the pulverized coal burning power station, production of materials for glass, material preparation for china and porcelain, production of aggregate to use construction of building, waste treatment, and so on. Also, the type of pulverization or crushing equipment is diverse.

#### (2) Pulverization or Crushing Equipment<sup>2) 3)</sup>

The pulverization or crushing equipment is categorized into three types according to the type of force that the machine exerts on the solid to crush it finely. That is, the equipment that uses pressure by sandwiching the material to be crushed between two surfaces, the one that uses impact by hitting the material with a hammer so that it is crushed instantly by a high speed collision of a hard body, and the one that uses shear force exerted perpendicular to the direction of sear force<sup>2) 3)</sup>.

(Rough Crusher)<sup>2) 3)</sup>; The rough crusher is appropriate for pulverization or crushing the solids of 1,500-100 mm to a size of 500-25 mm, and includes the jaw crusher, gyratory crusher, crushing roll, impact crusher and disintegrator<sup>2)</sup>. The jaw crusher is a machine which crushes solid materials by a strong pressure applied between a fixed jaw and a reciprocating movable jaw as if human masticates foods by mouth, and is also called the masticating crusher. The gyratory crusher is also called the revolving crusher, and crushes the material by rotating and gyrating a conical rotator eccentrically set within an inverted conical ring to constantly change the distance between them and supplying the

material to be pulverized or crushed from above <sup>3)</sup>.

(Medium Crusher) <sup>2)3)</sup>; The medium crusher is used to pulverize or crush solid materials of 500-6 mm to a size of 50-1 mm, and includes the cone crusher, roll mill, hammer mill and disk mill. The hammer mill is an impulse crusher which pulverizes or crushes the material by hitting it with a high speed hammer.

(Fine Crusher) <sup>2)3)</sup>; The fine crusher pulverizes or crushes solid materials of 12.5-1 mm to a size of 2-0.1 mm, and includes the conical ball mill, tube mill, rod mill, roller mill, stamp mill, edge runner, vibration mill, and impact mill.

(Super-Fine Crusher) <sup>2)3)</sup>; The super-fine crusher is used to pulverize or crush solid materials of 6-0.1 mm to a size of 0.1-0.001 mm, and includes the hydrodynamic energy mill, colloid mill, and friction disk mill.

(Shearing Machine) <sup>3)</sup>; The shearing machine includes the knife cutter, rotary cutter, and screw cutter.

### 6.2.3 Polishing Process

Polishing is an operation that finishes the surface of material by using the polishing machine such as the belt sander, drum sander and wide belt sander. The belt sander polishes the surface of material by moving the polishing paper or cloth with 2-4 pulleys. The drum sander polishes the material by rotating a cylinder, wrapped around with the polishing paper or cloth, at high speed.

The wide belt sander is a machine to polish the material by upper and lower drums wrapped with an endless polishing paper. These machines are used in the factories for paper and pulp manufacturing, furniture manufacturing, and plywood manufacturing, and produce dusts <sup>2)</sup>.

### 6.2.4 Sieving of Particles

The sieve discriminates between smaller particles, which pass through small holes, and larger particles, which do not. It is used to classify the dust that corresponds to a mesh of the sieve by moving the dust on it. The mesh and thickness of the sieve are specified by the Japanese Industrial Standard (JIS). If the dust is classified by stacking a sieve with a larger mesh on another sieve with a smaller mesh, its particle size distribution can be obtained.

For industrial purposes, the rotation sieve which has a sieve of cylindrical wall and the flat sieve which uses a moving flat sieve are used. The dust is generated at the particle feed portion, on the mesh of a sieve, and at the particle discharge portion. The protection measures such as a cover and a dust collecting equipment are installed.

### 6.2.5 Transportation of Particles <sup>2)</sup>

For the transportation of particles, belt conveyers and bucket conveyers are used, and they also scatter dusts. The belt conveyer is a transportation facility which has an endless belt stretched between two pulleys on both ends of a frame and transfers loads on the belt by moving it continuously. This facility is quite economical for transporting loose materials such as coal, ore, or gravel, but sometimes accompanied by dust scattering. Also, the bucket conveyer is a facility to transport loose materials from the lower to higher positions vertically or at a steep

angle. Buckets are attached at a constant distance to a chain or a belt stretched upward, and loose materials thrown into the bucket are continuously transported upward. When it is used for transporting dusts, they are scattered around. The protection cover is installed or the dust is collected by a dust collecting equipment while arranging an enclosure hood.

#### 6.2.6 Pile of Particles

Industrial raw materials such as coal and ore are often stored in the open air as a pile, but some are lost through the scattering of dust. For instance, it is reported that about 6.4 mg per 1 kg per year will be lost from the pile of coal. Also, about 13.2 pounds (1 pound=0.4536 kg) per acre of pile area (1 acre=4046.6m<sup>2</sup>) per day of rock and gravel is reported to be lost from the rock pile operated 24 hours a day<sup>2)</sup>. Measures such as the water sprinkling or the chemical sprinkling by a sprinkler are used.

## 6.3 Chemical Process

### 6.3.1 Introduction

The chemical industry is an essential factor for the prosperity of humankind, but it has been causing many air pollution problems because it deals with various chemical substances. As the kind of chemical industry is diverse, only some typical examples of incidence of the emission of air pollutant will be described below.

### 6.3.2 Cement Industry <sup>1) 2) 3)</sup>

Dust scattering from the cement industry has been often a subject of claims. Cement is a generic name of inorganic colloid containing plaster, lime, magnesia, volcanic ashes, etc. The currently so called cement is the Portland cement or the mixed cement, which is mainly composed of the former, and its manufacturing method is summarized as, "to thoroughly mix mainly lime material and magnesia material in proper proportion, then to obtain clinker by firing it until part of it will melt, and to pulverize or crush it while adding some gypsum." There are a dry process and a wet process in the manufacturing method of cement.

#### (1) Dry Process

In the dry process, lime stone and clay are dried by a rotary dryer, such as shown in Fig. 6.3.1, until their water content becomes less than 1%. Then, mixing them at an appropriate ratio considering their composition, the clinker is produced by firing them in the rotary kiln. The heat source of the rotary dryer is the combustion gas of coal or oil, and the raw material is continuously fed into one end of the cylindrical rotary furnace and the dried material is brought out from the other end. In this case, the exhaust gas of the rotary kiln is sometimes used as a heat source. Dried materials for cement is then continuously fired in the rotary kiln, shown in Fig. 6.3.2, which is made of a steel cylinder, lined with fire bricks and inclined by 3-5 degrees from the horizontal. That is, the materials fed from the upper end of the kiln will move downward in the furnace which is rotating at 1-2 rpm, and burned at the lower end at a temperature of about 1,450°C for 1-4 hours until they are partly melted. The cement clinker formed will be discharged from the lower end of the kiln at a temperature of about 1,100°C.

Heat is recovered by the boiler and suspension heater attached to the kiln. The rotary kiln, shown in Fig. 6.3.2, is attached with a suspension heater that exchanges heat by flowing the materials and burnt gas through a chain of several cyclones. The rotary kiln is the largest source of air pollutant in the cement industry, and generates air pollutants such as cement dust, sulfur oxides, nitrogen oxides and heavy metal fumes.

#### (2) Wet, Lepol Process

In the wet process, the product is manufactured by crushing and mixing the materials as a slurry of about 35% water content, without drying, with ball mill or compartment mill, and burning with the rotary kiln.

In the Lepol process, 10-14% of water is added to the material obtained from the mixing process of the dry process, and pellets of 2.5-5 cm in diameter are made by the pelleting machine. Then the pellet is dried and fired

by the exhaust gas of the rotary kiln to obtain clinker.

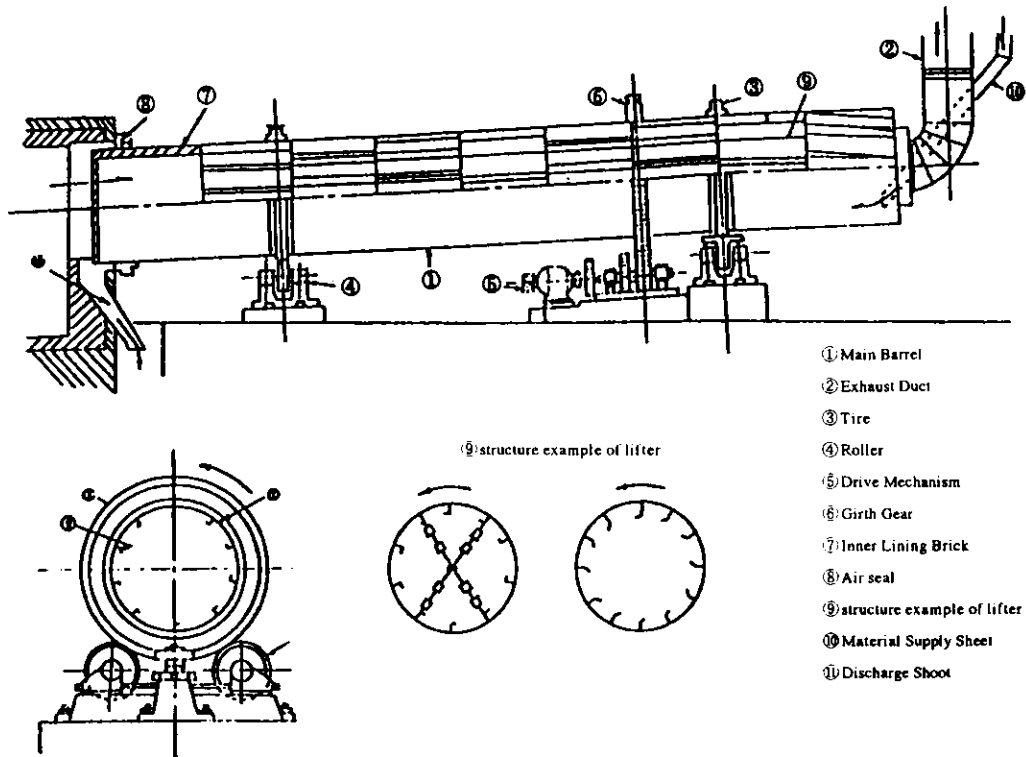


Fig.6.3.1 Rotary Dryer

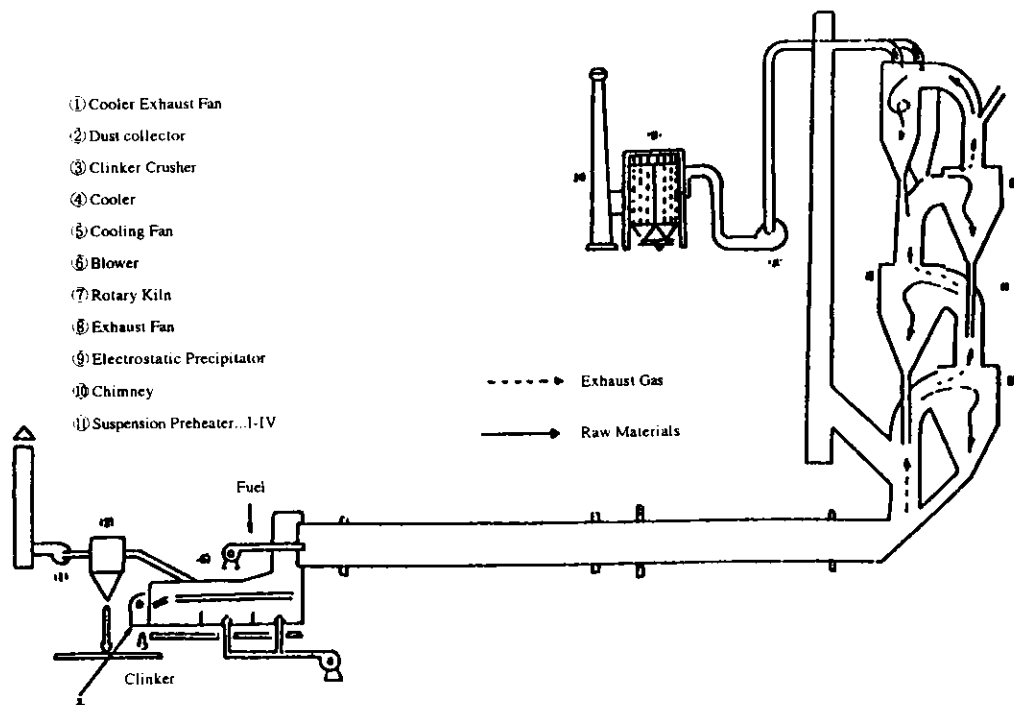


Fig.6.3.2 Rotary Kiln



### (3) Cement Manufacturing

The clinker obtained from the burning process using kiln is cooled to a room temperature by the air quenching cooler, and then cement is obtained by adding 4-6% of gypsum and crushing them with a ball mill or a tube mill. Here, the gypsum is added as a coagulation retarder for the cement. The cement industry is attracting attention, because it acts as a receiver of gypsum obtained by desulfurization plant for flue gas with excellent environmental stability, as described in Chapter 7, and also it uses scrapped tires as a fuel or flyash as a raw material.

### (4) Air Pollutants

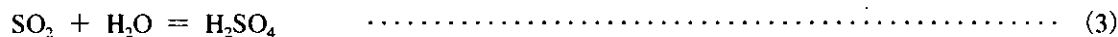
The major air pollutant in this industry is the dust generated from processes such as crushing and mixing, silo storage, transportation, burning process, and drying, and the sulfur oxides and nitrogen oxides are also generated from the rotary kiln and rotary dryer. Particularly, the concentration of nitrogen oxides in the exhaust gas of the rotary kiln is quite high. The pollutant, that is attracting much attention recently, is the particle, and it is presently handled by the electrostatic precipitator. The sulfur oxides due to fuel containing sulfur are absorbed by the raw materials for cement, and the ash enters into the cement if coal is used as a fuel. The concentration of dust from the rotary kiln in the dry method is 25-130 g/m<sup>3</sup> and the dust is collected by the electrostatic precipitator.

In the case of the wet process, it is reported that the dust of 84.5 g/m<sup>3</sup> discharge concentration has been processed by an electrostatic precipitator to achieve the collection efficiency of 97.8%. Also, there is data that, in the case of rotary dryer, the dust collection efficiency is 99.93% for the exhaust concentration of 42.9g/m<sup>3</sup><sup>1)</sup>. Also, data is reported that the concentration of nitrogen oxides in the exhaust gas of the rotary kiln is 158-1,469 ppm for the dry process and 503-1,927 ppm for the wet process, converting to the concentration for 0% oxygen<sup>1)</sup>.

### 6.3.3 Sulfuric Acid Industry<sup>4)5)</sup>

In the production of sulfuric acid, the contact process is adopted. In this process, the elemental sulfur is oxidized by air to sulfur dioxide, then the latter is oxidized to sulfur trioxide using vanadium pentoxide as a catalyst. Then it is made into contact with sulfuric acid to obtain highly concentrated sulfuric acid. Sulfur dioxides has been obtained by burning iron sulfide before, but is now obtained by the method using elemental sulfur which is a byproduct of desulfurization of the heavy oil.

The primary chemical reaction of this process proceeds as Eq.(1) to (3) below:



The major air pollutants produced are sulfur dioxide and acid mist, and the amount of sulfur dioxide emission is about 20-70 pounds (1 pound is equivalent to 0.4536 kg) per 1t of acid production, and that of acid mist is 0.3-

7.5 pounds per 1 t of acid production if there is no removal system. However, the latter is reduced to 0.02-0.2 pounds if the removal system is installed<sup>4)</sup>.

#### 6.3.4 Nitric Acid Industry <sup>4) 5)</sup>

In the production of nitric acid, a method using ammonia as a raw material is adopted. In this process, ammonia is oxidized by air in high temperature to form nitrogen monoxide, at the existence of platinum catalyst. Nitrogen dioxide, obtained by oxidizing the mixture of this nitrogen monoxide and air, is then absorbed into the water to form nitric acid. The primary chemical reactions of this process are shown in Eq.(4) and (5): <sup>5)</sup>



The major air pollutants emitted from this process are nitrogen monoxide and nitrogen dioxide exhausted from the absorption tower. The amount emitted is, converting it to nitrogen dioxide, about 57 pounds per 1t of 100% nitric acid production. Besides, it is reported that about 10 pounds of nitrogen dioxide equivalent per 1,000 pounds of strong acid production is discharged from the nitric acid concentrator.

The removal efficiency of a catalytic burner for processing the exhaust gas from the absorption tower is reported as 36-99.8% with average 80%, and that of an alkali scrubber is reported as 90% <sup>4)</sup>.

#### 6.3.5 Chlorine Production <sup>4) 5)</sup>

Chlorine is produced by the electrolysis of sodium chloride water solution, and it is formed at the positive electrode. Fig.6.3.3 shows a schematic of the diaphragm method. High temperature chlorine formed is cooled, and then washed by nitric acid to dry. It is then liquefied and shipped. Major cause of chlorine release is the discharge of gas that is not liquefied. When there is no protection system, the amount of chlorine emission is 2,000-16,000 pounds per 100 t of liquefied chlorine production for the mercury method (concentration of 20-50 ppm), and 2,000-10,000 pounds for the diaphragm method. Other than that, it is reported that 400 pounds of chlorine is discharged from the water absorption tower, 450 pounds from the vent hole of tank trucks, and 1,200 pounds from the vent hole of storage tank <sup>4) 5)</sup>.

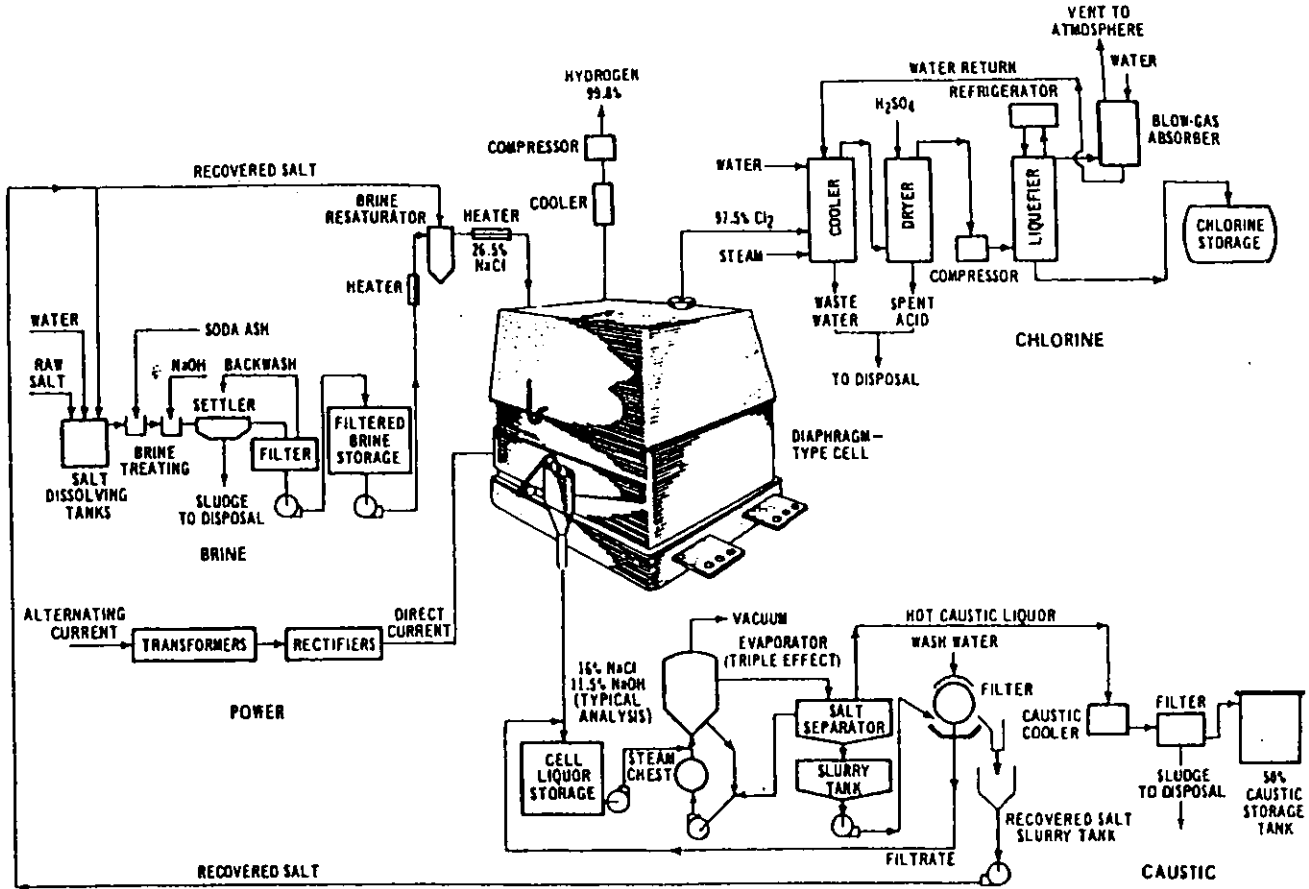


Fig 6.3.3 Process of Sodium Chloride Water Solution Electrolysis

## 6.4 Metal Refinery

### 6.4.1 Introduction

Generally, the iron buried as oxides is a very important metal for the life of humankind, and the scale of its refinery process is very large. Also, the copper buried mainly as sulfide is important, and the scale of its refinery is relatively large too. Both processes have been the source of air pollution. Fluorine is used in the refinery of aluminum and chlorine is used for titanium and silicone. These are also causing air pollution problems.

### 6.4.2 Integrated Steel Works

#### (1) Ore

The earth is a planet abundant in iron, and hematite ( $\text{Fe}_2\text{O}_3$ ), magnetite ( $\text{Fe}_3\text{O}_4$ ), etc. are used for the iron refining<sup>1)</sup>. Also, coal is required for the process, but Japan has little natural resources and is dependent on the import for those raw materials. Further, other materials such as limestone are also required, and the integrated steel work is usually provided with port facilities.

#### (2) Facility Outline<sup>2-4)</sup>

Large scale production of iron has been started since the invention of the blast furnace in the 15th century. At first, charcoal was used as reduction agent, and the lack of charcoal was a restriction for iron refining by the blast furnace, because forests were cut down. In 18th century, the desulfurization of coal by coaks oven became possible. So, today, a large scale integrated steel works with blast furnaces for iron refining using coal coke as reduction material which annual production capacity of, for example 12 million tons became possible to operate. And these integrated steel works consist of the material facilities and as the sintering furnace and coaks oven, the blast furnace and its associated facilities, the steel refining facility from pig iron such as converter or open-hearth furnace, the work furnace to make steel sheets or pipes, and the system for transportation and energy supply and control that connect with each other. Japan is the largest iron manufacturing country in the world with annual production capacity of 180 million tons of total iron and is refining 100 million tons of steel.

#### (3) Blast Furnace<sup>2-4)</sup>

The blast furnace is a symbol of the integrated steel works, and visitors of the plant will be deeply impressed by its sight rising high among other facilities. The blast furnace is, as shown in Fig.6.4.1, a tall cylindrical furnace, a little thicker at the lower part. It is consisted of a main body made by steel outer plate lined with brick its inside and a hot blast stove that sends hot air to the main furnace. A large blast furnace with a production capacity of 4,000 tons of pig iron per day is now in operation. Generally, the daily production rate of the blast furnace is about 0.8-1.0 ton per  $1 \text{ m}^3$  of inner volume of the furnace. The raw materials such as iron ore (sintered ore), coke, lime, fluorite, etc. are thrown in from the top of the blast furnace, and air, heated to about  $700^\circ\text{C}$  by a hot blast stove, is blown into the furnace from a lower tuyere to promote reduction reaction in the furnace. The blast furnace gas, mainly

composed of carbon monoxide, which is generated in the furnace and has a calorific value of about  $1,000 \text{ kcal/m}_N^3$  is used as a fuel for the hot blast stove.

A reduction environment is produced in the blast furnace, and there occurs such processes as the reduction of iron ore and melting of generated iron, the melting of limestone and fluorite and the dissolution of impurities such as sulfur and phosphorus contained in the melted iron into the melted limestone and fluorite. The separation of them occurs as heavy melted iron sinks to the bottom of the furnace and melted limestone rises over it. The temperature inside the furnace is about  $1,600^\circ\text{C}$ , and the situation resembles well with that of the age of first segregation of the earth when it was hot and the central core, mantle and crust were formed. The melted iron (pig iron) accumulated in the bottom of the furnace is taken out from a tap hole at the lower part of the furnace 5-7 times a day, and then the molten limestone (slag) floating over molten pig iron is also brought out from a slag hole alternately. Besides the exhaust gas from the hot blast stove, air pollutants discharged from the blast furnace include dust and fume formed when taking out the pig iron and slag and the leak from the furnace top. The concentration of nitrogen oxides in the exhaust gas of hot blast stove is 6.27-1.11 ppm converting to 0% oxygen when the blast furnace gas is used as a fuel, and is 34.8-97.7 ppm when the mixture of the blast oven gas and the coke furnace gas is used as a fuel <sup>2)</sup>.

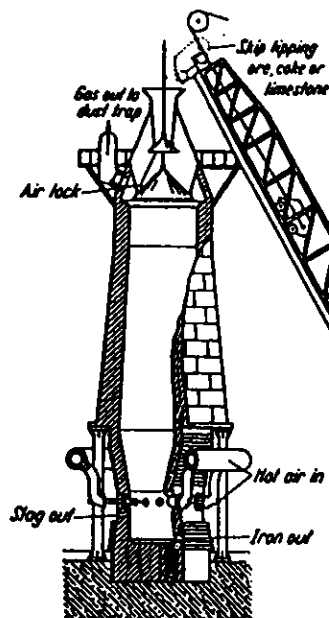


Fig.6.4.1 Blast Furnace

#### (4) Cokes Oven <sup>2)4)</sup>

The cokes oven is a facility to obtain coke by carbonization of coal, and is made of bricks. It is composed of a carbonization part and a combustion part. Coal is charged into the former and the coke furnace gas generated in it is burnt in the latter to make coke through heating and carbonization while shutting down the air.

About a hundred coke furnaces are usually constructed side by side, each of which the size of carbonization part is 400-450 mm wide, total length 8-6 m, total height 4-6 m and the amount of charged coal 8-27 tons. The temperature of combustion part is about  $950-1,300^\circ\text{C}$  and the coal is made into coke in 14-24 hours <sup>2)4)</sup>. The wet

quenching method has been used before in which coke is pushed out from each oven one by one and quenched by sprayed water, but the dry quenching method is recently used that quenches coke by taking it out into a steel vessel and quenching it with nitrogen gas under a tightly closed condition. The latter method has advantages because thermal energy can be recovered and the discharge of dust from steam exhaust does not occur. Besides dust produced at the wet quenching process, air pollutants from the combustion of cokes oven gas are generated in this method. Sulfur oxides is not produced in the combustion part of cokes oven gas as it is well refined, and also dust is not produced because the process is a gas phase combustion. However, the nitrogen oxides is a problem. There is data that the concentration of nitrogen oxides in the cokes oven exhaust gas is about 284-1,844 ppm when the coke oven gas is used as a fuel periodically in accordance with a cycle of taking out coke from the furnace.<sup>2)</sup>

#### (5) Sintering Furnace Machine <sup>2) 4)</sup>

The shape of iron ore for iron refining is not always uniform, and much powdery ore is also included. Therefore, the temperature distribution in the blast furnace becomes not uniform and the ventilation of the furnace is deteriorated if the raw iron ore is introduced in the furnace as it is. Furthermore, sulfur contained in the iron ore enters into the pig iron and also deteriorates its characteristics. Accordingly, the ore is sintered so that the raw materials are utilized efficiently by adjusting their particle size and improving the ventilation of the furnace, and sulfur is removed. In the sintering process, powder ore, sulfuric acid slag, iron sand, mill scale, blast furnace dust, scrap, etc. are mixed with limestone, converter slag, etc., and coke is also mixed in them as a fuel. The mixture is sent into one end of the sintering furnace, and transferred through it on the belt of the pallet that travels between two sprockets. It is quenched by air when the belt moves to the other end of the sintering furnace and the sintering process is completed<sup>2) 4)</sup>. The primary air pollutant emitted from the sintering process is oxides of sulfur, and about 71% of sulfur contained in the raw iron ore is discharged as sulfur oxides. This amount corresponds to a major part of the discharge of sulfur oxides from the integrated steel work, and its concentration is said to be about the same as that of the exhaust gas from heavy oil combustion<sup>4)</sup>. Also, there is data that the amount of nitrogen oxides discharged, when converted to NO<sub>2</sub>, is 76.2-369 kg per 1 ton of ore sintered<sup>2)</sup>.

#### (6) Converter, Heating Furnace, Etc. <sup>2) 4)</sup>

In the integrated steel work, there are many combustion facilities including the converter, open-hearth furnace, electric furnace, working furnace for steel, and boiler. The converter, shown in Fig. 6.4.2, is a pear-shaped furnace with an opening on the top, and is a typical facility in the integrated steel work along with the blast furnace.

As shown on the left of Fig. 6.4.2, it can be tilted to take out the melted steel and slag in its inside. In the basic oxygen process of steel manufacturing, which succeeded to improve the efficiency and is shown in Fig. 6.4.2 for the case of an upward oxygen blowing converter (LD converter), fine iron oxide fume of about 0.2  $\mu$  m in diameter is discharged, and the release of red cloud of high concentration, as high as 70-80 g/m<sup>3</sup>, rising high in the sky, has attracted much attention. However, this problem is almost solved by the dust collector at the present, and it is highly appraised as a great achievement of air pollution measures in the steel industry. In other cases of air pollution measures in the integrated steel work, the electrostatic precipitator and desulfurization/denitration equipment have been successfully applied. In Japan, high quality steel products can be refined quickly in the LD

converter, and the open-hearth furnace is not used now.

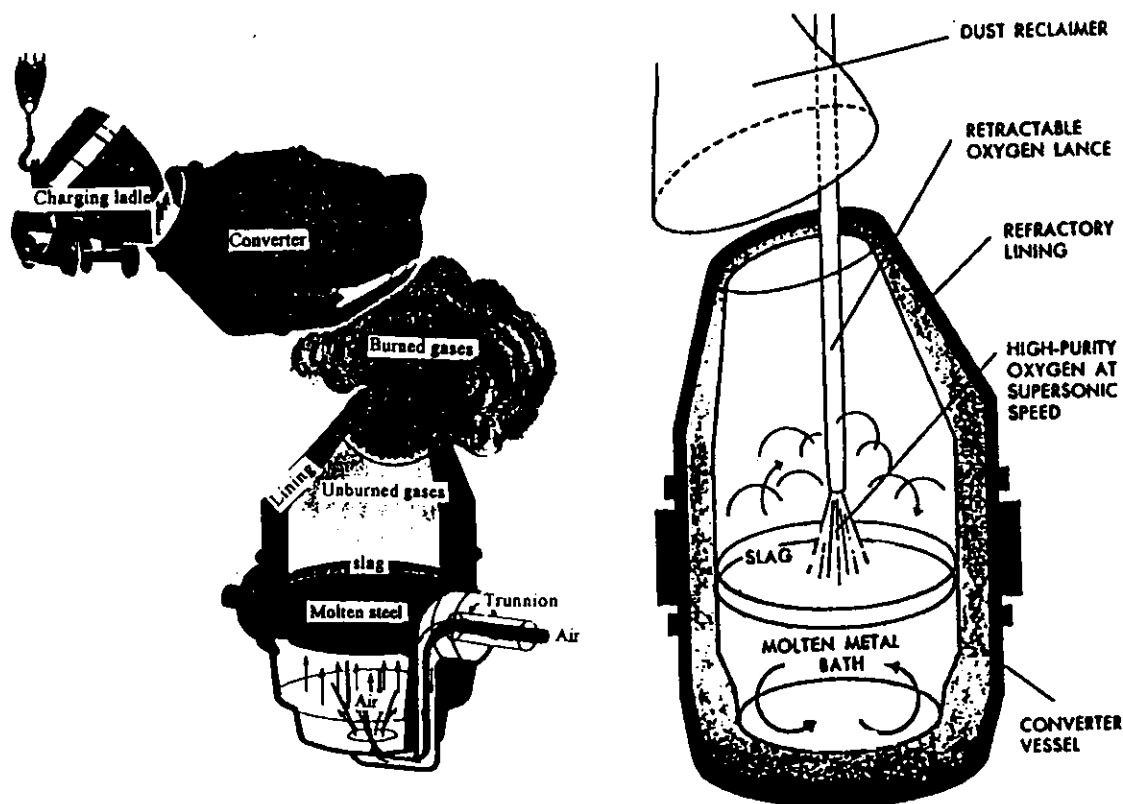


Fig. 6.4.2 Converter Furnace

### 6.4.3 Copper Refinery <sup>5)</sup>

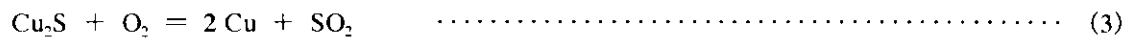
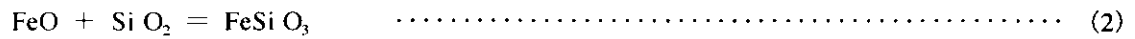
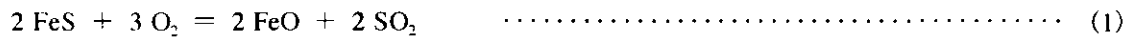
Copper is most abundantly mined as sulfide ore such as iron containing chalcopyrite ( $\text{CuFeS}_2$ ), chalcocite ( $\text{Cu}_2\text{S}$ ) and covellite ( $\text{CuS}$ ), although it is also mined as native copper ( $\text{Cu}$ ) or cuprite ( $\text{Cu}_2\text{O}$ ) <sup>1) 5)</sup>. As a content of copper in the copper ore is very small, the refined ore obtained by flotation process to separate useful ores from the mother ore is dried and roasted in the multi-stage roasting furnace to burn excess sulfur, arsenic, antimony, etc.. In this process, all of the copper and most of the sulfur combined with iron are retained in the ore and excess sulfur is discharged in the atmosphere as sulfur dioxide. Then, the roasted ore obtained from the roasting process is heated and melted in the reverberatory furnace or self-melting furnace at  $1,370\text{-}1,500^\circ\text{C}$  by burning coal or heavy oil. Impurities are removed as a slag and the copper content is increased.

Through this process, copper is converted to cuprous sulfide and iron to ferrous sulfide. Those are melted together to form a matte containing 38-45% copper. Then, this matte is changed into the converter along with silica and alumina.

In the first stage of the converter process, ferrous sulfide is oxidized by air blown into the converter, and sulfur dioxide is formed. This sulfur dioxide then combines with silica and alumina to form a slag, and pure cuprous sulfide is left in the bottom of the furnace.

The slag is removed by tilting the converter, and then refining is continued by raising it upright. Cuprous

sulfide is decomposed to remove sulfur as sulfur dioxide, and the melted copper in the furnace is taken out. Major chemical reactions in the copper refining process are shown in Eq. (1) to (3). Blister copper obtained in the converter is further refined by electrolytic refining to obtain pure copper <sup>5)</sup>.



Major air pollutants from the copper refinery process are sulfur dioxide and particles. In the exhaust gas of the self-melting furnace, 20-30 /m<sub>N</sub><sup>3</sup> of particles are contained and collected by the cyclone or electrostatic precipitator.

#### 6.4.4 Zinc Refinery

The major ore containing zinc is zinc blende (ZnS), so zinc is also mined as a sulfide ore like copper. In the refinery of zinc blende, the refined ore obtained through the concentration process is roasted to remove sulfur as sulfur dioxide, as shown in Fig.6.4.3. As sulfur dioxide is an air pollutant, it is used to manufacture sulfuric acid. However, if its concentration is small, it is converted to plaster by the exhaust gas desulfurization equipment. Zinc is then sent to the exuding/dissolution process and refined by electrolysis as Fig.6.4.3.



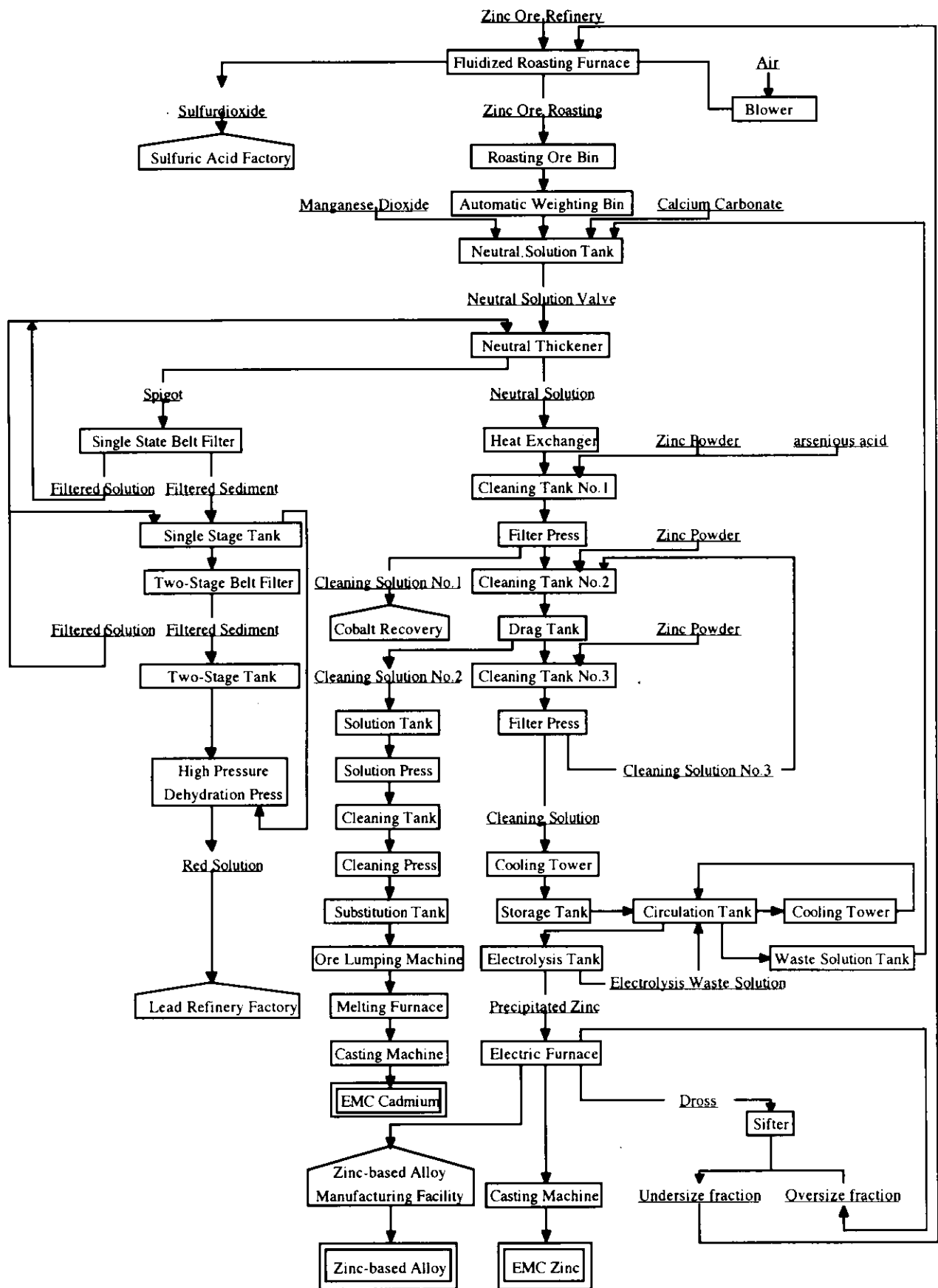


Fig.6.4.3 Zinc Refinery

#### 6.4.5 Aluminum Refinery <sup>6)</sup>

The major ore for aluminum production is bauxite ( $\{HAlO_2\}_2 \cdot H_2O = Al_2O_3 \cdot 2H_2O$ ) <sup>1)</sup>.

Bauxite is converted first into sodium aluminate by dissolution process with sodium hydroxide solution. Then, alumina ( $Al_2O_3$ ) is obtained by Bayer process, in which aluminum hydroxide obtained by hydrolysis of sodium aluminate is calcinated, and it is then refined by electrolysis under the bath of molten salt containing fluorine. This process had been widely used in Japan before, but is now almost abandoned since the energy crisis because the price of electricity increased. The refinery has been transferred to overseas, and the domestic work of aluminum industry is restricted to only working process now.

This refinery work has discharged hydrogen fluoride in the molten salt electrolytic process and caused air pollution problems. The electrolytic refinery is proceeded by melting alumina in the high temperature molten bath of cryolite ( $AlF_3 \cdot 3NaF$ ), which is a double fluoride of aluminum and sodium, and running the electricity between cathode (carbon) placed in the lower part of electrolytic furnace and carbon anode inserted into the upper part of the furnace. Here, the operating voltage is 4.7 volts and the temperature of electrolysis is about 950°C. The anode is oxidized and consumed by oxygen contained in alumina. Either the soderberg type anode which is forming an electrode by burning carbon paste or the Prebake type anode which uses an electrode burned in advance is used. The amount of air pollutants emission is less for the latter type. The specific weight of molten bath at melted condition is 2.08-1.96, and that of aluminum is 2.28-2.24. Thus, the aluminum generated is separated from the molten bath. Carbon monoxide, tar, alumina particles, and hydrogen fluoride are generated in this process, and, for example, it is reported that 40 mg/m<sup>3</sup> of hydrogen fluoride, 100 mg/m<sup>3</sup> of tar, 90 mg/m<sup>3</sup> of particles are contained in the exhaust gas of the electrolytic furnace and are controlled <sup>6)</sup>.

#### 6.4.6 Refinery of Titanium and Silicon

Titanium is a super heat resisting metal with an excellent anti-corrosion characteristics, and its alloy is used as a material for gas turbine blade. Also, silicon, germanium, and zirconium are quite useful as a material for semiconductors and special purpose materials. Those elements are easily combined with chlorine to form chloride, and the chlorides of these elements are refined by hydrogen reduction after removing impurities through distillation. Primary air pollutant emission by those refinery processes is chlorine.

## 6.5 Petroleum Refinery

### 6.5.1 Petroleum <sup>1) 2)</sup>

Petroleum is generally considered to be formed by the sedimentation of organic materials originated from ancient animals buried under the seabed, but there are many different theories. In 1890, a Russian N.V. Sokoloff proposed a theory of space origin for petroleum in which he insisted that hydrocarbons in the petroleum has been existing since the early stage of the space formation because trace methane is found in the meteorite. A volcanic theory has been also proposed because there are volcanic eruptions in the oil well regions and hydrocarbons are present in the volcanic gases. Furthermore, a theory of earth's depth origin was presented in which petroleum is thought to be made as carbon dioxide solved in the water deep underground of the earth reacts with alkaline metals to form acetylene and then petroleum. Although those inorganic theories of petroleum origin had some rationality in reasoning, they didn't correspond with the conditions of petroleum turn out, and the theory of organic origin attracted general supports. The theory of land plant origin, in which the difference in the formation of petroleum and coal was explained by the difference in heat and pressure subjected to buried plants, has little ground concerning to chemical proofs.

Further, the theory of coal origin was not supported because coal and oil were not found simultaneously. The theory of oil shale origin had been attracted some support, but it required an explanation of the origin of oil shale itself. Also, there are many supporters of the theory of sea weeds origin, in which sea weeds are transformed to petroleum by heat and pressure. The theory of animal origin insisted that fish, shellfish, and microorganism have been transformed to petroleum. The theory of fish origin proposes that petroleum was formed as ancient marine animals were buried underground and their protein was dissolved by decomposition and fermentation action and the fats remained formed petroleum. In 1880, Engler in Germany produced petroleum by dry distillation of fishes at 10 atm and 320°C. K. Kobayashi in Japan proved experimentally that petroleum could be easily formed under atmospheric pressure by mixing Japanese acid clay and fishes, and claimed that Japanese acid clay has some relationship with the formation of petroleum. Many theories and experimental results have been reported about the origin of petroleum, and the existence of organic nitrogen compounds in the petroleum is considered to be a strong basis for the animal origin theory and fish origin one is now widely supported.<sup>1)</sup> In this theory, the organic material sediments on the seabed is considered to be decomposed by anaerobic bacteria, releasing most of their oxygen, sulfur, and nitrogen. Hydrocarbons formed by decomposition is then considered to dissolve into water and condensed in the sedimentation rock. The theory explains that those were gradually accumulated in the space of rock dome and formed an oil field. As for the natural gas, it is considered that there are two kinds of gas, namely that formed simultaneously with petroleum in the rock dome and that formed from anaerobic decomposition of plants. The former is thought to be a main resource.

Petroleum is a mixture of hydrocarbons which could be classified by the molecular structure and number of carbons, and contains small amount of organic nitrogen and sulfur compounds. Hydrocarbons in petroleum include paraffins, expressed as a general chemical formula of  $C_{2n}H_{2n+2}$ , naphthenes  $C_nH_{2n-6} + 6H$ , olefins  $C_nH_{2n}$  and benzenes  $C_nH_{2n-6}$ . Also, the structure of various hydrocarbons is very complex as there are compounds with straight or

branched carbon chain, ring structure of five or six elements, or multiple union <sup>1)</sup>.

Hydrocarbons in the petroleum can be separated by distillation utilizing the difference of molecular weight.

Table 6.5.1 summarizes the composition of each petroleum distillate <sup>2) 3)</sup>.

Table 6.5.1 Composition and Boiling Point of Various Oil Distillates <sup>2) 3)</sup>

Distillate	Typical Hydrocarbon	Boiling Point (°C)
Natural Gas	CH <sub>4</sub>	-161
Liquefied Gas	C <sub>3</sub> H <sub>8</sub> , C <sub>4</sub> H <sub>10</sub>	-44 ~ +1
Petroleum Ether	C <sub>5</sub> H <sub>12</sub> , C <sub>6</sub> H <sub>14</sub>	30 ~ 60
Aviation Gasoline	C <sub>5</sub> ~ C <sub>9</sub>	32 ~ 150
Automotive Gasoline	C <sub>5</sub> ~ C <sub>12</sub>	32 ~ 210
Naphtha	C <sub>7</sub> ~ C <sub>12</sub>	100 ~ 200
Kerosene	C <sub>10</sub> ~ C <sub>16</sub>	177 ~ 290
Light Oil	C <sub>12</sub> ~ C <sub>18</sub>	205 ~ 316
Lubricating Oil	C <sub>15</sub> ~ C <sub>24</sub>	250 ~ 400
Heavy Oil		300 ~

## 6.5.2 Petroleum Refinery

### (1) Outline

Crude oil is distilled to separate each hydrocarbon component by the difference of boiling point into similar groups because of the convenience of usage. The distillation is a process to separate mixed liquid into its component by utilizing the difference of boiling point and volatility. Also, in the production process of fuels and lubricating oils, the conversion of some components, which have little value in the aspect of demand, into other products of higher value by chemical reaction, as well as a refinery process to remove impurities and hazardous components from the product, is performed to maintain the balance of demand and supply for different distillate <sup>4) 5)</sup>.

### (2) Oil Refinery

The structure of oil refinery is roughly divided into a refinery facility and an utility facility. Facilities for the normal pressure distillation (topping), normal/reduced pressure combination, reduced pressure distillation (vacuum flash, vacuum distillation), catalytic reforming, cracking, coking, visbreaking, hydrogenation refinery of kerosene, desulfurization of heavy oil, solvent dewaxing, solvent extraction, cleaning of naphtha, cleaning of kerosene, cleaning of lubrication oil, recovery of sulfur (desulfurization) belong to the former. The facilities for harbor, boiler and oil storage belong to the latter. The scale of oil factory is now very large, and a typical capacity of a single oil refinery is almost 300 thousand barrels (1 barrel is 159 liter) per operating day <sup>3)</sup>.

### (3) Oil Storage

As the oil refineries in Japan depend their supply of crude oil on the import, they have port facilities and store imported oil in the outdoor tanks. The storage tank is divided into a fixed roof tank and a floating roof tank. Incidentally, the salt component is removed from crude oil before the distillation process by the electric demineralization or chemical demineralization process <sup>4)</sup>.

#### (4) Distillation

The distillation is a basic operation of the oil refinery, and divided into the natural pressure distillation and reduced pressure distillation (vacuum distillation, vacuum flash). The natural pressure distillation is the first stage of oil refinery, and is called topping. Crude oil is heated by the heating furnace (pipe still), but maintains a liquid state without being vaporized because the pressure in pipe still is high. It is then blown into the flash zone of the fractionator (rectification column) which has a dozen stepped racks with many bubble-caps and holes, in 45-90 cm intervals. As its pressure is atmospheric, crude oil except for its heavy components is vaporized and the heavy components descends the tower and are taken out from the bottom. The vaporized oil ascending the fractionator is gradually condensed at each step rack, in the order of higher boiling point components, and flows down to lower step racks, because the temperature is lower as it goes upward inside the column. As the temperature of lower step rack is higher than that of upper step racks, light components are again transferred to upper racks. Inside the fractionator, the volatilization and liquefaction of oil are repeated and the same effect as re-distillation is realized. Various different distillates can be obtained by extracting the distilled liquid from the appropriate part of the rack and cooling it by the heat exchanger. For heavy oil, it is distilled at reduced pressure because it will be decomposed thermally if distilled in the normal pressure. When the pressure is reduced, the boiling point is also reduced and the distillation can be performed at relatively low temperature to prevent thermal decomposition. The principle of reduced pressure distillation is similar as that of natural pressure distillation, and a similar type of fractionator is used in which the pressure is reduced to 30-80 mm by a steam ejector. In this case, the fractionator becomes generally large in diameter and shows a short and thick appearance<sup>4)</sup>. The outline of crude oil distillation process is shown in Fig.6.5.1<sup>5)</sup>.

#### (5) Conversion

If the oil is sent directly to the market as distilled, the ratio of oil products is restricted by the composition of crude oil. Then, the supply could not always respond to the demand, and the demand and supply will not be balanced appropriately. Lack or excess of supply will occur for certain products. Therefore, it is desirable to convert some oil distillates (fraction) that could be excessive in supply into more useful products by chemical reaction. This process is called the oil conversion. The conversion had been started since when the demand for gasoline increased and the direct distillate gasoline (straight run gasoline) alone could not fill the demand. The oil conversion process includes, for instance, the thermal cracking that produce gasoline by pressurized decomposition of heavy oil at a temperature of about 500°C, catalytic reforming that produce high octane gasoline by catalytic decomposition of high boiling point distillates heavier than kerosene, hydrogenating decomposition (hydrofinishing) that converts unsaturated hydrocarbons to saturated hydrocarbons or removes sulfur and nitrogen compounds<sup>4)</sup>. The hydrogenating decomposition will be described more in detail later, because it is very important in the measures for air pollution.

#### (6) Treating

As impurities and hazardous materials are contained in the crude oil distillates, the substances such as compounds of sulfur, nitrogen and oxygen, resins, asphalt, and aromatic compounds should be removed from them. This process is called the treating, and includes such operations as cleaning by acid/alkali, absorption refining, solvent extraction, and hydrogenation refining<sup>4)</sup>.

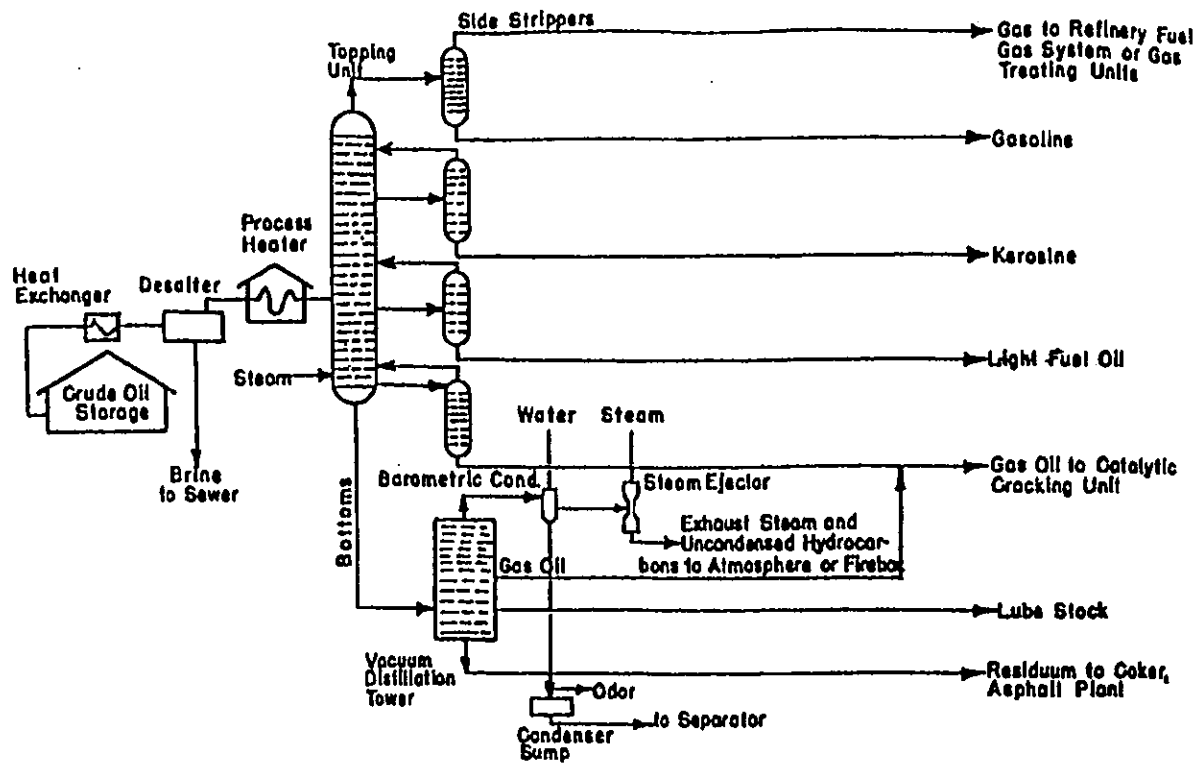


Fig.6.5.1 Outline of Crude Oil Distillation Process

(7) Petroleum Products and Shipping

Many kinds of petroleum product are existing including fuels such as gasoline, kerosene, light oil, jet fuel, heavy oil, and liquefied petroleum gas, lubricating oil, grease, and oil pitch. Fuels are shipped by tankers, tank train, and tank lorries, while lubricating oil and grease are shipped in cans <sup>4) 6)</sup>.

6.5.3 Desulfurization of Heavy Oil

(1) Hydrogenating Desulfurization

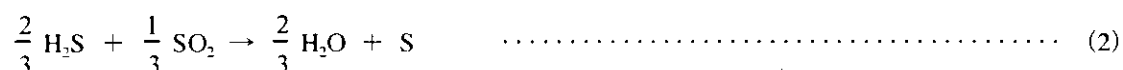
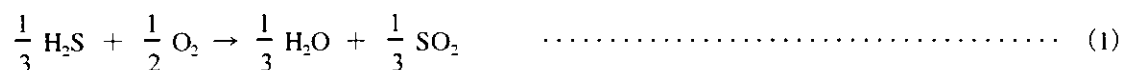
The hydrogenating desulfurization process performed to remove sulfur from heavy oil is very important in the oil refinery operation as a measure for the air pollution control. Compounds of sulfur based on thioalcohol (mercaptane), sulfide and thiophene are contained in the crude oil. Generally, the oil produced in the Middle East contains more sulfur than that produced in Southeast Asia. Also, distillates with higher boiling point contains more sulfur, and more than 80% of sulfur in the crude oil remains in the residual oil at natural pressure. Of course, sulfur remains in every class of distillate (fraction) and is desulfured, but the most important process for air pollution control purposes is the hydrogenating desulfurization of residual oil obtained by topping. This process utilizes the reaction of sulfur compounds and hydrogen at high pressure to form hydrogen sulfide, under the action of catalyst such as Co-Mo/Al<sub>2</sub>O<sub>3</sub>. The hydrogenating desulfurization of heavy oil, which is the most important among the hydrogenating desulfurization, is divided into a direct desulfurization and an indirect desulfurization. The former method desulfurs the entire residual oil and is currently possible to reduce the sulfur content to as small as 0.1%. In

the latter method, the residual oil is separated into reduced pressure light oil and reduced pressure residual oil by a reduced pressure so that the metal contamination of desulfurizing catalyst could be reduced, and only the reduced pressure light oil is desulfured by hydrogenation.

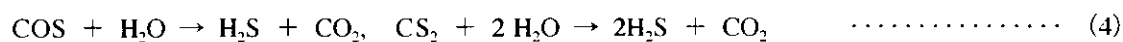
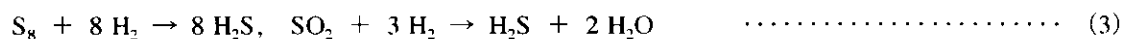
Therefore, the efficiency of desulfurization for the entire natural pressure residual oil is relatively low, and the sulfur content of desulfured heavy oil could be reduced to only about 1% or so. Presently, the technology of direct hydrogenating desulfurization of reduced pressure residual oil is already established.

## (2) Gas Cleaning and Sulfur Recovery

Although most of the gas released from the hydrogenating desulfurization equipment is hydrogen, some hydrogen sulfide and hydrocarbons are also contained. Accordingly, hydrogen sulfide is absorbed by the substance such as aliphatic alkanol amine and recovered after performing separation and condensation processes. A typical process of converting the recovered hydrogen sulfide into elemental sulfur is the Claus method. In this method, one third of hydrogen sulfide is burned in the combustion reaction furnace to form sulfur dioxide and then it is reduced to elemental sulfur by hydrogen sulfide, as shown in Eq.(1) and (2). As the conversion ratio of sulfur in the combustion reaction furnace is relatively low with 60-70%, the unreacted gas is further reacted in the subsequent 2-3 stages of catalytic reactors.



Several thousands ppm of hydrogen sulfide and sulfur dioxide are contained in the tail gas discharged from the Claus equipment, and small amount of COS, CS<sub>2</sub>, and sulfuric acid mist are also contained. The SCOT (Shell Claus Off-gas Treater) process is used to remove those substances. In this process, sulfur dioxide and others are reduced by hydrogen as shown in Eq.(3), and COS and others are decomposed by hydrolysis to recover hydrogen sulfide as shown in Eq.(4), and to recycle the latter to the Claus equipment.



The concentration of unreacted hydrogen sulfide in the exhaust gas from the absorption tower is about 300 ppm, and it is emitted from the combustion furnace as sulfur dioxide. Using the above described Claus process and tail gas treatment process in combination, the recovery efficiency of elemental sulfur from hydrogen sulfide reaches to 99.5-99.8%<sup>7)</sup>.

#### 6.5.4 Air Pollutants

The sources of air pollutant in the oil refinery to be noted are the storage tanks for crude oil and its products. The tanks are classified into a fixed roof type and a floating roof type, and both types of tank release hydrocarbon vapor as a result of the evaporation of oil. In the former type of tank, a roof equipped with internal pressure control valve is fixed to a top of cylindrical steel shell. The oil vapor is filled in the space between the oil surface and roof, and it is released to the atmosphere as the valve opens when the atmospheric temperature, and thus pressure of the space, increases. Also, when the temperature decreases, the gas inside the tank contracts to draw in the outside air. At the same time, the oil vaporizes to saturate the gas phase and the loss occurs. In the fixed roof tank, a certain amount of stored oil is lost by internal gas expansion and contraction caused by daily temperature change, and it is called the breathing loss. The vapor release from the fixed roof tank is also caused by the change in oil level which occurs when oil is supplied to or discharged from the tank. The loss caused by those operations is called the working loss. The floating roof tank has been constructed to prevent this working loss, and all the large crude oil storage tanks now used in the oil refinery belong to this type. The floating roof tank has a cylindrical steel shell and a floating roof that floats on the surface of stored oil. The floating roof that contacts with outside air covers the oil surface completely, and the roof and inside wall of the tank are sealed. The floating roof moves up and down following the movement of oil surface caused by supply and discharge of the stored oil. Evaporation of oil is minimized by the seal against the inside wall and the amount of evaporation is reduced to about 0.2-0.1% of that for the fixed roof tank. Major evaporation loss is caused by the wind around the tank and the evaporation of oil adhered to the inside wall remained when the oil surface lowers as the stored oil is discharged. There is data that about  $0.48 \times 10^{-3}$  kg of oil per 1 kl of discharged oil is released in the air through this loss <sup>6)</sup>.

Such an evaporation loss occurs during the process of loading of the oil fuels, and the oil vapor leaks through flanges, valves and pumps of oil pipeline in the oil refinery facilities. Generally, the evaporation loss is said to differ depending on the maintenance of oil refinery facilities <sup>5)</sup>.

Furthermore, air pollutants are generated in the combustion process of heating furnace (pipe still), catalyst recovery furnace and boiler in the oil refinery. Particularly, carbon particles and fine particulate metal salts could be contained in the exhaust gas of the catalyst recovery furnace. The source substance of offensive odor in the oil refinery is mainly hydrogen sulfide and mercaptane. The sources and counter measures for these substances are shown in Table 6.5.2 <sup>8)</sup>.



Table 6.5.2 Sources and Counter Measures for Offensive Odor in Oil Refinery

Equipment/Facility	Source	Counter Measure
Refining Facility	Offensive odor occurs at steam purge for the open inspection of the facility.	Condensation→Separation of Gas Phase→Gas Phase→Combustion→Liquid Phase→Drain
Tank	<p>① Offensive odor substances evaporate associated with evaporation loss from the fixed roof tank. Crude oil and crude naphtha containing offensive odor substances are generally stored in the floating roof tank.</p> <p>② Hydrogen sulfide is generated from the liquid sulfur tank.</p> <p>③ Characteristic smell is generated from the vent of asphalt tank.</p>	<p>① Absorption, adsorption, etc.</p> <p>② Absorption by sodium hydroxide solution.</p> <p>③ Combustion, absorption, etc.</p>
Shipping Facility	<p>① Hydrocarbon smell is generated at the loading of gasoline to the lorry.</p> <p>② Offensive odor is generated from hold and vent of the ship at oil loading works.</p>	<p>① Condensation, absorption balance line, etc.</p> <p>② Absorption by sodium hydroxide or kerosene.</p>
Drain Treatment Facility	Offensive odor substances dissolved into the drainage come up from oil separator, pressurized float up tank, and activated sludge aeration tank.	Attachment of cover to the tank, absorption or adsorption after sucking in by blower.

## 6.6 Transportation

### 6.6.1 Overview

The biggest air pollution source in the transportation sector is the motor vehicles. Among pollution caused by auto exhaust, that caused by CO was revealed from the beginning, but CO pollution has been kept below the environmental quality standards through various measures regulating the emission of exhaust gas. The motor vehicles, at present, is considered to be the biggest source of NO<sub>2</sub> and suspended particulate matters pollution, and the strengthening of measures against them is in demand.

### 6.6.2 The present situation of the motor vehicles as a facility of transportation

From the mid-1960's, motorization advanced rapidly in Japan, and the motor vehicles, whose position is rising, is becoming a major source of increasing amounts of air pollution, noise and vibration. When looking at the freight volume by transportation, motor vehicles are accounting for a steadily increasing amount year-by-year, rising from 15% in 1960, to 39% in 1972 and to 51% in 1993. Also, although the transportation of travelers has been dull recently, the ratio occupied by the motor vehicles is increasing yearly, with the ratio growing from 23% in 1960 to 51% in 1972 and to 60% in 1993.

Next, when looking at vehicle use, we see remarkable development with number of motor vehicles in operation 810,000 units in 1965 growing to 2,910,000 (3.59 times) in 1975, 4,820,000 (5.95 times) in 1985 and 6,810,000 (8.4 times) in 1994. The increased growth in passenger cars of many variations has been phenomenal. Also, the total distance traveled by vehicles is increasing in accordance with the growing number of vehicles as in 1994 it was 2.86 times that of 1971.

### 6.6.3 The present situation of the motor vehicles as a pollution source

#### (1) Pollutant emissions from motor vehicles

The motor vehicles, as a pollution source, is separated into gasoline engines and diesel engines, but there are differences in these two engine types as shown in Table 6.6.1, creating different characteristics of the exhaust gases and different countermeasures against the different emissions.

The discharge of pollutants from gasoline-engine vehicles is roughly from three areas as shown in Fig. 6.6.1. Exhaust emission is what after the gasoline is burned in the engine, is discharged from the muffler in the form of CO<sub>2</sub>, NO<sub>x</sub> and H<sub>2</sub>O as the products of complete combustion, and CO and HC as the products of incomplete combustion. Next is blow-by gas which leaks into the crankcase through a gap between the piston and cylinder wall and contains unburned fuel, mostly HC. Also, there is the evaporative emission. Because gasoline is a highly volatile fuel, it evaporates from the fuel supply equipment such as the fuel tank and the carburetor, polluting the atmosphere.

Table 6.6.1 Differences between gasoline engines and diesel engines

Category	Gasoline engine	Diesel engine
Fuel & characteristics	Gasoline High volatility	Light oil Volatility is low, but percentage of sulfur is high
Ignition & burning method	It mixes air with the vaporized fuel, (mixed air) beforehand, then the mixture is compressed and ignited by an electric spark.	Fuel is jetted into a chamber where air has been heat insulation adiabatic compressed to a high temperature to ignite and burn the fuel.
Burning characteristic	The mixing is continually adjusted so the fuel burns completely and the ratio of fuel and air does not change.	The air is always constant. As for the fuel, it fluctuates according to the output, big changes in the ratio of air and fuel.

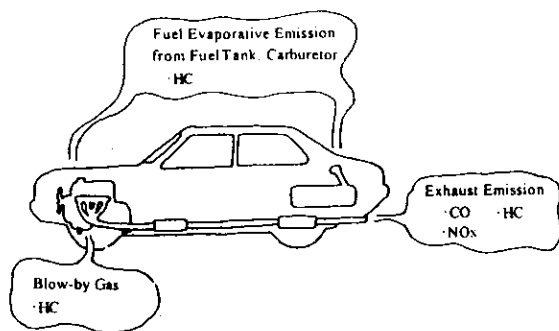


Fig.6.6.1 Emissions from Gasoline-fueled Vehicles

The pollutants discharged from diesel powered vehicles are shown in Fig.6.6.2. Because diesel fuel's volatility is lower than gasoline's, pollution of the air from emission evaporation after the fuel is burned is remarkably low. Also, as only air is pushed into the piston ring part in the early stages of burning, blow-by gas emissions are low resulting in a remarkably low concentration of HC. Therefore, most of the emissions from diesel engines that pollute the air are discharges from the exhaust. As shown in Table 6.6.1, with the diesel engine, an air-excess condition in burning occurs mostly and there is little discharge of CO and HC. However, on the other side is much NOx emission, and, also, black smoke and particulate matter cannot be avoided because of the partial lack of oxygen in the latter part of the burn period. Moreover, because there is a higher percentage of sulfur in diesel fuel than in gasoline, SO<sub>2</sub> is discharged, too.

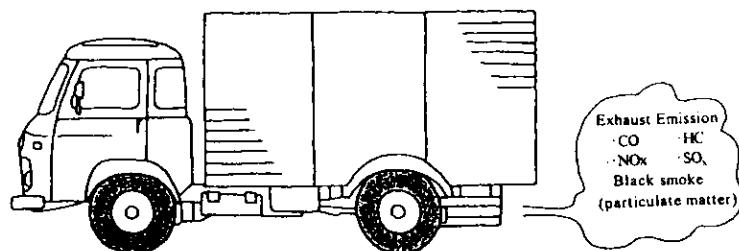


Fig.6.6.2 Pollutant emissions from Diesel-powered Vehicles

## (2) Pollutant emission level

The type of vehicle, its weight and running condition, among others, mainly change the pollutant which is discharged from the vehicle. Table 6.6.2 compares the emission level of various pollutants at an average running condition (average speed of 18.2 km/h) in the Tokyo area. Looking at the type of vehicle in Table 6.6.2, the emission volume of each pollutant increases as the size of the vehicle increases. This tendency is conspicuous with NO<sub>x</sub> and particulate matter (PM). When attempting to see the ratio of NO<sub>x</sub> emissions, we find that, with a gasoline-engine passenger car at 1, a 10-ton diesel powered truck comes in at 31 and its PM emission is at 55. As for CO, there is originally little emission volume in a diesel vehicle, so that in a 10-ton diesel-powered truck is almost the same as a gasoline passenger car. As for HC, remarkably low emission volume of gasoline-engine passenger cars makes the comparative emission ratio of diesel-powered trucks is, however, almost the same as that of heavy-duty gasoline-fueled vehicles.

Table 6.6.2 Emission level by hazardous substances and by vehicle category (Unit: g/km)

Hazardous substances	Gasoline-fueled passenger cars	Medium-duty gasoline fueled vehicles 750kg	Heavy-duty gasoline fueled vehicles 1 ton van	Diesel- powered trucks			
				2 t	4 t	8 t	10 t
NO <sub>x</sub>	0.26	0.39 (1.5)	0.96 (3.7)	3.25 (12.5)	3.99 (15.3)	5.13 (19.7)	8.05 (31.0)
CO	3.99	5.76 (1.4)	15.84 (4.0)	1.63 (0.4)	1.88 (0.5)	2.03 (0.5)	3.56 (0.9)
HC	0.19	0.20 (1.1)	1.39 (7.3)	0.78 (4.1)	1.12 (5.9)	1.11 (5.8)	1.86 (9.8)
PM	0.012	0.010 (0.8)	0.022 (1.8)	0.173 (14.4)	0.365 (30.4)	0.505 (42.1)	0.656 (54.7)

Note: The numbers in the parenthesis are the ratios with 1 standing for gasoline-fueled passenger cars.

## (3) The contribution ratio of motor vehicles as a pollution source

Fig.6.6.3 shows the NO<sub>x</sub> emission per pollution source on a nationwide level. The contribution of motor vehicles accounts for 42%, including both controlled and uncontrolled vehicles. The uncontrolled vehicle is equipped with special characteristics and includes industrial machines (forklifts, etc.), construction machines (hydraulic shovels, bulldozers, wheeled loaders, etc.) and agricultural machines (tractors, combines, etc.). Most NO<sub>x</sub> emissions occur from these vehicles during their working periods. This pollution source ratio changes somewhat when it occurs in a metropolitan area. The emission rate of NO<sub>x</sub> per pollution source in metropolitan areas is shown in Fig.6.6.4. The emission rate of motor vehicles exceeds 60% for both controlled and uncontrolled vehicles. Also, the NO<sub>x</sub> contribution rate according to the category of motor vehicle is shown in Fig.6.6.5, where it can be seen that the emission rate is high for the freight vehicle sector such as ordinary sized trucks next to the uncontrolled vehicle category.

The SO<sub>x</sub> emission rate for motor vehicles is increasing (55% of all the SO<sub>x</sub> emission volume is from motor vehicles according to the 1990 emission volume calculation for the Tokyo area) mostly because of the reduction in the emission volume of factories. However, as the percentage of sulfur in light oil has been reduced in recent years, the emission contribution rate seems to be declining.

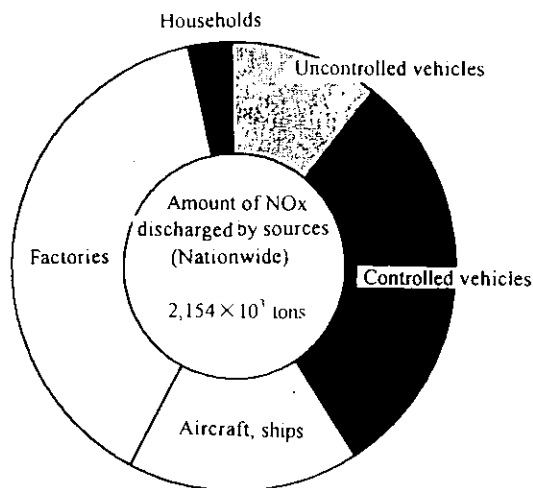


Fig.6.6.3 Contribution to NO<sub>x</sub> emissions by source (Nationwide)

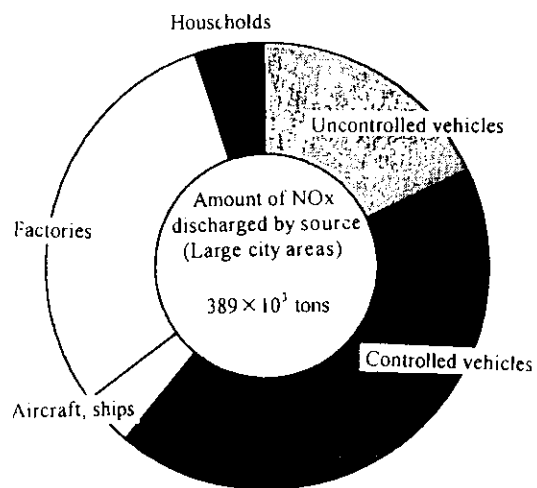


Fig.6.6.4 Contribution to NO<sub>x</sub> Emissions by Source (Large city areas)

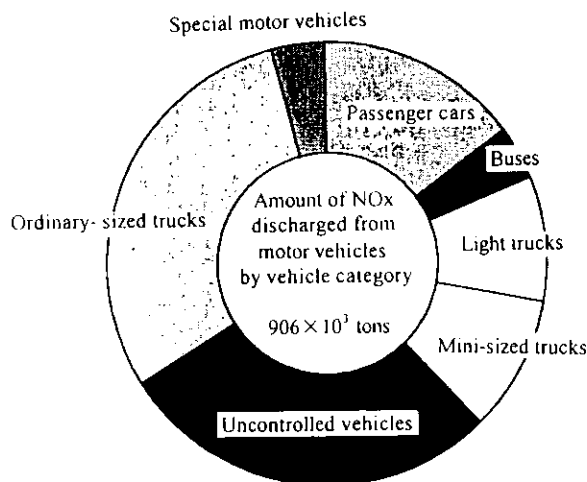


Fig.6.6.5 Contribution to NO<sub>x</sub> emissions from motor vehicles by vehicle category (Nationwide)

#### 6.6.4 Other modes of transportation

##### (1) Aircraft as a pollution source

When treating aircraft exhaust gas as a pollution source on the ground, an altitude of 1,000 meters is set generally for measuring purposes. In dealing with the discharge of exhaust gas for aircraft, this includes the landing, the parking and the reaching of 1,000 meters on takeoff. This mode of landing and take off for aircraft is called the landing/takeoff (LTO) cycle and the mode of operation for this question is classified into approach, idle, take off and climb. Therefore, to calculate the exhaust gas volume from aircraft, the duration should be understood according to each mode and the pollutant emission factor (the pollutant discharge volume per unit of time and engine mode) of the concerned aircraft. The duration according to this mode depends on the airport and aircraft type. However, the domestic research data on the pollutant emission factor in various engine types is scarce and the US's EPA published value for the pollutant emission factor per engine classification is depended upon. When calculating the NO<sub>x</sub>

emission volume for Haneda Airport using these numerical values, the volume becomes 400 tons annually, accounting for 2.4% of the NOx discharge volume for the whole Tokyo area. Also, as for the average NOx discharge volume per aircraft, it reaches 8.7 kg per 1 LTO cycle, equal to the discharge volume of a 10-ton truck running at an average speed of 18.2 km/h for 1,080 kilometers.

#### (2) Maritime vessel as a pollution source

When evaluating maritime vessel as a pollution source, it is necessary to examine it by dividing it into the vessel being in motion or being anchored in the harbor. Also, when anchored, the emission source depends on whether the loaded or unloaded. The hazardous substance discharge volume from a vessel can be calculated when the conditions are understood such as the vessel's operational mode observation rate on its sailing route and the longshore craft's ratio of idle time, its boiler and its diesel load factor. As shown in Fig.6.6.4, maritime vessel, along with aircraft, in its role as a NOx pollution source contributes 4% of the whole for the metropolitan area. The personal contribution rate of ships and aircraft depends on the scale of the harbor and the airport, etc. In the NOx emission volume calculation results for 1990 in the Tokyo area, the volume for ships accounts for 2.7% of the whole volume and, in Yokohama, this ratio is 9%.

#### 6.6.5 Indicated problems

There still are many problems in grasping the pollutant emission situation in the transportation sector. First, it is said that large-scale equipment is necessary to grasp the actual state of the pollutant discharge from motor vehicles or vessels and there are few made which have a capacity of measuring exhaust gas. Second, these transportation facilities are the pollution source of hazardous chemical species such as benzene, 1,3 butadiene, aldehyde and benzo(a)pylene. Therefore, grasping the actual state of the discharge is becoming an urgent business.

## 6.7 Sources of offensive odors

### 6.7.1 State of offensive odor complaints

When the number of complaints annually by type concerning the seven major pollutants as compiled by the Environmental Disputes Coordination Commission are examined, offensive odors are second only to noise, occupying a total of approximately 17% of complaints as a whole. This has not changed in the more than 20 years since these surveys began <sup>1)</sup>.

When the sources of these offensive odor complaints are examined for 1995, the most common area is the "services and others" category, at 2,930 complaints, or 26.0% of the total, followed by the "livestock and farming industry" at 1,824 complaints (16.2%). In third place comes "private housing, apartments, and dormitories" at 1,481 complaints (13.1%). Further, the food processing industry, chemical plants, and other manufacturing plants within the manufacturing industry occupy a total of approximately 1/4 of all complaints, while as before, offensive odors are one of the largest causes of complaint (Fig.6.7.1). Moreover, in the "service industry and others" category, the majority of complaints are to do with restaurants, field burning, automobile repair plants, dry cleaners, and so forth, and in the same way, in the "other manufacturing plants" category, the majority of complaints are addressed against paint factories, metal products manufacturing plants, woodworking plants, and so on <sup>2)</sup>. Given these trends, the outstanding feature of offensive odor complaints in recent years has been a tendency towards an increase in the percentage of offensive odors caused by daily living, such as ordinary private housing, apartments, and dormitories, sewage, and tap water, and the offensive odor complaints relating to urban living account for 46.2% of all complaints as a whole.

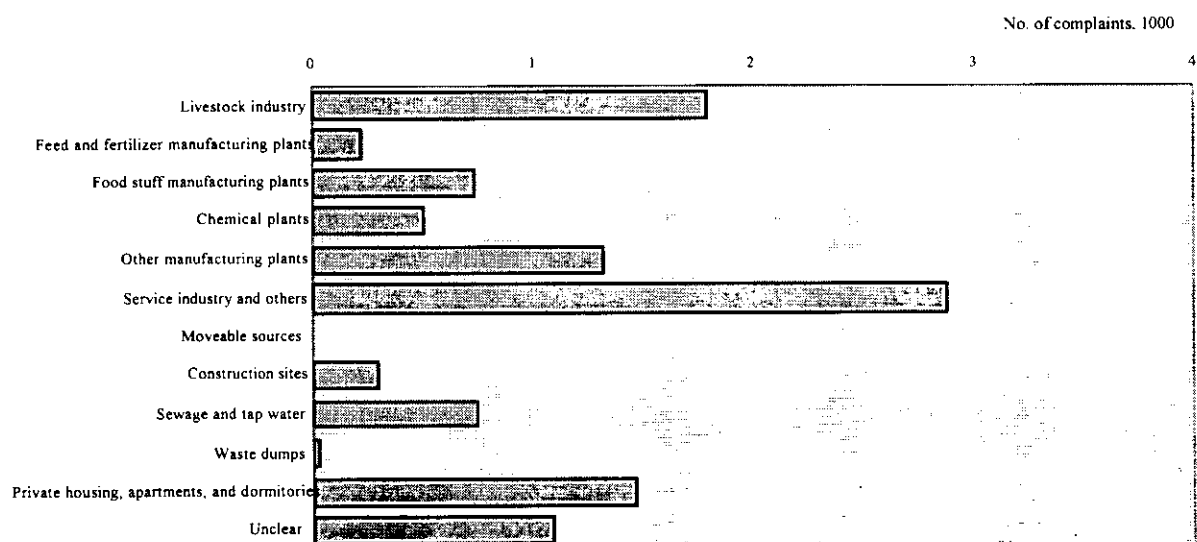


Fig.6.7.1 Offensive odor complaints in Japan (1995)

### 6.7.2 Worksites generating specific offensive odor substances

The Offensive Odor Control Law regulates as its target 22 proscribed substances such as ammonia, hydrogen

sulfide, fatty acids, and so forth, and these substances are called specific offensive odor substances. The main sources of these specific offensive odor substances are shown in table 6.7.1<sup>3)</sup>.

Table 6.7.1 Main sources of specific offensive odor substances

Ammonia	Livestock and farming industry, chicken excrement drying sites, compound fertilizer manufacturing, starch manufacturing, dead animal processing plants, fish entrails and bone processing plants, feather processing plants, waste disposal plants, raw sewage treatment plants
Methyl mercaptane	Craft pulp manufacturing, chemical plants, fish entrails and bone processing plants, waste processing plants, raw sewage treatment plants, sewage treatment plants
Hydrogen sulfide	Livestock and farming industry, craft pulp manufacturing, starch manufacturing, cellophane manufacturing, viscous rayon manufacturing, dead animal processing plants, fish entrails and bone processing plants, feather processing plants, waste processing plants, raw sewage treatment plants, sewage treatment plants
Methyl sulfide	Craft pulp manufacturing, dead animal processing plants, fish entrails and bone processing plants, waste processing plants, raw sewage treatment plants, sewage treatment plants
Methyl disulfide	Craft pulp manufacturing, dead animal processing plants, fish entrails and bone processing plants, waste processing plants, raw sewage treatment plants, sewage treatment plants
Trimethyl amine	Livestock and farming industry, compound fertilizer manufacturing, dead animal processing plants, fish entrails and bone processing plants, fishing industry canneries
Acetaldehyde	Acetaldehyde manufacturing plants, acetic acid manufacturing plants, vinyl acetate manufacturing plants, chloroprene manufacturing plants, tobacco manufacturing plants, compound fertilizer manufacturing, fish entrails and bone processing plants
Aldehydes Propion aldehyde n-Butylaldehyde abutyl aldehyde n-Valeraldehyde Isovaler aldehyde	Paint factories, other metal manufacturing plants, automobile repair plants, printing plants, fish entrails and bone processing plants, oil and fat food processing plants, transport machinery and equipment manufacturing plants
Solvents Isobutanol, Ethyl acetate Methyl isobutyl ketone Toluene, xylene	Paint factories, other metal manufacturing plants, automobile repair plants, woodworking plants, fiber plants, other machine manufacturing plants, transport machinery and equipment manufacturing plants, foundries
Styrene	Styrene manufacturing plants, polystyrene manufacturing plants, SBR manufacturing plants, FRP manufacturing plants, cosmetic plywood manufacturing plants
Propionic acid	Fatty acid manufacturing plants, dye plants, livestock sites, dead animal processing plants, starch manufacturing plants
n-Butyric acid, n-Valeric acid, Isovaleric acid	Livestock sites, starch manufacturing plants, dead animal processing plants, fish entrails and bone processing plants, livestock food processing plants, raw sewage treatment plants, waste disposal plants

### 6.7.3 Impact of the main offensive odor generating worksites

The examination of emissions of offensive odors generated by offensive odor generating worksites, and the relationship to the extent of local environmental impact by these offensive odors, is important from the point of view of discussing anti-offensive odor measures. Generally, in order to investigate the environmental impact caused by



sources of offensive odors, methods are adopted for calculating the environmental concentration using various diffusion algebraic formulae. However, in the case of offensive odors, understanding specific sources and the amount generated is problematic, and there are many cases where there is a disparity with reality. Shigeta used the TOER (Total Odor Emission Rate) and have come to calculate the impact of offensive odors experimentally as shown in Table 6.7.2<sup>4)</sup>. Moreover, the OER is a value that multiplies the amount of gas emitted (m<sup>3</sup>/min.) by the odor concentration of their source, and the TOER, when there are multiple sources within the same worksite, is the sum of all the various OERs<sup>4)</sup>.

Table 6.7.2 Relationship between TOER and range of effects

TOER	Causes of offensive odor pollution Condition	Typical examples of originating industries (No offensive odor protection law)	Area of impact
Max. 10 <sup>4</sup>	Only in special circumstances	Bakeries Distilleries	
10 <sup>5-6</sup>	Although offensive odor pollution is only happening on a small scale at present, includes the possibility.	Paint and solvent factory Printing and ink plant Leather factory, FRP works Feed and fertilizer plant Sewage treatment plant	Maximum distance reached by offensive odor is 1 to 2 km, offensive odor complaints are centered on a 500 m radius, with no complaints from 1 km or more away.
10 <sup>7-8</sup>	Offensive odor pollution is happening on a small to medium scale.	Foundries Raw sewage treatment plants Pig and chicken farms Petroleum chemical plants	Maximum distance reached by offensive odor is 2 to 4 km, offensive odor complaints are centered within a 1 km radius.
10 <sup>9-10</sup>	Offensive odor pollution is happening on a large scale.	KP plants Cellophane plants Dead animal processing plants Rayon plants	Maximum distance reached by offensive odor is within 10 km, offensive odor complaints are centered within a 2 to 3 km radius.
10 <sup>11-12</sup>	A few examples at the largest offensive odor source.	Large-scale KP plants with no anti-pollution measures.	Maximum distance reached by offensive odor is 10 km, with damage occurring within a 4 to 6 km radius.

#### 6.7.4 Main worksite offensive odor sources

In the case of offensive odors, pinpointing the sources is not necessarily an easy task. Predictable sites vary, and an odor generated from an unexpected site can have a major impact on the local surroundings. Mr. Takahashi<sup>5)</sup> has compiled a report on the sources and concentrations of offensive odors generated in sewage treatment plants. The state of generation of offensive odors in sewage treatment plants is shown in Table 6.7.3. Further, Mr. Tanigawa<sup>6)</sup> has reported on the offensive odors generated by the treatment and disposal of urban waste and, as classifications of those odors, has broken them down into offensive odors generated by waste itself, offensive odors generated by waste processing (gasses emitted by burning, compost processing, landfills, etc.), and other (exhaust gas from garbage trucks, waste water treatment facilities, etc.). The main sources of offensive odors from urban waste processing and disposal are shown in Table 6.7.4.

Table 6.7.3 Offensive odor sources and odor concentrations at sewage treatment plants

Offensive odor source	Minimum value	Maximum value	Geometric mean
Reserve aeration tank	730	7,300	2,300
Grit chamber	14	2,300	210
Screen combers	980	2,300	1,500
Gravity thickener concentration tank	230	230,000	6,200
Vacuum filters	<10	310	28
Pressure filters	<10	10	<10
Belt press filters	5,500	13,000	7,300
Centrifugal dehydrator	—	—	55
Multihearth furnace exhaust gasses	3,100	55,000	16,000
Precipitation	18	41,000	220
Aeration tank	10	1,900	110
Sludge cleaning tank	<10	980	120

Table 6.7.4 Main sources of offensive odors in waste treatment and processing

Waste treatment and processing	Offensive odor sources	Types of offensive odor
Collection	Waste dumps Dump trucks	Offensive waste odors, offensive automobile gas emissions Offensive waste odors, offensive automobile gas emissions
Transportation	Dump trucks	Offensive waste odors, offensive automobile gas emissions
Intermediate processing	Urban waste Incinerator facilities Platform Waste pits Stacks Ash pits Environmental countermeasure facilities Garbage hauling vehicles	Offensive waste odors, offensive automobile gas emissions Offensive waste odors Offensive odors from incinerator gasses Offensive odors from ash pits Offensive waste water odors, offensive pharmaceutical odors Offensive automobile gas emissions, offensive waste odors Offensive ash incineration odors
Final disposal	Landfills	Offensive landfill odors, offensive waste odors, offensive ash incineration odors. Offensive motor vehicle exhaust gas, offensive waste water odors

The main sources of offensive odors generated by other offices are shown below by worksite.

Table 6.7.5 Main sources of offensive odors generated by other offices

Worksites	Main sources of offensive odors
Livestock and farming industry:	Pig pens, henhouses, excrement and its processing facilities, fertilizer dumps, fertilizer drying facilities.
Chemical processing plants:	Cookers, presses, dryers, and their waste steam, waste water processing equipment, raw material dumps, concentration equipment drains.
Paint factories:	Paint booths, etching, glazing kilns.
Print works:	Drying equipment built into printing machines, printing parts, cleaning work on plates and rollers, printing ink, mixing work such as with dilutants, ink and solvent store rooms.
Food processing plants:	Waste water, burning of raw materials and packaging, fumigants, boiler smoke, raw material dumps.
Petrol refineries <sup>7)</sup> :	Gas by-products from distillation equipment, decomposing equipment, hydroxide refining equipment, and sulfur separating equipment, sulfur recovery equipment, storage facilities, flare stack, waste water.
Chemical plants <sup>8)</sup> :	Reaction facilities, distillation facilities, absorption and adsorption facilities, drying facilities, burning facilities.
Sewage treatment plants <sup>9)</sup> :	Grit chamber, sedimented basin, aeration tank, inspection hoppers, Grit hoppers, concentrated sludge tank, sludge storage tank, mud elimination tank, water extractors, sludge incinerators.
Raw sewage treatment facilities <sup>10)</sup> :	Insertion pores, insertion rooms, aeration tank, elimination tank, concentration tank, sludge processing facilities, sludge incinerators.
Craft pulp manufacturing <sup>10)</sup> :	Distillation threshers, evaporators, recovery boilers, lime kilns.

## 6.8 Emission factors for atmospheric pollutants

### 6.8.1 Outline <sup>1)</sup>

In order to rationalize the air pollution counter measures in a given region, it is essential to estimate the total quantity of air pollutant emissions by kind of pollutant as well as human activities. For this estimation, the air pollutant emission factors calculated from air pollutant emission due to human activities such as fuel combustion or industrial production and these activities have been utilized. Further, this technical constant, which are necessary for estimating the total regional emissions of air pollutants, can be used to predict the concentration of air pollutants by means of the theory of atmospheric diffusion, and are also employed both in establishing plans for the regional reduction of air pollutants emission and in the running of environmental impact assessment systems.

In 1970, the government of the United States of America announced the estimation results of total amount air pollutant emissions on fire kinds of pollutants in nation wide in 1968 <sup>2)</sup>. Further, the American government issued a report <sup>3)</sup> by R. L. Dupley concerning the air pollutant emission factors that considered these calculation standards, and which were translated into Japanese by the Japanese Fuel Association <sup>4)</sup>. In this report, R. L. Dupley defined the emission factor as follows: "The emission factor for air pollutants is a statistical average of the rate at which a pollutant is released to the atmosphere as a result of some activity, such as combustion or industrial production. The emission factor thus relates the quantity of pollutants emitted to some indicator such as production capacity, quantity of fuel, or vehicle miles traveled by autos." thereby introducing both the overview of causes that generate air pollutants such as fuel combustion, various manufacturing processes, means of transport, and so forth, and the emission factors that they cause. Further, in the introduction to this report <sup>3)</sup>, Dupley wrote that this collated data from already-published technical papers, and that the calculation basis had been compiled and edited by M. Mayer in 1965 <sup>5)</sup>.

On the other hand, the MITI, Agency of Industrial Science and Technology in Japan examined the state of smoke and soot emissions from 271 plants nationwide in 1960, and published their results in 1961, but in this report <sup>6)</sup>, the quantity of soot and dust emissions from cement kilns was calculated as the quantity of soot and dust emissions per cement clinker production t. That result was 0.3 to 3.3 kg/t, and stated that with the wet process, there were many cases where the results were 1.8 kg/t or less, and with the dry process, there were many cases where the results were 1.0 to 3.3 kg/t, and that although no specific air pollutant emission factor was included, it can be inferred from the American government that this concept was held from the first. In spite of which, there have been no similar activities by MITI to be seen since.

At any rate, in the same way as the American government, which was noted previously, the MITI report <sup>6)</sup> also based its calculations on a air pollutant concentration in exhaust gas that had already been measured, but thereafter, in the 1970s when the emission factor was considered important in Japan, no technical reports concerning the sources of air pollution source were to be seen, and the formation of an independent organization for actually measuring this data deserves special mention.

## 6.8.2 Air pollutant emission factors from stationary sources

### (1) Calculating air pollutant emission factors from stationary sources in Japan

It was when the environmental pollution control programs were planned in 1970 that the total quantity of emissions of air pollutants in a fixed region in Japan were estimated, but if taken separately from the aforementioned production and technology unit's report <sup>6)</sup>, the emission factors collated from the actually measured data can be said to be the first ever for Tokyo <sup>7)</sup>.

In 1971, the pollution control bureau for Kanagawa Prefecture obtained formulae pertaining to the emission of sulfur oxide and the quantity of heavy oil burnt in 40 major factories in both Kawasaki and Yokohama cities, and introduced regulations for reducing the total quantity of sulfur oxide using a proportional model, and these relevant formulae can also well be called emission factors <sup>8) 9)</sup>.

In 1972, the Air Quality Bureau, Environment Agency established the "Investigation Committee for Air Pollution Due to Photochemical Reaction," added the "Emission Source Sub-Committee," and began examining and investigating on the air pollutant emission factors from stationary combustion sources <sup>1) 10)</sup>. As stated previously, this activity led to the formation and promotion of an independent organization for the measuring of air pollutants in exhaust gas, and this led to its continuation, in 1973, as the "Investigation Committee for Source Generation Air Pollution" <sup>1)</sup>. Next, in 1976, measurement data was also collected by local governments and, for example, the relationship between nitrogen oxides emissions and the amounts of burning of fuel was established with regard to power plant and industrial boilers, as shown in Fig. 6.8.1. Further, the formulae pertinent to such load fluctuations, emissions of NO<sub>x</sub>, and quantity of fuel combustion were also established, and the emission factors for NO<sub>x</sub> concerning boilers were also obtained, as shown in Table 6.8.1. Moreover, the mean proportion of fuel combustion and emissions relating to soot and dust, NO<sub>x</sub>, CO, and hydrocarbon species was calculated both by fuel and by type of source and, for example, the emission factors as shown in Table 6.8.2 were analyzed <sup>1) 11-13)</sup>.

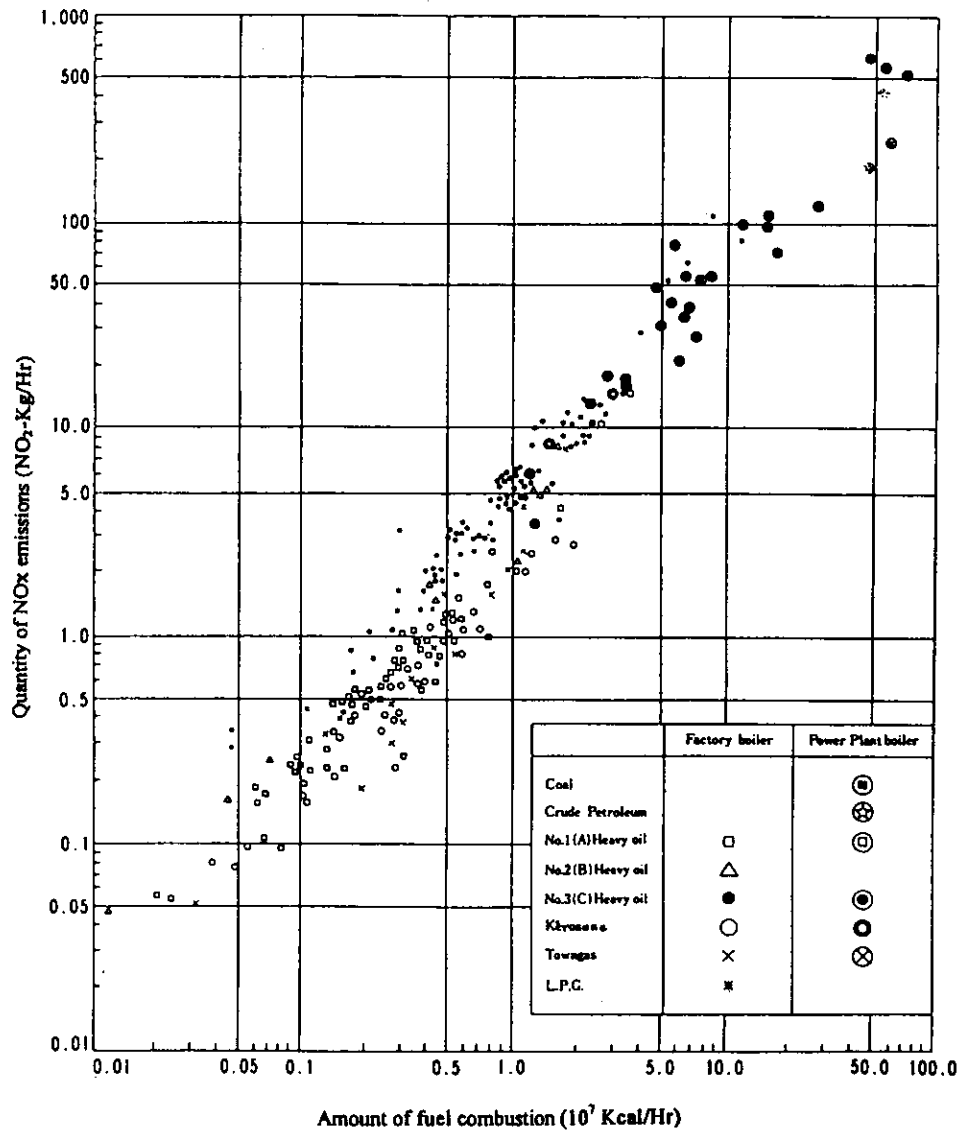


Fig.6.8.1 Relationship between quantity of NOx emitted by industrial boilers used in power generation and amount of fuel combustion.

Table 6.8.1 Emission factors for NOx from industrial boilers and power plant.

Rating of fuel combustion (10 <sup>7</sup> Kcal/h)	NOx emission factor (NO <sub>2</sub> -Kg/10 <sup>8</sup> Kcal)			
	Load 0.4	Load 0.6	Load 0.8	Load 1.0
10	49.1	57.3	64.1	69.8
30	49.7	58.1	64.9	70.7
50	50.0	58.5	65.3	71.2
70	50.2	58.7	65.6	71.5
90	50.3	58.9	65.8	71.7
100	50.4	58.9	65.9	71.8

Table 6.8.2 Examples of emission factors from stationary combustion sources

Combustion facilities	Kind of fuel	Emission factor (Kg/10 <sup>6</sup> Kcal)					
		Soot and dust	NOx	Carbon monoxide	Hydrocarbons		
Power-plant boiler	C heavy oil	4.7	63.5	78.9	37.1		
	A heavy oil		50.7				
	Crude oil		56.5			3.3	89.0
	LNG		38.9				
	Coal		128.0				
Industrial boiler	C heavy oil	12.2	56.6	7.5	0.11		
	B heavy oil	5.9	40.5	7.1	1.12		
	A heavy oil	6.4	25.2	3.5	0.43		
	kerosene	5.6	20.7	17.3	0.08		
	LPG		32.4		0.09		
Glass melting furnace	C heavy oil	48.1	251.0	32.5	0.25		
	B heavy oil		257.0				
	A heavy oil		121.0				
Cement kiln	Dry process	9473	127.0	83.2	4.64		
	Wet process	11782	162.0	12.2	1.63		
	Lepol	14414	212.0	246.0	1.42		

(2) Examples of calculated results of air pollutant emission factors

Thereafter, the government of the United States of America also calculated the emission factors for many different air pollutants, and published their findings<sup>14)</sup>. These examples are shown in Table 6.8.3 and 6.8.4<sup>14)</sup>.

Table 6.8.3 Emission factors for stationary combustion sources (Fuel oil burning, no controls)

		Soot and dust	Sulfur dioxide	Anhydrous sulfuric acid	Carbon monoxide	Nitrogen oxides
(amount of air pollutants emitted - Kg/Amount of fuel oil firing-kℓ)						
Boiler type	For power generation (Heavy oil firing)		19S	0.34S	0.6	8.0
	For industrial use (Heavy oil firing)		19S	0.24S	0.6	6.6
	For industrial use (Kerosene firing)	0.24	17S	0.24S	0.6	2.4
	For commercial use (Heavy oil firing)		19S	0.24S	0.6	6.6
	For commercial use (Kerosene firing)	0.24	17S	0.24S	0.6	2.4
	For domestic use (Kerosene firing)	0.3	17S	0.24S	0.6	2.2

Note: S: Sulfur content in the fuel (%)

Table 6.8.4 Evaporation loss of hydrocarbon from gasoline supply

Emission source		Emission factor (Emitted gasoline vapor-mg/Amount of gasoline fuel-l)
Supplied by underground tank	Splash fill (On the supply pipe surface)	1440
	Submerged fill	840
Underground tank venting loss		120
Automobile tank fuel supply loss	(No control)	1320
	(Control)	132
	Spillage	80

(3) Use of emission factors of air pollutants relating to stationary sources

Thereafter, the emission factors shown in both Table 6.8.1 and in Table 6.8.2 were used in estimating the total quantity of air pollutant emissions of NO<sub>x</sub>, etc., in Japan nationwide <sup>1)</sup> and, moreover, were used in establishing a plan for reducing the total emission of NO<sub>x</sub>, thereby proving to be very useful to Japan in her pursuit of air pollution control programs.

Further, as was stated previously, the emission factors have also been used in creating a regional diffusion model <sup>15)</sup> for air pollutants, in order to evaluate the environmental impact.

6.8.3 Air pollutant emission factors from mobile sources <sup>16)</sup>

The quantity of air pollutant emissions from automobiles differ depending upon the model of car (engine type [gasoline, diesel], size [weight of vehicle, load] quantity of exhaust), running type (driving pattern), and type of exhaust gas control measures. These different factors are made as equitable as possible, the overall composition is thoroughly understood, and the emission factors are vitally important as basic data for calculating the proportion that different types of vehicle contribute to pollution.

(1) Relation between driving and air pollutant emissions (general trends)

The general trends of the relationship between driving and air pollutant emissions are shown in Table 6.8.5. Moreover, for calculating the emission factors, it is vitally important to thoroughly and effectively understand the mean of the types of vehicle, chassis weight, load, emissions, and driving pattern.

Table 6.8.5 Relation between air pollutants and driving

CO, HC, Fuel costs	Depends on mean vehicle speed.
Diesel vehicle NO <sub>x</sub>	The quantity of low speed emissions during urban driving is high. On the expressway, however (when there are no traffic jams), conversely, the quantity of high speed emissions is high.
Gasoline vehicle NO <sub>x</sub>	Does not rely heavily on mean vehicle speed. At times of high load (during acceleration, during high speed driving), emissions are high.

(2) Overview of the procedure for calculating emission factors

In order to calculate the emission factors, first of all a survey is taken of the actual driving used by test cars on roads in the area targeted for calculation, and the driving results obtained are analyzed and a representative driving pattern compiled.

Next, the calculations of the emission factors for the target vehicle are set on the chassis dynamo meter, the compiled driving pattern is recreated and, during a mock run, the quantity of atmospheric pollutants emitted in the exhaust gasses is measured. In addition, the measurements obtained are collated and analyzed, and the emission factors thereby calculated.

(3) Actual driving survey using test vehicles in the target area

An actual driving survey is conducted using a test vehicle with the aim of understanding the automobile driving



conditions on roads in the area targeted for calculating the emission factors. The driving method used follows regular automobile driving trends, the test vehicle is driven, and vehicle speed the idling time, cruise time, amount of both acceleration and deceleration in time blocks of, for example, every 0.5 seconds are recorded. And both the running time and roads are considered under multiple traffic conditions before a decision is reached.

(4) Driving pattern sampling

A sample of the mean driving pattern (taken to be approximately 20 minutes of ordinary driving) for vehicle speed, idling time, cruising, amount of both acceleration and deceleration, and the time period is taken from the results recorded above. In addition, in the case of Tokyo, a single pattern is sampled for the urban expressway, and the mean of 10 patterns (4.5 to 45.0 km/h) sampled for ordinary roads.

(5) Experiments to measure exhaust gasses from the chassis dynamo meter

The representative driving pattern is recreated on the chassis dynamo meter, a mock test run is conducted, and the exhaust gasses are analyzed and measured. The model of vehicle measured is selected following consideration of the type of vehicles driven in the survey test area. In addition, the chassis dynamo meter is fixed to a roller device on the automobile motor axle, the driving pattern is established, and the device recreates engine running conditions while the vehicle in question remains stationary. Although there is running resistance to the roller, the load that the mechanism bears depends upon the vehicle speed. Because running resistance differs depending on the model of the vehicle, if the running resistance in question is discordant, then actual measurements are necessary.

(6) Compiling emission factors

The measurement results are adjusted depending on the model of car. In addition, if the test is held in Tokyo, a regression curve is normally taken to be the emission factor. Fig.6.8.2 shows the results of emission factors calculated for NOx in Tokyo in 1994.

(7) Calculating air pollutant emissions

The emission factor for automobiles is always represented as (g/vehicle · km). In addition, there are many times when “vehicle” is omitted. Moreover, in order to calculate the quantity of emissions, the model of vehicle (vehicle · km/h) corresponding to the emission factor is necessary. The method for calculating (vehicle · km/h) is to identify the amount of traffic (y vehicles/h) over a fixed distance (x km), and to then multiply these to obtain (x · y vehicles km/h), namely, the total quantity of air pollutant emissions is obtained by Eq. (1) and (2).

$$\text{Quantity of emissions per model of car (kg/h)} = \text{fixed distance (x Km)} \times \text{amount of traffic per model of car (y vehicles/h)} \times \text{emission factor per model of car (g/vehicle · Km)} \dots\dots\dots (1)$$

$$\text{Total quantity of emissions (Kg/h)} = \text{model A (Kg/h)} + \text{model B (Kg/h)} + \dots\dots\dots (2)$$

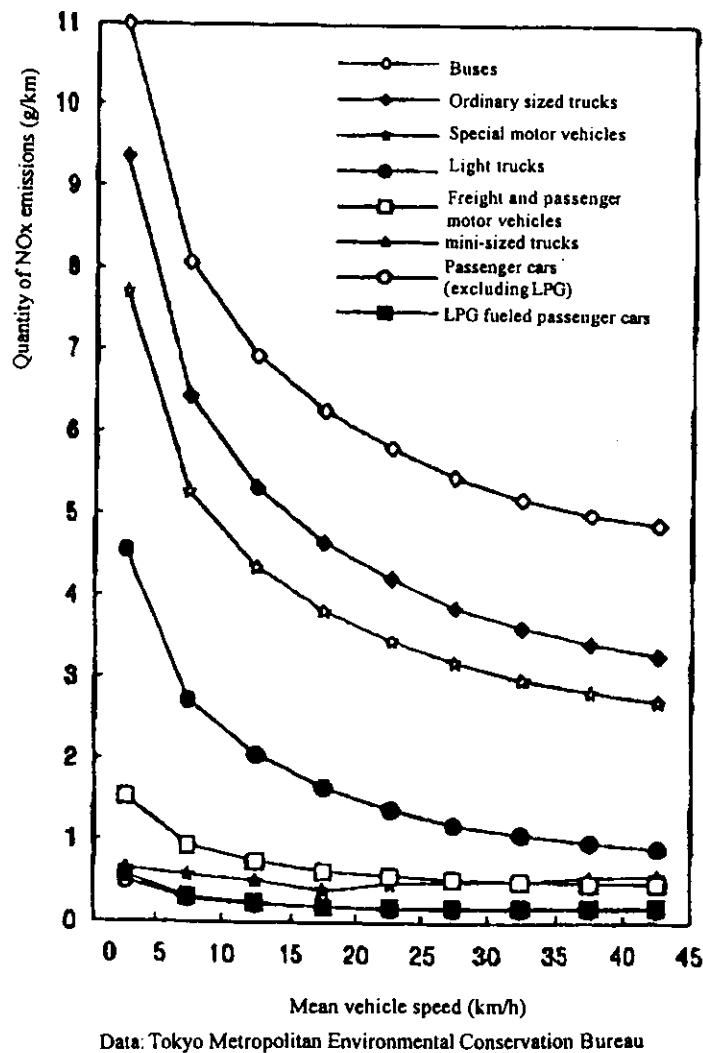


Fig.6.8.2 NOx emission factors (fiscal 1994)

(8) General noteworthy items

Because the emission factors are both regional and mean values, in order to use a fixed route and time period, it is necessary to be cautious. Because the emission factors are necessary to the very end for calculating the quantity of emissions, if such data as, for example, the amount of traffic, necessary for these calculations is insufficient, no matter how great a level of accuracy is demanded, it is virtually meaningless, or else have to be taken as the units of the emission factors that correspond to the amount of traffic flow data.

#### (6.1) Literature cited

- 1) Kurokawa, M.; Combustion Engineering, pp.1-14, Gihodo (1953)
- 2) Tsuji, S.; Combustion Technologies for Environmental Protection, Association of Japanese Thermal Energy Technology (1973)
- 3) Shibata, K.; Power Station and Substation, Denki Shoin (1972)
- 4) Air Quality Bureau, the Environmental Agency of Japan; Report on the Air Pollutant Emission Factor from Stationary Combustion Sources (1977)
- 5) Himi, Yasuji; Future of Air Environment Protection and the Role of Science and Technology, J. of Environmental Pollution Control, 21, 310-312 (1985)
- 6) Hagiwara, H. and Tamaki, T.; MHD Power Generation, Sogo Kagaku Shuppan (1975)

#### (6.2) Literature cited

- 1) Japan Society of Powder Technology Ed.; Dictionary of Particle Engineering Terms, Nikkan Kogyo Shinbunsha (1981)
- 2) Air pollution control division, Air Quality Bureau, the Environmental Agency of Japan, Ed.; Explanation of Dust Generating Facility, Gyousei (1985)
- 3) Hatta, S.; Introduction to Chemical Engineering, Kyoritsu Shuppansha (1951)
- 4) Himi, Y.; On the Air Pollutant Emission Factors from the Stationary Sources, *J. Japan Soc. Air Pollut*, 21, 465-485 (1986)

#### (6.3) Literature cited

- 1) Air Quality Bureau, the Environmental Agency of Japan; Report on the Air Pollutant Emission Factors from Stationary Combustion Sources (1977)
- 2) Ministry of International Trade and Industry; Industrial Location and Environmental Protection Bureau ed., Law and Technology of Environmental Protection, Industrial Pollution Control Association of Japan (1978)
- 3) Himi, Yasuji; On the Air Pollutant Emission Factors from the Stationary Sources, *J. Japan Soc. Air Pollut*, 21, 465-485 (1986)
- 4) R.L. Duplay, Trans. Fuel Association; Sources of Air Pollutant-Formation Process and Emission Factor-, Yokogawa Shobo (1971)
- 5) Shoji, T.; Acid, Alkali and Fertilizer, Sangyo Tosho (1948)
- 6) U.S. Environmental Protection Agency; Atmospheric Emissions from Chlor-Alkali Manufacture (1971)

#### (6.4) Literature cited

- 1) Iwasaki, J.; Applied Mineralogy, Uchida Rokakuho (1943)
- 2) Air Quality Bureau, the Environmental Agency of Japan; Report on the Emission Factor of Air Pollutant from Stationary Combustion Facilities, pp.312-313 (1976)
- 3) Himi, Y.; On the Air Pollutant Emission Factors from the Stationary Sources, *J. Japan Soc. Air Pollut*, 21, 465-485 (1986)

- 4) J.J. Scheneman, W.D. High, W.E. Bye; Air Pollution Aspect of the Iron and Steel Industry, U.S. Department of Health Education and Welfare, Public Health Service (1963)
- 5) Tanabe, T. et. al. trans.; Shlaeger, Introduction to Metallurgy, Maruzen (1971)
- 6) Morinaga, T.; Aluminum Refinery, Nikkan Kogyo Shinbunsha (1968)

#### (6.5) Literature cited

- 1) Igi, S.; Natural Petroleum and Artificial Petroleum, Kyoritsu Shuppan (1952)
- 2) Iwamoto, S., Trans.; Fundamental Chemistry for Understanding Environment, Tokyo Kagaku Dojin (1989)
- 3) The Japan Petroleum Institute, ed.; Oil Refinery Process, Revised, Miyuki Shobo (1972)
- 4) Morita, Y.; Petroleum Refinery-Theory and Engineering, Shokodo (1972)
- 5) U.S. Department of Health, Education, and Welfare, Public Health Service; Atmospheric Emission from Petroleum Refineries, A Guide for measurement and Control (1960)
- 6) Himi, Y.; On the Air Pollutant Emission Factors from the Stationary Sources, *J. Japan Soc. Air Pollut*, 21, 465-485 (1986)
- 7) Nippon Sekiyu Co., Ltd.; Petroleum Handbook 1994, p.245 (1994)
- 8) The Japan Petroleum Institute, ed.; New Encyclopedia of Petroleum, p.857 (1992)

#### (6.6) References

- 1) Air Quality Bureau, Environment Agency; Motor Vehicle Environmental Pollution Control in Japan, July 1996.
- 2) The Tokyo Metropolitan Research Institute for Environmental Protection; Air Pollution by Motor Vehicle Exhaust Gas, Environmental Topic Series No.8, 1995.
- 3) Bureau of Environmental Protection Tokyo Metropolitan Government; The Report of Vehicles Total Distance Traveled, and Amounts of Pollutants Emitted from Vehicles in Tokyo (Summary Estimate) March 1996

#### (6.7) Literature cited

- 1) Yamamoto, Shinichi; Improving the Offensive Odor and related Laws, *Environmental Management* 32, 137 to 144 (1996)
- 2) Kuratani, Hidekazu; Recent Trends in Offensive Odor Complaints, *Industry and Environment*, March issue, 57 to 60 (1997)
- 3) Offensive Odor Law Research Council; Offensive Odor Control Law Handbook, Revised Edition, Gyosei (1996).
- 4) New Environmental Management Facilities Lexicography Committee; Air Pollution Prevention Equipment, Industrial Survey Council (1996)
- 5) Takahashi, Masahiro; Status of Offensive Odor Problems in Sewage Treatment Plants, *Offensive Odor Research* 25, 1 to 7 (1994)
- 6) Tanigawa, Sho; Status of Preventive Measures Against the Generation of Offensive Odors Concomitant with

Urban Waste Processing and Disposal, *Offensive Odor Research* 27, 1 to 8 (1996)

- 7) Tazawa, Shinji; Preventive Measures Against Offensive Odors in Oil Refineries, *Industrial Pollution* 29, 430 to 436 (1993)
- 8) Yoshimura, Tsunenori; Preventive Measures Against Offensive Odors in Chemical Plants, *Industrial Pollution* 29, 437 to 442 (1993)
- 9) Ohgoshi, Yoshio; Preventive Measures Against Offensive Odors in Sewage Facilities, *Industrial Pollution* 29, 443 to 455 (1993)
- 10) Ishikuro, Ryukichi; Basic Preventive Measures Against Offensive Odors and Current Reality, Ohm Co., 1997

#### (6.8) Literature cited

- 1) Himi, Yasuji; On the Air Pollutant Emission Factors from Stationary Sources, *Journal of Japan Society for Atmospheric Environment*, 21, 465-485 (1986)
- 2) U.S. Department of Health, Education and Welfare, Public Health Service ; Nationwide Inventory of Air Pollutants Emissions 1968 (1970)
- 3) Dupley, R.L.; Compilation of Air Pollutants Emission Factors, U.S. Department of Health, Education and Welfare, Public Health Service, Bureau of Disease Prevention and Environment Control, National Center for Air Pollution (1968)
- 4) R. L. Dupley (author), Japanese Fuel Association (trans.); Sources of Air Pollutants-Generation Process and Emission Factors (1971)
- 5) Mayer, M.; A Compilation of Air Pollutants Emission Factors for Combustion Process, Gasoline Evaporation, and Selected Industrial Processes, U.S. Department of Health, Education and Welfare, Public Health Service, National Center for Air Pollution (1965)
- 6) Agency of Industrial Science and Technology; Report on Survey into Soot and Dust (1961)
- 7) Tokyo Metropolitan Pollution Bureau Regulation Department; Survey into Calculations of Air Pollutant Emission Coefficients (Stationary Sources) (1973)
- 8) Kanagawa Prefecture Pollution Prevention Bureau; Kanagawa Prefecture Pollution Prevention Prescriptions, etc., Point-by-Point Explanation, Imperial Local Government Association (1972)
- 9) Himi, Yasuji; Results of Air Pollution Control Measures in Kanagawa Prefecture, *Industrial Pollution*, 16, 461-469 (1980)
- 10) Japan Thermal Energy Technology Association; Report of Survey into Pollutant Sources Caused by Photochemical Reactions, Quantity of Emissions of NO<sub>x</sub> and Hydrocarbon Species in Stationary Combustion sources, 1972 Environment Agency-Entrusted Survey (1973)
- 11) Air Quality Bureau, Environment Agency; Report of Survey Regarding Air Pollutant Emission Factors from Stationary Combustion Sources (1976)
- 12) Himi, Yasuji et. al.; Air Pollutant Emission Factors from Stationary Combustion Sources, Proceedings of The Fourth International Clean Air Congress, pp.913-918 (1977)
- 13) Himi, Yasuji; Air Pollutant Emission Factors from Stationary Combustion Sources, Extended Abstracts, Japan-China Symposium on Air Pollution, pp.173-178 (1991)

- 14) U.S. Environmental Protection Agency; *Compilation of Air Pollutant Emission Factors, Volume 1: Stationary Point And Area Sources, Fourth Edition*, pp.1.3-1, pp.4.4-15 (1985)
- 15) Himi, Yasuji; Responses to Regulation of Total Emission of NO<sub>x</sub>, *Energy Saving*, 10, (7) 10-17 (1983)
- 16) Tokyo Metropolitan Pollution Bureau Air Quality Department; *Vehicle Pollution Handbook* (1980)

## Chapter 7 Air Pollution Control Technology

### 7.1 Basic Ideas Behind Air Pollution Control

#### 7.1.1 Overview

Reducing the emission of air pollutants into the atmosphere is indispensable for air pollution control. Many technologies are applied toward this purpose and the technology for the collection and treatment of pollutants such as dust collector and exhaust gas desulfurization and denitrification equipment is spreading widely. However, these are not the only means of air pollution control. Among many other control techniques are the improvement of fuels, combustion management, energy saving and process control, as mentioned later. The basic ideas employed by these techniques toward the air pollution control described here.

#### 7.1.2 Basic considerations on air pollution control

It is sometimes possible to control the amount of air pollutant emission through rational management of the production process as follows. There are many cases where pollutants are emitted into the atmosphere, because the process management is improper, so it is desirable to check first about whether or not the production process is rational. A conventional flow sheet has depicted in its center a flow of materials starting from the raw materials and ending in the final products in the manufacturing process, but Professor Motoji Shimpo<sup>1)</sup> of Nagoya University has introduced the idea of a negative flow sheet which depicts centrally (the influencing objects) the environment. In short, it is the point of view that the process control should be considered, covering not only the flow of all the materials in the manufacturing process, but the discharging of the contaminants. The main point is that keeping the contaminant emissions at a minimum during production is the first necessity to control air pollution. This idea also exists in the pollutant release and transfer register (PRTR) which was introduced by Professor Kohei Urano of Yokohama National University. Generally, what should be considered is that the process with a larger amount of raw materials which are not put into products has a higher contaminant discharge factor, and more contaminants are discharged in a highly complicated process which incorporates multiple steps in the production process.

#### 7.1.3 Air Pollution Control Equipment

The adoption of the air pollution control equipment must be considered when it is impossible to regulate the quantity of pollutant emission to the level of air pollution emission standards even if the manufacturing process is thoroughly rationalized and the fuel and raw materials are chosen carefully based on the above idea, and when the emission of pollutants which are not regulated still could cause air pollution. Generally, it should be recognized that the air pollution control equipment such as dust collector, exhaust gas desulfurization equipment and heavy oil desulfurization equipment are all just pollutants collectors and sufficient care should be used in its employment. In other words, it is considered that the environmental pollution is induced when there are an accumulation of materials

and a polarization of many things. The collected pollutants would be dispersed by nature if left untreated, and cause environmental pollution. Therefore, changing the point of view, it should be recognized that the air pollution control equipment which is a material collection device is useful for a temporary lowering of environmental pollution, but does not exterminate the cause of environmental pollution if the contaminants it collects are dumped carelessly. Moreover, it should be considered that in the operation and management of air pollution control equipment some energy is necessary and this acquisition means is a factor in contaminant emissions.

In 1823, the alkaline industry in England which adopted the Leblanc soda process flourished, brought about damage from hydrogen chloride polluting the air. Then, taking this opportunity, the Alkali Act was established and regulation of hydrogen chloride was instituted. In 1863, this was solved by developing a control by processing the exhaust gas via washing in. It is believed to be the world's first chemical discharge control. But this was criticized as "only changing air pollution into water pollution" because it discharged the drainage into rivers<sup>3)4)</sup>. Thus, maintaining environmental safety, including a utilization method with air pollutants which were collected by an air pollution control equipment, is important and it seems that a consideration of the collected contaminant as resources is also essential. Moreover, when talking about the consideration of resources, a consideration of the material as resources which are used in the air pollution control is also important. For example, in the case of dry process exhaust gas denitrification equipment which reduces NO<sub>x</sub> in catalytic reaction with ammonia, the consideration should be agreed upon for the limitation of ammonia supply, and it is the same with the wet process exhaust gas desulfurizing equipment which uses ammonia as a neutralizer. Furthermore, it should be recognized that, when ammonium sulfur is added to fertilizer, it acidifies arable soil, and the problem of damage to environmental protection remains<sup>5)</sup>. Also, the sulfuric acid manufactured by the dry method exhaust gas desulfurization equipment promotes the use of ammonium sulfur in fertilizer.

In Japan, especially, sulfuric acid is in surplus. At this point, gypsum is imported due to a shortage and the limestone-gypsum process has a higher priority in the consideration of resources and environmental safety. Moreover, it is an excellent technique to collect air pollutants for reusing the flyash which was collected with an electrostatic dust collecting equipment as a raw material for cement, for reusing the fumes in the exhaust gas from converter which was collected with a bag filter as a raw material in iron manufacturing, or for reusing to a crude oil tank the evaporating gas which was collected through washing with kerosene in the gasoline forwarding facilities. In this way, to use an air pollution control equipment, it is necessary to sufficiently examine the benefit to environmental safety and the use of collected materials considering the restrictions on the resource and energy. Accompanying this is the necessity to consider the transportation means of by-product materials and the materials control needed for equipment operation<sup>6)</sup>.

#### 7.1.4 The establishment of an air pollution control organization

And, as for the technical activity, the establishment of an air pollution control organization centered on technical expertise is indispensable and much thought should be given as a software part of the control technology. This sometimes presses the change to the conventional view of manufacturing technique. There are examples of a thermal power plant which had to spare much of the site area for air pollution control equipment and of a coal firing



thermal power plant which had to construct exhaust gas desulfurizing equipment in the coal storage space which had been the hard core of a thermal power plant. Also, there is an electric power companies which among the many electronic engineers chemical engineering experts is appointed as director to smooth the management of the exhaust gas desulfurizing equipment. Thus, for effective management of air pollution control organization, it should be attested that air pollution control technology has been broadly influential, exceeding the conventional academic boundaries of the engineering field <sup>7)</sup>.

## 7.2 Transitions in Countermeasures to Air Pollution

### 7.2.1 Introduction

Methods to prevent air pollution include;

- (1) Separation of damaged areas and pollution sources. (aim at reduction by widening the distance)
- (2) Heighten ventilation pipes and chimneys to avoid high concentrations on ground level.
- (3) Attachment of dust collecting equipments and toxic gas treatment equipments. (automobiles: three way catalytic converter)
- (4) Change over to better quality fuel. (automobiles: low-pollution vehicles) as well as many other measures.

To take these countermeasures into definite action, the following should be implemented.

- (1) Conclude agreements (agreement on environmental pollution control, etc.) with stated goals and detailed measures.
- (2) Set and enforce limits through laws, ordinances, etc.
- (3) Create standards of guidance, guideline and guideline principle, set and enforce numerical targets. Along with other possibilities, significant results could be effected by a combination of measures.

### 7.2.2 Examples of instruction and agreements

In 1951, the Ube City Assembly gathered representatives from business, the assembly itself and academia and created a mayoral advisory organ entitled the "Smoke and Soot Prevention Committee". Under the leadership of this committee, despite the fact that coal usage rose over 79% between 1951 (monthly consumption of 85,600 t) and 1962 (monthly consumption of 153,600 t), the amount of soot and dust decreased by one third from 55.9 t per month per km to 18 t per month per km. (The city utilizes nearly 40 dust collecting equipments and uses the soot and dust to make cement admixture which has been sold over the past ten years for a total of nine hundred million yen.) The Ube measure is referred to as the "Ube Method". There are many other examples of methods through which large scale pollution-causing businesses and local government entered into agreements.

### 7.2.3 Gradual introduction of restrictions and results.

#### (1) Measures against soot and dust

When in 1932 the "Osaka Smoke and Soot Prevention Act", which restricted black smoke concentration to under Ringelman's levels 3, was promulgated it was the beginning of the introduction of restrictive measures to control air pollution. The measures adopted in this plan were to burn anthracite coal and nurture boiler engineers. Progress was curtailed by the war, etc., thereafter. After WW II, in 1955, the "Tokyo Metropolitan Soot Prevention Ordinance" was established and restrictions based on Ringelman's smoke chart were begun. Further, in 1962, the government set the "Smoke and Soot Regulation law" which introduced regulations limiting mass concentration of soot and dust emissions based on the Ringelman's smoke chart and the definitions of JISZ8808.

Based on these measures, restrictions on soot and dust were implemented in Tokyo, Kawasaki, Osaka, Kita-Kyushu, etc. However, air pollution caused by sulfur oxides continued unabated and the addition of pollution from auto exhaust gases, etc., made the situation more complicated and grave.

(2) General measures against air pollution

In order to prevent such air pollution, the "Air Pollution Control Law" was enacted in 1968 and restrictions on sulfur oxides were introduced. Also, in 1970, new types of air pollution problems including photo-chemical smog and pollution from lead as well as that from auto exhaust gas, hydrocarbon species and other hazardous substances (Cd, Cl, HCl, F, HF, Pb, NOx, etc.) inspired the setting of restrictions on these substances as well. Also, materials resulting from synthesis, resolution and other chemical treatment methods that could adversely affect the health of human beings and the environment (ammonia, etc., 26 different types) were tagged "specified substances" and measures in case of accidents were made compulsory. Additionally, structure, use and management standards were introduced for the particulate dust due to the pulverization or crushing, sorting, and accumulation, etc., of materials. Later, in 1974, the "Areawide Total Pollutant Load Control for Sulfur Oxides", and in 1986, the "Areawide Total Pollutant Load Control for Nitrogen Oxides" were designated to and enacted in each area. Also, in December of 1989, the asbestos emission restriction was introduced. Table 7.2.1 lists the various restriction measures, etc., included in the Air Pollution Control Law. In May of 1996, benzene, trichloroethylene, and tetrachloroethylene were made specified substances and came under restrictions.

Table 7.2.1 Air Pollution Control Law restriction measures

Restricted substance	Substance example	Form at occurrence source	Facility	Restriction standards	Restriction measures, etc.	
Smoke and dust	Sulfur Oxides	SO <sub>2</sub> , SO <sub>x</sub>	material combustion	smoke and soot emitting facilities	Smoke and soot emission standards (restrictions on amount, K level method by area, restriction standard on total emission)	order for improvement, direct penalties, etc.
	Soot and dust	soot, etc.	same as above	same as above	same as above (concentration restrictions based on facility type and size)	same as above
	Hazardous substances	NOx, Cd, Pb, HF, Cl <sub>2</sub> , HCl, etc.	material combustion, synthesis, decomposition, pressurization, etc.	same as above	same as above (concentration restrictions based on type of material and facility type. General restriction standards are in place for NOx.)	same as above
	Designated hazardous substances	(not yet designated)	material combustion	same as above	same as above (amount restrictions, K level method)	same as above
Dust	Designated dust	asbestos	dissection, cutting, grinding, etc.	specified dust emitting facilities	restriction standards (concentration restrictions, site limits)	order for improvement
	Common dust	cement dust, coal dust, steel dust, etc.	material pulverization, sorting, accumulation, etc.	common dust emitting facilities	structure, use and management standards	order to meet standards
Automobile exhaust gases	CO, HC, Pb, NOx, etc.	running of automobiles	specified automobiles	approval limitations (consideration of security standards)	traffic regulations, order for maintenance, etc., (according to other laws)	
Designated materials	C <sub>6</sub> H <sub>5</sub> OH (phenol), C <sub>5</sub> H <sub>5</sub> N (pyridine), etc.	accidents occurring during material synthesis and other chemical treatment	treatment methods specified facilities (not designated by government ordinance)	none	orders to take measures in case of accident	

(3) Results of measures against air pollution

A. Fig.7.2.1 presents the results of the stationary emission source measures enacted by Tokyo (sulfur dioxide, suspended dust).

B. Fig.7.2.2 presents the results of the auto exhaust gas restrictions (Carbon Monoxide).

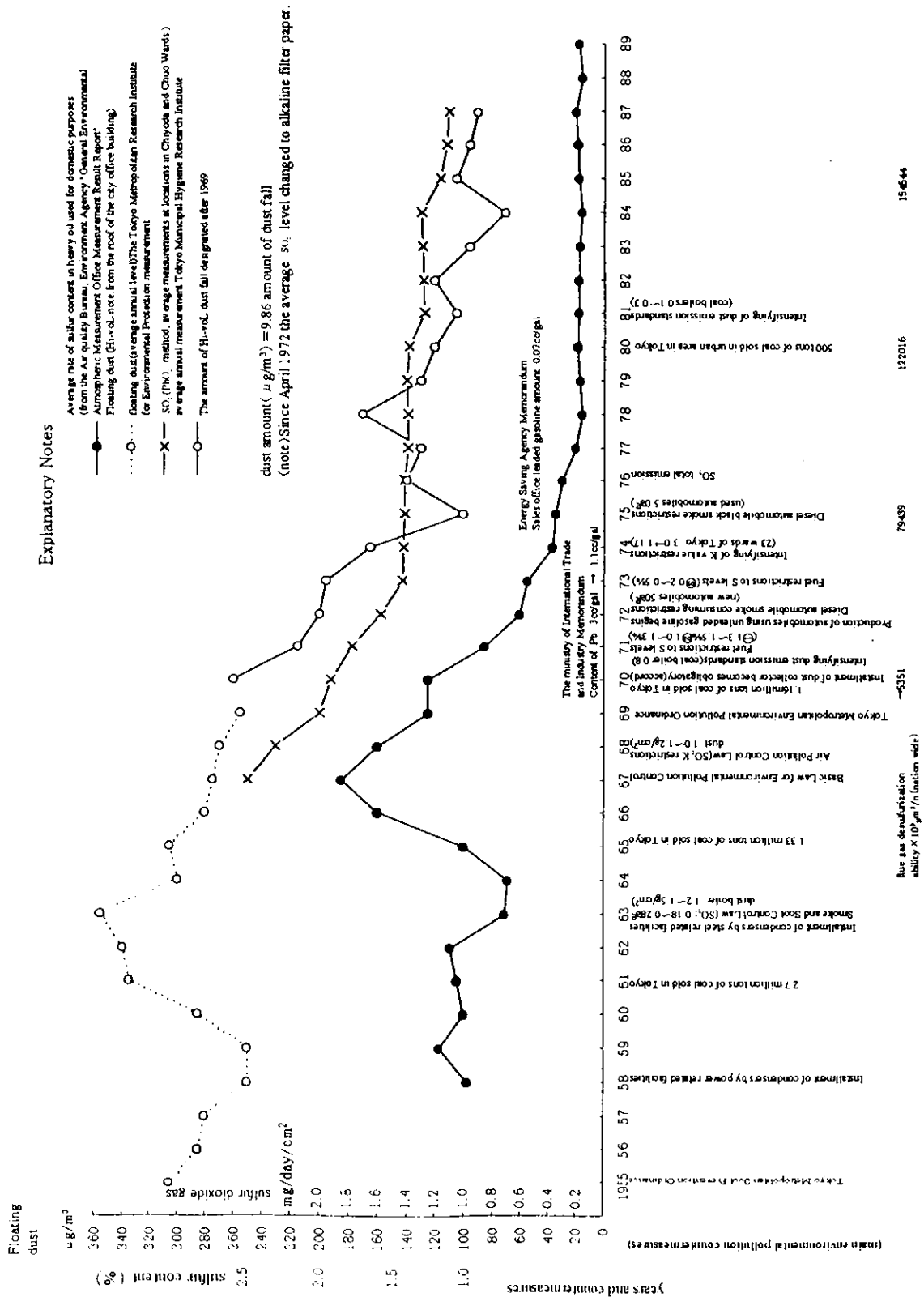


Fig. 7.2.1 Conditions of air pollution in cities over the past 35

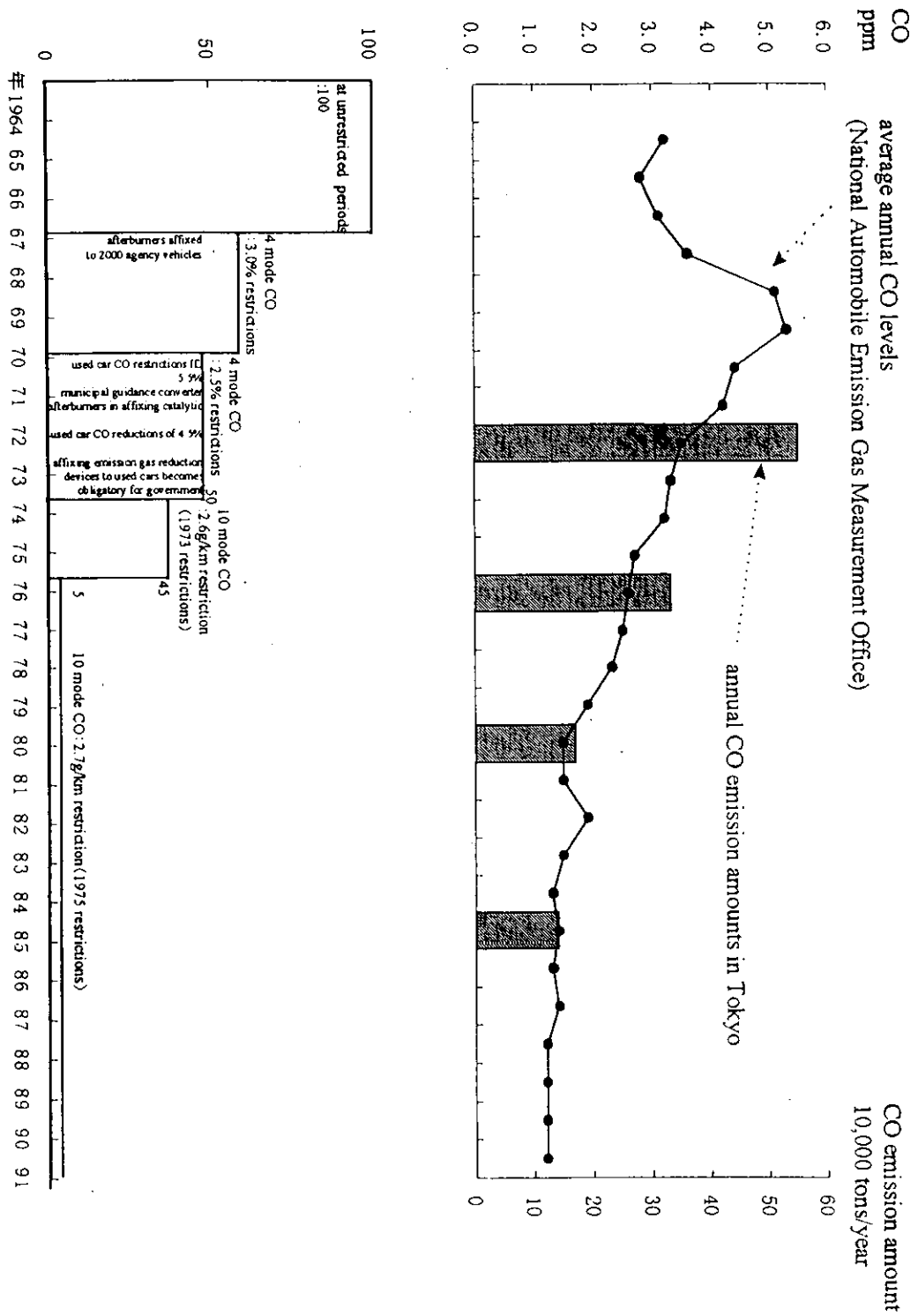


Fig. 7.2.2 Changes in the average annual CO levels and automobile

## 7.3 Better fuel countermeasure

### 7.3.1 Introduction

By the use of better fuel controls, the creation of air pollutants is reduced. Ways to convert types of fuel (changing from burning coal to burning heavy oil or gas) and the method of using the better quality fuels within the same fuel type (switching from those with high sulfur content to low sulfur content) are being adopted.

### 7.3.2 The necessity of better fuel countermeasure.

Even if emission standards are kept in urban regions with concentrations of offices and commercial buildings, because the sources of the emissions are concentrated by each building's boiler, there is high concentrate pollution.

It is difficult to install flue-gas desulfurization equipment because areas which have become crowded do not have much space and are small in scale. Therefore, in areas such as these seasonal fuel usage standards are effective.

### 7.3.3 Fuel usage standards as a Air Pollution Control Law

There are the following two types of these standards

- (1) Seasonal standards: Regulate the amount of sulfur in fuel in 14 urban areas including Sapporo and Tokyo.
- (2) Standard for area-wide total pollution load control: Regulate the facilities smaller than objective facilities for areawide total pollutant load control, in 24 areas including Tokyo and Osaka, setting standards to match areas with less than 0.3% sulfur.

### 7.3.4 The effects of using better fuel

- (1) Gaseous fuels (natural gas, LPG etc.)
  - a) Burns with little excess air, burns efficiently.
  - b) Because they do not contain sulfur or ash, they do not create sulfur oxides or soot.
  - c) Because they do not contain nitrogen they do not create Fuel NO<sub>x</sub> (however we must pay attention to Thermal NO<sub>x</sub>).
- (2) Liquid fuels (heavy oil, light oil, kerosene etc.)
  - a) Heavy oils create soot, but not as much as coal.
  - b) Because heavy oils contain sulfur and nitrogen, they create sulfur oxides and nitrogen oxides. In this case, flue-gas desulfurization and denitrification are performed.
  - c) Low-sulfur heavy oils, made by removing sulfur by desulfurization, produce few sulfur oxides and nitrogen oxides. Desulfurization can be thought of as performing denitrification at the same time. Refer to Fig.7.3.1 for the relationship between sulfur and nitrogen in heavy oils.

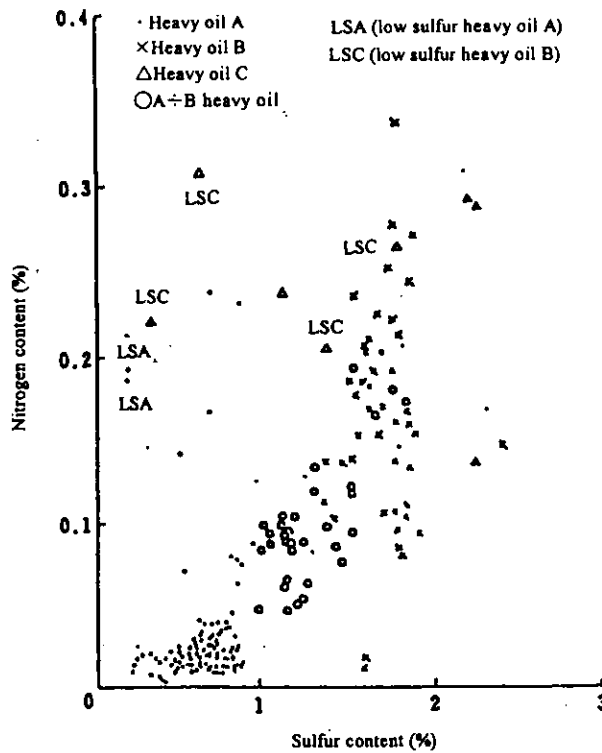


Fig.7.3.1 Relationship of sulfur and nitrogen in heavy oils.

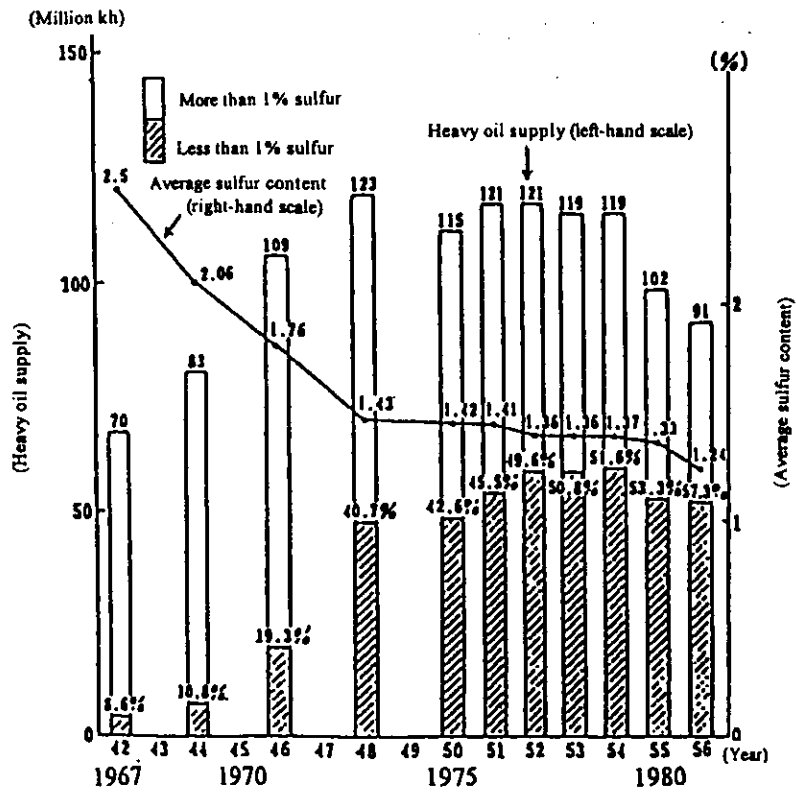
(3) Solid fuels (bituminous coal, anthracite coal, lignite, coke)

- a) Because they contain a lot of ash and sulfur, large quantities of soot and sulfur oxides are produced.
- b) Because they also contain nitrogen, they also produce nitrogen oxides.
- c) It is difficult to control burning, skill is required.
- d) Large furnaces use pulverized coal firing and fluidized combustion, and by the exhaust treatment gas, soot and dust sulfur oxides and nitrogen oxides are removed.

7.3.5 Supply of high-quality fuels.

According to statistics going back to 1967, the amount of sulfur in fuels has decreased from 2.5% in 1967 to 1.33% in 1980. Fig.7.3.2 shows the supply of heavy oils and average sulfur content by type of sulfur. Also, the current level is approximately 1.1%. This is how all regions in Japan achieved the environmental standards for sulfur dioxide in 1985.





Note: according to fuel oil federation information

Fig.7.3.2 Supply of heavy oils and average sulfur content by type of sulfur.

## 7.4 Combustion control and energy saving

### 7.4.1 The meaning of combustion control<sup>1)</sup>

Combustion control refers to the operation of combustion furnaces so that the combustion efficiency is improved by rationalizing the combustion method and equipment maintenance.

In the time when coal was the primary fuel the main purpose was for heat economy and to prevent soot, but in the time of liquid fuels the primary factor of combustion control is the quality of the design of furnaces which use automatic combustion controls.

Also, because of the photochemical smog problem, combustion control must also take into account control of NO<sub>x</sub> creation, and fuels, furnaces and combustion technology have been improved.

### 7.4.2 Trends in combustion control and energy saving in Japan

#### (1) Application of low air fuel ratio combustion

The measurement and control of oxygen concentration in the burnt gas have been energetically used to properly control the amount of excess air, achieve complete combustion and reduce the amount of heat loss by exhaust gas. That is, reducing the amount of heat retained by excess air by reducing the amount of air in the furnace. In principle, as shown in Fig. 7.4.1<sup>2)</sup>, heat loss from unburned CO and H<sub>2</sub> is caused by insufficient air, and heat loss is also required to heat the excess air, so by controlling burning to the minimum amount of oxygen the total heat loss is minimized. However, currently the oxygen control target is higher than the minimum levels because of the problem of intruding air.

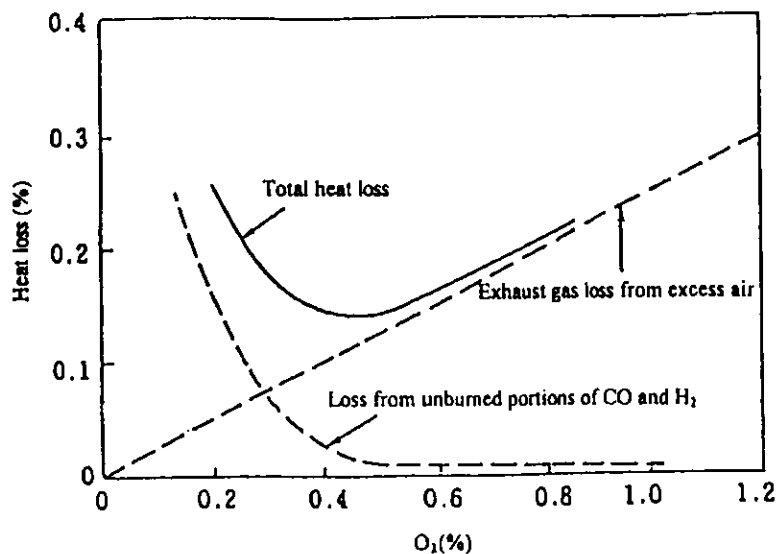


Fig. 7.4.1 Relationship between O<sub>2</sub> concentration and total heat loss

Table 7.4.1<sup>3)</sup> shows the standard air fuel ratio for various furnaces. By targeting these various levels we should burn using low air fuel ratio combustion to rationalize energy saving to the extent technically possible.

Table 7.4.1 Standard air fuel ratio

(1) Boiler

Section	Load ratio (%)	Standard air fuel ratio			
		Solid fuel	Liquid fuel	Gaseous fuel	Blast furnace gas and other by product gasses
Electrical power industry use	75~100	1.2~1.3	1.05~1.1	1.05~1.1	1.2
Other	Amount of steam in excess of 30 t per hour	1.2~1.3	1.1~1.2	1.1~1.2	1.3
	Amount of steam more than 10 t but less than 30 t per hour	—	1.2~1.3	1.2~1.3	—
	Amount of steam less than 10 t per hour	—	1.3	1.3	—

(2) Industrial furnaces

Section	Standard air fuel ratio
Metal casted melting furnaces	1.3
Continuous steel slab heating furnaces	1.25
Metal heating furnaces other than continuous steel slab heating furnaces	1.3
Continuous heat treatment furnaces	1.3
Gas generating furnaces and gas heating furnaces	1.4
Fuel oil heating furnaces	1.4
Thermal resolution furnaces and improved quality furnaces	1.3
Cement kiln	1.3
Aluminum and lime calcination furnaces	1.4
Continuous glass melting furnaces	1.3

By using low air fuel ratio combustion heat loss by exhaust gas is reduced and we can reduce the amount of fuel consumed. Furthermore, with less combustion air and less exhaust gas, forced draft fans (FDF) and induced draft fan (IDF) are used less, so operation costs are reduced.

In addition, low air fuel ratio combustion is effective for the following measures, and are implemented as part of combustion control.

① NO<sub>x</sub>

Because of the low concentration of oxygen in the combustion the creation of NO<sub>x</sub> can be controlled.

② Low temperature corrosion

With low air fuel ratio combustion, by controlling the oxidation reaction from SO<sub>2</sub> to SO<sub>3</sub>, the creation of SO<sub>3</sub>, which causes low temperature corrosion, can be relatively reduced.

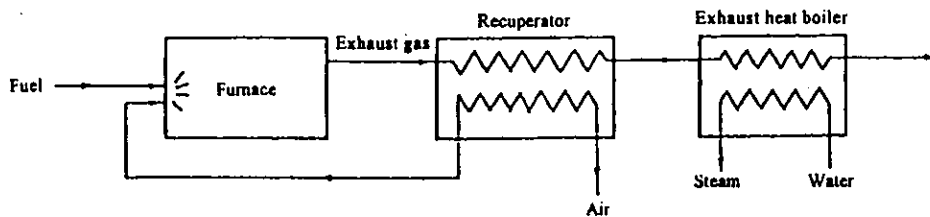
③ Acid smat

By burning fuel the soot created becomes a nucleus, and the sulfur in the fuel is absorbed in the sulfuric acid soot through burning, and frost forms at a temperature close to the fuel gas dew point, which is called acid smat. If low air fuel ratio combustion is used the creation of SO<sub>3</sub> and sulfuric acid are controlled, creating less acid smat.

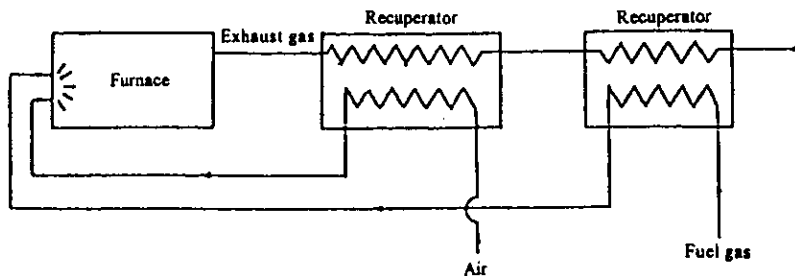
(2) Preheating of combustion air and gaseous fuel .

An effective method for addressing energy saving is close at hand at the furnace; preheating the unheated combustion air or fuel using waste heat. Currently however, mostly combustion air is preheated. Preheating of fuel should be undertaken upon sufficient consideration of thermal decomposition and safety, especially liquid fuels are not preheated for energy saving, except for being preheated to the most suitable evaporation temperature. Accordingly, preheating fuel for energy saving is limited to gaseous fuels.

Preheating air or gaseous fuels uses a recuperator (exhaust heat recovery heat exchanger). Recuperators are made from metal or tile (ceramic), the heat from the exhaust gas passes through a partition and is transferred to the air or gaseous fuel. An example of the arrangement of heat recovery by a recuperator is shown in Fig.7.4.2<sup>4)</sup>.



(a) When increasing creation of steam in the exhaust boiler by preheating the combustion air



(b) When preheating both fuel and air

Fig.7.4.2 Sample arrangement of exhaust heat recovery with a recuperator

(3) Recent developments in energy saving

Technology to make a large impact on energy saving in industrial furnaces area, a regenerative burner system has been developed<sup>5)</sup>. This technology recovers more than 80% of exhaust heat in industrial heating furnaces and is very effective for energy saving, enabling an almost 50% fuel reduction. Also, the low NO<sub>x</sub> combustion technology, FDI (Fuel Direct Injection) combustion has been developed which can sufficiently use highly pre-heated air.

In the thermal electric power area, efforts are being made to increase the output scale and improve thermal efficiency, and especially for improving heating efficiency is important to control the amount of CO<sub>2</sub> emission output in addition to lost heat. When burning fuel in compressed air combustion gas is created, and this expansive power is used to turn the gas turbine which creates electrical power, and the remaining heat from the gas turbine is

used to turn a steam turbine. This combined cycle power generation is said to be 48-49%, more efficient than traditional thermal electric power generation. Also, in the case of burning coal, we are trying to introduce new technology for pressurized fluid boiler and compound coal-gas electric generation.

#### 7.4.3 Items to mention

Using the second oil shock as an opportunity, Japan's untiring efforts regarding energy saving continue, but recently more efforts are required to improve thermal efficiency with regard to CO<sub>2</sub> countermeasures. Producing a combined form of second generation energy which has at least 2 types of useful energy from one source, and the adoption of a cogeneration system to supply energy is one option.

In developing countries in general the average thermal efficiency is about 30%, so there is much room for improving utilization of lost energy. These countries should first implement measures for low-air fuel ratio combustion and exhaust heat recovery. By doing this they will be able to significantly reduce energy lessened at the same time energy costs.

## 7.5 Process Management

### 7.5.1 Overview

It is important to decrease the amount of pollutant emission at the emission source in order to control air pollution. Thus, control equipment is installed and it collects contaminants, reducing the pollutant emission volume through combustion management. In the same way, it is possible to reduce the pollutant emission volume by adopting a rational production process control. Because many kinds of production processes exist, examples of air pollution countermeasures through process control follow.

### 7.5.2 Method through Cleaning of a Workplace

It would seem that such an example as that of the 1960's would not exist in Japan at present, however, in the 1960's, a small-scale factory ran a process where 1,400 tons of a solid raw material were pulverized, in every month scattering fine particles in the neighborhood and it became a problem when the residents complained. Fine particles from the factory's crusher deposited on the floor and scattered with a concentration in the factory's atmosphere at 50-500 mg/m<sup>3</sup> near the crusher, and lowering the visibility in the factory. In this event, there was an increase in visibility by the cleaning of the deposited dust progressed. Cleaning seems to be important in the case of process control.

Many dumptrucks are taking on the haulage of earth and sand in land reclamation sites to be used for housing, land improvement sites and earth and sand excavation sites, and the environmental impact is remarkable. Then, the earth and sand adhering to the tires of these trucks during transportation are causing a scattering of dust, raising complaints along their routes. In addition, earth and sand falling onto the road from the truck beds are also scattering dust. Therefore, the dumptrucks which are departing a site are put into a shallow pool where the tires are thoroughly washed, and there are cases where efforts are being made to cover the load to prevent earth and sand from falling onto the road.

### 7.5.3 Method through Management of Raw Materials and Fuels Used during the Process

Sometimes, the clay used as a raw material at glassworks contains some fluorides which causes hydrogen fluoride emission into the atmosphere from the glass melting furnace. Therefore, the quality of the clay is controlled and clay with little fluorine content is chosen as a raw material, which should reduce the volume of hydrogen fluoride emission.

Moreover, the organic solvent which has been used to clean the painting devices after painting ship hulls in a large dockyard was thrown away. Or, there is the example where, because the amount of solvent used was more than really necessary, the amount of solvent used was decreased, thereby reducing the volume which evaporated into the air during the process.

There also is the example where the sulfur contained in the coal is controlled, by mixing of the different kind

of coal and hold the electrical resistivity of dust to the appropriate level, in order to keep the high dust collecting efficiency of the electrostatic precipitator at a high level.

#### 7.5.4 Strengthening of the Process Maintenance and Management

There are many couplers, flanges and bends as well as many control valves in the pipe system which transports volatile organic compounds. Also, pumps and compressors are necessary to transport these compounds, too, and the packing material for these connections is indispensable. However, in some cases, the volatile organic compounds evaporate through this packing material.

For example, there is data showing that an average of 0.15 g/h of hydrocarbons leaks from the pumps of a naphtha transportation pipe system and that 0.031 g/h leaks from the valves of a hydrocarbon transport system <sup>1)</sup>.

Then, it is said that this quantity of evaporation is related with the maintenance and management of these facilities <sup>2)</sup>. It is especially desirable to strengthen the maintenance and management of the packing material in these processes in order to control air pollution.

#### 7.5.5 Improving Processes

At a subcontractor factory of a semiconductor maker, a metal pieces are put in many wide-mouth beakers and concentrated nitric acid was added to each beaker and heated in order to wash. A great deal of irritative nitric acid gas was discharged from beakers and leak from the building and caused complaints. The wide-mouth beaker was replaced by a flask with a return current condenser attached, or with a conical beaker with a clock plate covering the mouth, effectively canceling the problem of condensing the cooled nitric acid vapor and nitric acid consumption decreased, too <sup>3)</sup>.

At large paint facilities, the swing angle of the paint sprayer was maintained irrespective of the size of metal part to be painted. Thus, much paint was wasted when painting small metal parts, and more of the organic solvent in the paint vaporized than necessary. Therefore, a process control system was instituted that used a light sensor to detect the size of the metal part, controlling the swing angle accordingly, and this decreased the quantity of paint used and an organic solvent that vaporized. If the spray paint process using organic solvent-based materials is changed into one using water- or powder-based paint, then the vaporization of the organic solvent will decrease <sup>4)</sup>.

#### 7.5.6 Rationalizing Processes Through Combination

There was a case where a considerable amount of black smoke was discharged because, of the two adjacent boilers, one boiler was operating in an underloaded condition and the other was in an overloaded condition. Thus, the condition of the steam supply system of the two boilers was communicated by setting of connection steam pipe between two boilers and the load was controlled, which improved this situation.

In the manufacturing process of sesame oil, there were complaints of an offensive odor. This was dealt with successfully by changing the manufacturing process to use the exhaust from sesame oil manufacturing process as

combustion air for the boiler at the factory, which canceled the complaints. This situation is similar to the example of the spray paint process where the problem was the exhaust from the paint booth, which contained organic solvent, at the car manufacturing plant<sup>4)</sup>. Considering the process and plant location, the organic solvent exhaust can be used for energy if it is burned at a electric thermal power plant, making the amount of vaporization nil.

Considering the location of a blast furnace in steel mill and cement kiln in cement factory, if both facilities can construct near, it would be easy to supply the blast furnace slag as water dross in order to mix cement clinker. And blast furnace cement can manufacture double amount of cement clinker burned by rotary kiln in cement factory. Thus, the increased cement production is attempted and the basic burning fuel unit can be reduced.



## 7.6 Dust Collector

### 7.6.1 Introduction

Dust collecting is the operation which collects and separates the particles contained in processing gases. Generally, the dust collector is used in the last step of the production process and it is called an air pollution control equipment. However, it is used in various fields with divergent purposes such as preservation of the work environment, purifying exhaust gas, recovery of useful particles and improving the purity of processes. Generally, fine particles are generated during mechanical operations such as pulverizing in addition to soot particles occurring during the combustion process, and these are generically called "dust". Here, recent techniques for collecting the dust in exhaust gas are reviewed.

### 7.6.2 Changes in the dust collecting techniques in Japan

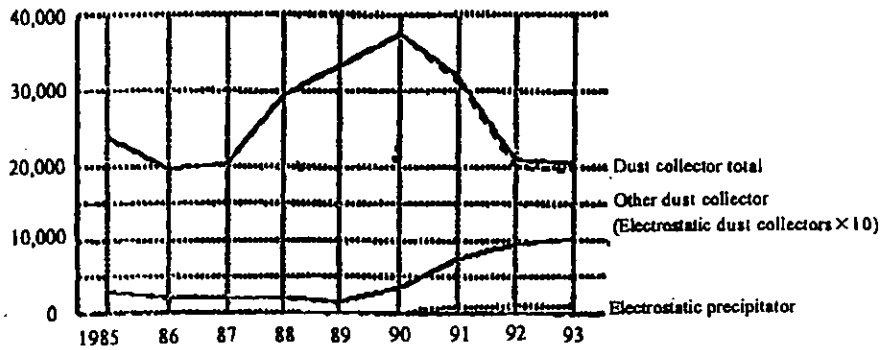
Dust collecting technology was mainly developed after the war, and, at present, the technology enables the separation and removal of minute particles to the sub-micron size. Air pollution control became regarded as highly important with the soot and dust emission standard strengthened time and again, and dust collecting techniques improved in response to this. There are limitations in dust collecting technology such as the theoretical possibility for separation and collecting of particles, especially as to particle size, and the dust collecting efficiency<sup>1)2)</sup>. For example, in the past, the emission standard for dust could be cleared with scrubbing dust collector such as scrubbers and cyclones which are centrifugal dust collectors, but, with today's severe standard, the situation has reached the point where highly efficient equipments such as filter dust separator like bag filters and electrostatic precipitator (ESP) must be utilized. Moreover, in recent years, with the high temperature/high pressure technology aiming for energy saving through the use of waste heat, new high temperature dust collecting equipments such as ceramic filters and packed-bag filters has started to appear.

### 7.6.3 Classification of dust collector

The dust collector uses one or several methods at the same time such as gravity, inertial force, thermal power, diffusion power, electric power to separate dust from inside exhaust gas then collect it in dust hoppers. The main devices for dust collecting are classified as (1) gravity dust collectors, (2) inertial dust collectors, (3) centrifugal dust collectors (cyclones), (4) scrubbing dust collector, (5) filter dust collectors (bag filters) and (6) electrostatic precipitator<sup>3)</sup>. Table 7.6.1 shows the general characteristics of these main devices, with (1) to (3) having the purposes of preprocessing the high concentration dust (primary treatment) and separating and collecting from large mists, and the purpose of (4) to (6) is to reach the emission standard. Fig.7.6.1 shows the change in the number of dust collecting units produced in Japan, of which the total number reflects the business conditions. Since hitting the peak in 1990, the number of units has been declining except that among the several thousand units of ESP and other dust collector, the bag filters have been on an increasing trend.

Table 7.6.1 The practical-use range of dust collector

Name	Type	Particle size ( $\mu$ m)	Pressure loss (mmH <sub>2</sub> O)	Collection efficiency (%)	Equipment cost	Operating cost
Gravity dust collector	Baffle chamber	1,000-50	10~15	40-60	Small	Small
Inertial dust collector	Looper	100~10	30~70	50~70	Small	Small
Centrifugal dust Collector	Cyclone	100~3	50~150	85~95	Medium	Medium
Scrubbing dust collector	Venturi scrubber	100~0.1	300~900	80~95	Medium	High
Sonic precipitator		100~0.1	60~100	80~95	Medium or higher	Medium
filter dust separator	Bag filter	20~0.1	100~200	90~99	Medium or higher	Medium or higher
Electrostatic precipitator		20~0.05	10~20	90~99.9	High	Small to medium



Source: The Ministry of International Trade and Industry: "The Machine statistic yearbook"

Fig. 7.6.1 Production of dust collector (Units)

#### 7.6.4 Development of new technology

##### (1) Filter dust separation technology

###### ① Ceramic filters<sup>9)</sup>

The base materials for ceramic filters are silica, alumina, zirconia and silicon carbide, because they have to collect dust in high temperature/high pressure exhaust gases that range in temperature from 300 to 1,300°C and at pressures from several atmospheric pressures (ATM) to 10 ATM and above. The shape is that of a porous cylinder made from woven or non-woven materials, or the cross flow as shown in Fig. 7.6.2 which can cover a very large filtration area. The advantage with this equipment is that it is compact, but the negative points include increased loss of pressure, due to the dust, the life cycle, and the shaking-off characteristic of dust.

###### ② Packed bed filters

The particle collection mechanism operates by thermodynamics accompanied by a temperature difference between the high temperature exhaust gas and the traveling particles in the packed bed, in addition to inertial force, diffusion, power interruption and gravity. The equipment which was developed for high temperature/high pressure dust collecting uses materials such as sand, gravel and ceramics several millimeters in size and is adaptable to high temperature exhaust gases of about 500°C. The dust collection efficiency is about 93% when the dust size is about

1  $\mu$  m and the pressure loss is at about the same level with the conventional bag filter.

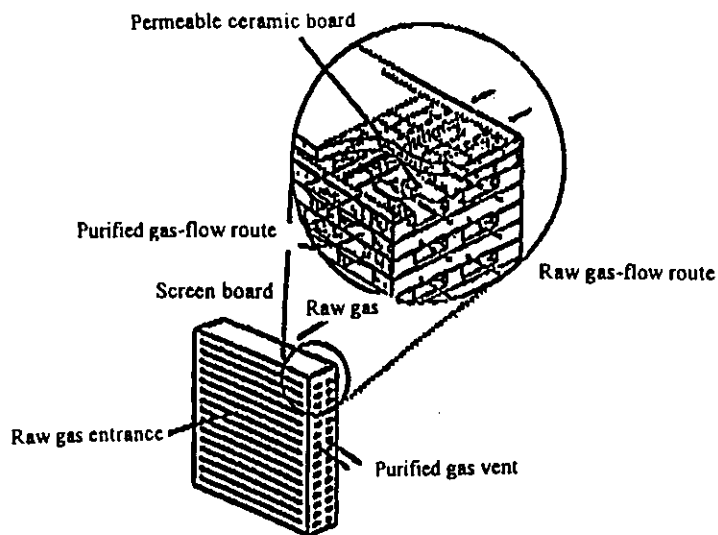


Fig.7.6.2 Ceramic cross flow filter

### ③ Electret filters <sup>10)</sup>

The mechanical particle collection mechanism of previous filters was not effective in collecting particles of around 0.1  $\mu$  m. The electret filter is made of fiber with a semi permanently polarized electric charge. In addition to the mechanical particle collector, the electret filter allows the induction force to work on un electrified particles, the electric force (coulomb force) and the induction force work on electrified particles. Therefore, particles of around the 0.1  $\mu$  m range are efficiently collected, and particles of other sizes can be collected, too, at a high degree of efficiency with the same pressure loss as that of the conventional bag filter. The electret filter is an energy-saving filter which collects particles with a high efficiency and low pressure loss.

## (2) Electrostatic precipitation technology

### ① Pulse-charge type

A reverse corona and a reverse ionization phenomenon occur in the collection of highly electrical resistant dust such as that from coal combustion and this becomes a problem with electrostatic precipitator, which causes a remarkable decline in the collection efficiency, one piece of equipment developed to counter this problem was the pulse electric charge type of electrostatic precipitator (ESP). The conventional ESP uses direct current voltage but this equipment utilizes pulse high voltage. With the pulse electric charge, the distribution of the corona electric current can be established uniformly over a wide area without a drop in the applied voltage through adjusting the pulse frequency. Energy saving can be expected from lower power consumption and stable dust collecting, and it is possible to suitably operate because the corona electric current does not experience a reverse corona due to highly electrical resistant dust.

② Moving electrode type

As shown in Fig.7.6.3, moving the dust collecting electrode continuously to keep the dust collector's electrode surface clean minimizes the electric field formed by the layer of dust adhering to the electrode and inhibits reverse ionization. The electrode moves slowly downward, and the adhering dust is detached by a revolving brush in the non-electrified area in the lower part, keeping the electrode clean.

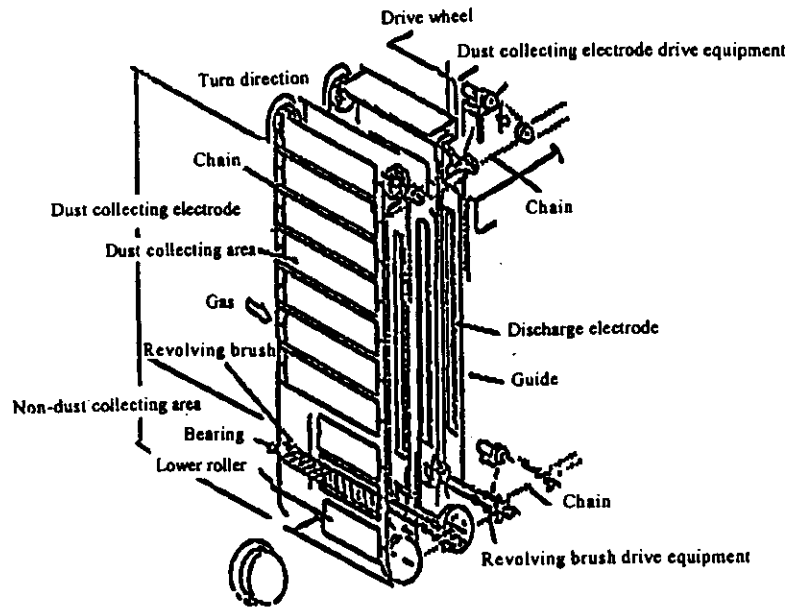


Fig.7.6.3 Structure of the moving electrode-type electrostatic precipitator

③ The high current velocity/wet process type <sup>11)</sup>

This one narrows the interval between the dust collecting electrodes facing each other across the discharge electrodes as shown in Fig.7.6.4, quickening the flow velocity of the raw gas and realizing a compact size for the equipment. The dust collecting area is reduced substantially by almost 1/2 which increases the base raw gas flow velocity more than twice that of the past (2-3 m/s) and the discharge electric current density more than several times that of the past (0.3 mA/m<sup>2</sup>).

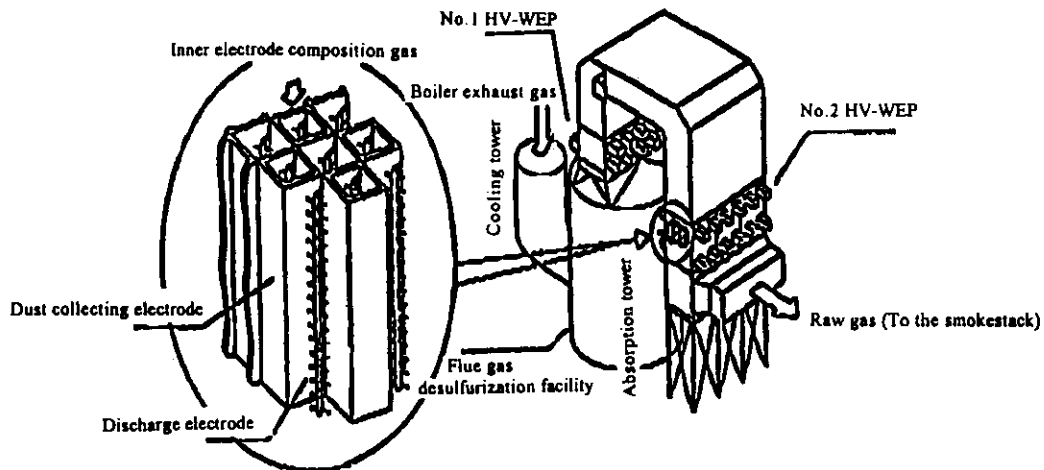


Fig.7.6.4 Outline of the high current velocity/wet process type electrostatic precipitator

### 7.6.5 Conclusion

Table 7.6.1 mentioned above compares the characteristics of the various dust collecting equipment. For a decision to be made on equipment selection, sufficient consideration is necessary involving the properties of the soot and dust emissions, the characteristics of the equipment, the equipment cost and cost performance. As new dust collecting technology advances steadily, there are many more than those mentioned above, please consult the reference literature and the related-industry standard given here.

Japan's dust collecting technology has reached a high technical level if it can be seen worldwide, but development of new technology in the future is important, too, and, while aiming for superior performance, precipitate technology should be expected to be energy-saving and resource-saving. Also, from the viewpoint of preserving the environment, it is necessary to examine together the reduction of the volume and the reusing of recycle from the enormous amount of dust collected.

## 7.7 Flue Gas Desulfurization Equipment and By-Products

In Japan, the rapid popularization of flue gas desulfurization equipment began in the 1970's. Environment Agency calculations in 1993 showed that 2,140 such units were in operation, processing a total raw gas flow of 217 million m<sup>3</sup>/h.

Flue gas desulfurization is categorized into two separate procedures, the wet method and the dry process. The majority of equipment which is currently in operation utilizes the wet method. The desulfurization ratio of the wet method is effective. Stable desulfurization results are obtainable even in conditions of load fluctuation.

Technically it is a well established method. The lime gypsum method is the main technique used for large capacity power plant boilers, such as coal fired thermal power stations utilized by electric companies. For independent power plant boilers used for general industrial purposes, the magnesium hydroxide method is widely used.

Table 7.7.1 illustrates the different types of flue gas desulfurization methods in use or under development and their related by-products.

The following sections describe major desulfurization processes, their unique characteristics and their by-products.

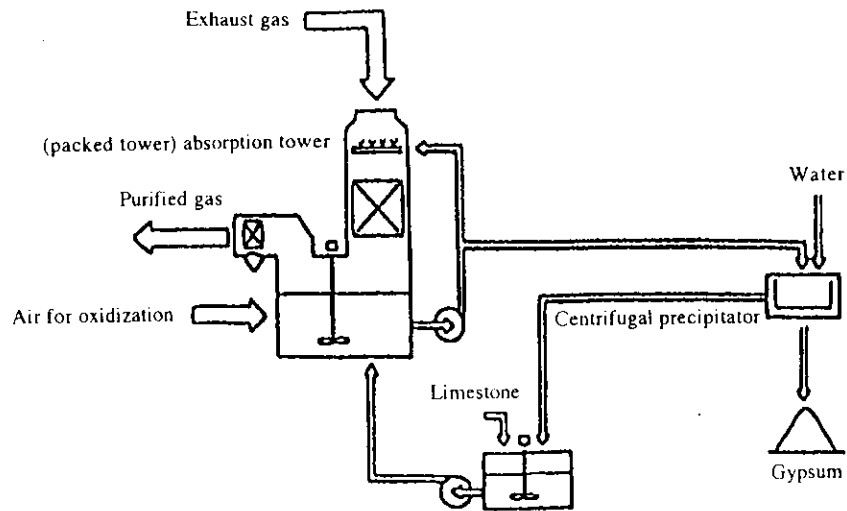
Table 7.7.1 Desulfurization Process and By-products

Process	Absorbing Agent	By-products	Usage
Lime gypsum Method	Limestone Slaked Lime	Gypsum	Gypsum Board Cement
Magnesium Hydroxide Method	Magnesium Hydroxide	Magnesium Sulfate	Discharge
Ammonia Method	Ammonia	Ammonium Sulfate	Fertilizer
Electron beam Method	Ammonia	Ammonium Sulfate	Fertilizer
Sodium Method	Sodium Hydroxide	Sulfur Soda Mirabilite	Pulp Dispersion Agent Chemicals
Simple Desulfurization Methods	Limestone, Slaked Lime	Gypsum + Coal Ash	Soil Melioration Agent Roadbed Agent

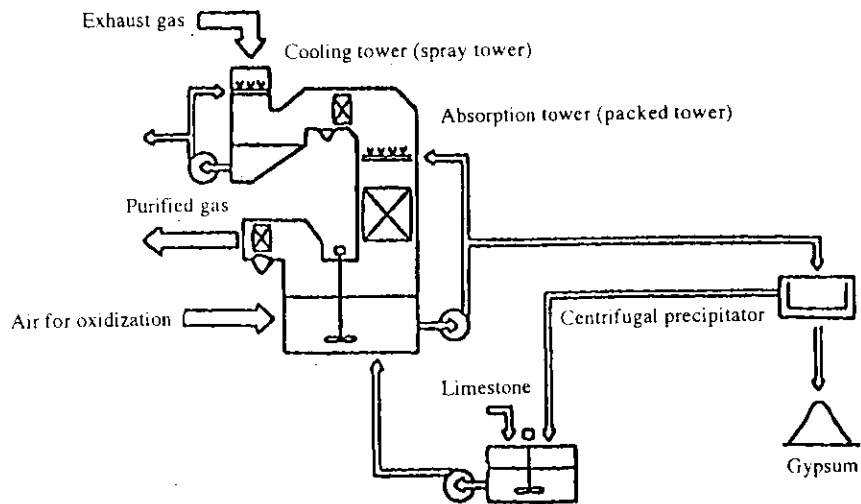
### 7.7.1 Various Processes of Desulfurization

#### (1) Lime Gypsum Process

Limestone and calcium hydroxide (slaked lime) particles, in the form of a slurry liquid, absorb SO<sub>2</sub>, creating a gypsum by-product. During the initial stages, there was scaling trouble as a result of gypsum adhering to the inner surface of the absorption tower. This was due to the low solubility of limestone and slaked lime. This problem has nearly been solved and this method has become the current mainstream of large-scale flue gas desulfurization equipment. The basic process consists of cooling the exhaust gas and absorbing SO<sub>2</sub> within the absorption tower. The created calcium sulfite is aerated in the oxidizer and the by-product is then retrieved as gypsum (2 tower type). In recent years, many systems utilize a method in which cooling, absorption, and oxidation is all performed in one tower. As illustrated in Fig. 7.7.1, the cooling tower can also be set up separately depending on the circumstances.



(a) Absorption oxidization one tower method (soot mixture one tower method)



(b) Cooling tower method (soot separation two tower method)

Fig.7.7.1 Standard Process for Lime Gypsum Method <sup>2)</sup>

As this procedure, which utilizes only one tower, immediately oxidizes the absorbed  $\text{SO}_2$  within the absorption tower, turning it into gypsum, the partial pressure level of the  $\text{SO}_2$  is reduced. The absorbed  $\text{SO}_2$  is reduced desorption. Even if the pH level of the absorption liquid falls into a lower domain of around 4.5-5, the absorption reaction of the  $\text{SO}_2$  still continues to progress. Also, the advantage of this one tower system is that it produces a smaller amount of the substance responsible for COD (Chemical Oxygen Demand) within the wastewater. This characteristic allows for a reduction the equipment for wastewater treatment. The purity of the gypsum, in comparison with methods which establish two separate towers for the cooling unit and oxidizer, declines slightly. However, the cost of the facility itself is reduced by 50-60% over the cost of conventional methods.

The construction of a separate cooling tower is determined by considering such factors as the properties of the exhaust gas and the purity level of the recovered gypsum. However, in the event that a separate cooling tower is built, the purity of the gypsum is over 95% and the dust collection efficiency is over 90%. Therefore from the standpoint of recovering a gypsum by-product this method is more advantageous. On the other hand, capital costs is higher and there is an increase in the amount of wastewater and sludge.

For absorption devices, packed towers or spray towers of a device with little pressure loss are frequently used. Moretana-tower (perforated plate-type tower) and jet bubbling devices are also utilized (Fig.7.7.2).

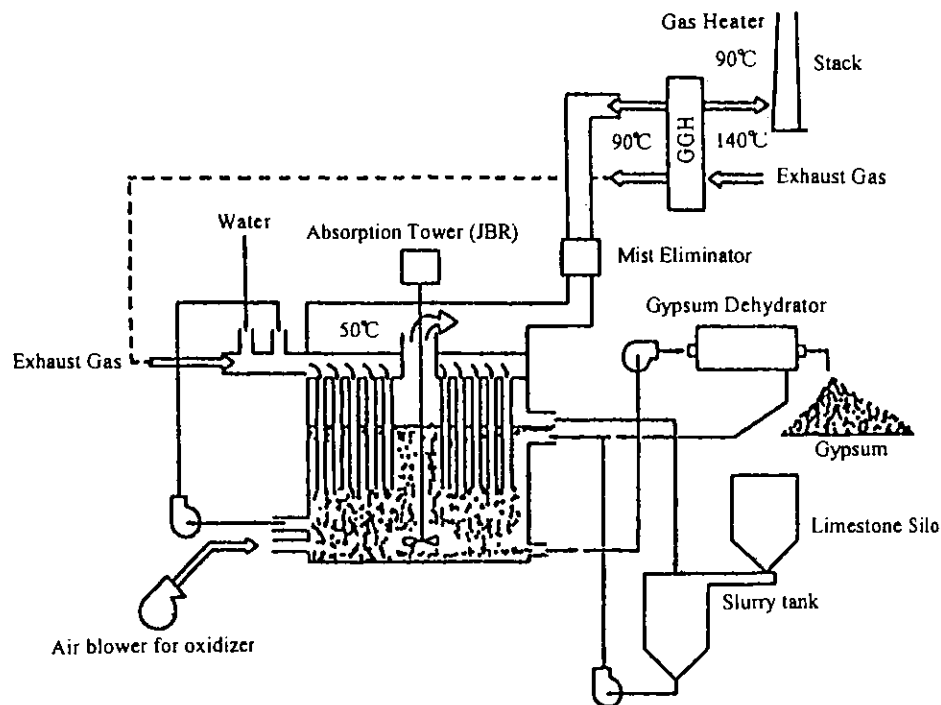


Fig.7.7.2 Jet Bubbling Reactor <sup>3)</sup>

## (2) Magnesium Hydroxide Process

The magnesium hydroxide absorbing agent, in the form of slurry, reacts with the  $SO_2$  and creates magnesium sulfite. Furthermore, once oxidized, it changes into magnesium sulfate which is harmless and highly soluble with water. This makes it possible to dispose of the drain-off or effluent. In comparison with sodium hydroxide, magnesium hydroxide is less harmful to the human body and a comparatively cheaper product to use. It is widely used in small to medium sized flue gas processors ranging in capacity of about 5~400,000  $m^3/h$ . Set up of the systems are prevalent especially in coastal areas where the absorbing agent can be drained. In Japan, there is a growing influence to make this a popular method alongside that of the lime gypsum method. The processing equipment consists of a system which unifies both the absorption tower and oxidizer into one unit. Furthermore, another method which incorporates a stack into this unit is also being adopted (Fig.7.7.3).



### (3) Sodium Scrubbing Process

This process utilizes sodium hydroxide or sodium carbonates as the absorbing agent. These substances are both highly soluble and can be used as an aqueous solution. There is no worry in regards to scaling and the both have a high absorption rate. Much of its achievements have been made in use with small scale flue gas desulfurization devices of less than 20,000 m<sup>3</sup>/h. In particular, its sodium sulfite and sodium sulfate by-products are used largely in paper and pulp factories.

### (4) Ammonia Scrubbing Process

#### (4-1) Ammonium hydroxide process

This process utilizes an aqueous agent consisting of ammonium hydroxide and absorbs the SO<sub>2</sub> while producing an ammonium sulfate by-product. In order to use the newly created ammonium sulfate aqueous agent as fertilizer, it must first be solidified. In order to do this a vast amount of energy is required. With the exception of facilities which use an excessive amount of ammonia, nearly no new facilities for this process have not built in recent years.

#### (4-2) Desulfurization by electron beam irradiation process

This desulfurization and denitrification process cools the exhaust gas down to 70~90°C. Ammonia is then injected into the gas, at the same time being irradiated by an electron beam. SO<sub>2</sub> and NO<sub>x</sub> found within the exhaust gas is used as fertilizer for the ammonium sulfate and ammonium nitrate, after which it is recovered by a dust collector. This process was developed in Japan. Substantiative testing is being performed in the US and Germany but it has yet to be used for practical application in the field. The unique characteristic of this process is that a fertilizer by-product is produced and there is no waste or wastewater. In the future a large-scale electron beam irradiation device will be manufactured. If a reduction in the system's energy consumption can be achieved, there may be high expectation placed in this method from the standpoint of its use of resources such as fertilizer.

### (5) Dry process, Semi dry process

While its rate of desulfurization is somewhat lower, the dry process and semi dry process was developed as a low cost simple process for desulfurization. There are two process: ① absorption of the SO<sub>2</sub> by limestone infused into a furnace heated to 900~1,200°C ② slaked lime slurry is sprayed into the flue where the SO<sub>2</sub> is absorbed and the by-product is collected along with the dust by a dust collector. Both of these process include such methods as spraying water after desulfurization or recirculate the dust collected by dust collecting equipments, substances which did not react to the absorbing agent, as absorber (Fig.7.7.4). While the equipment and facilities are relatively simple, reaction efficiency is low. There are also not many uses for the by-product produced. Also disposal is limited to certain areas.

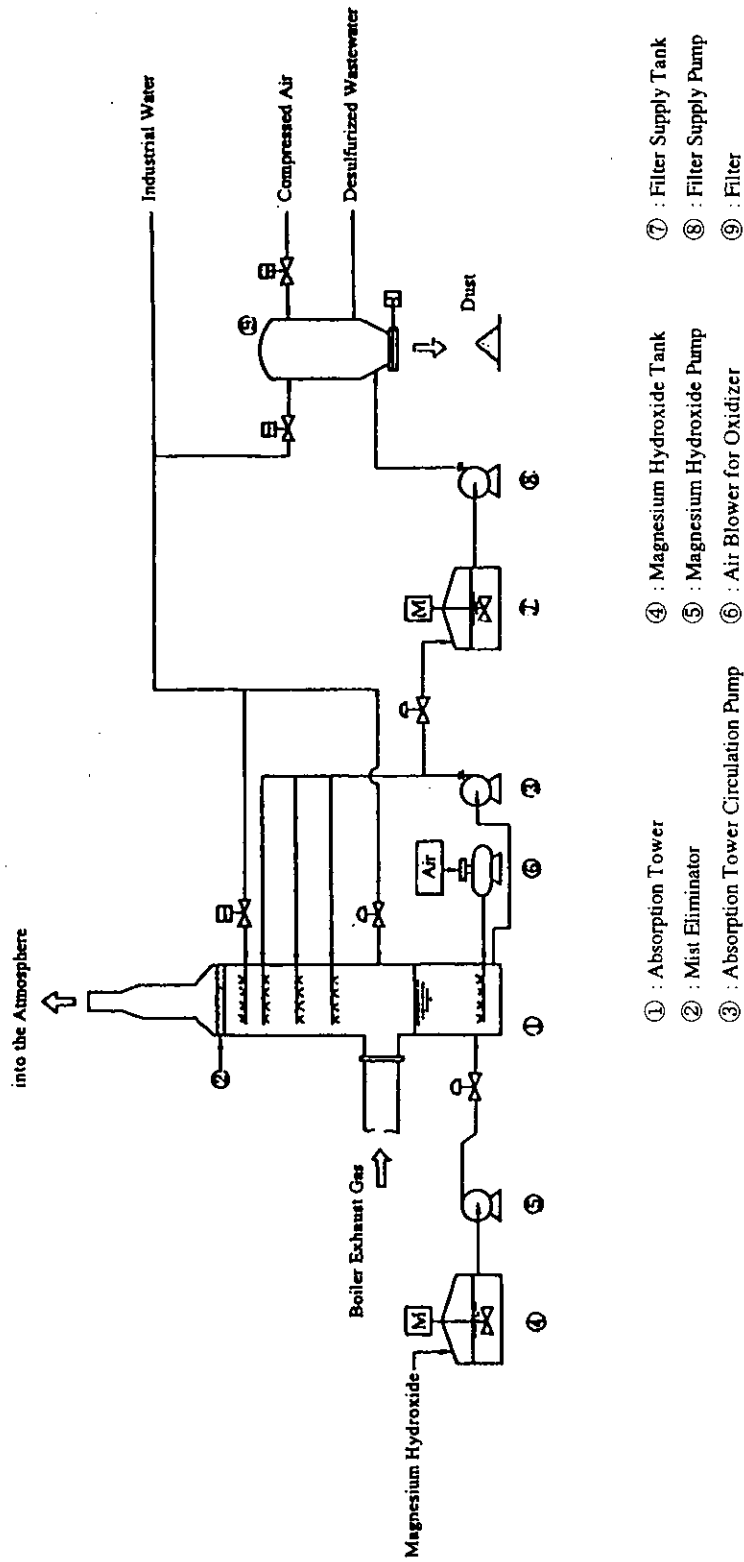
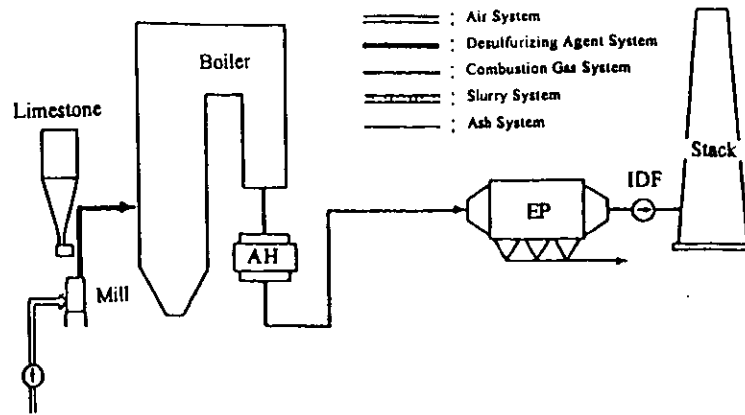
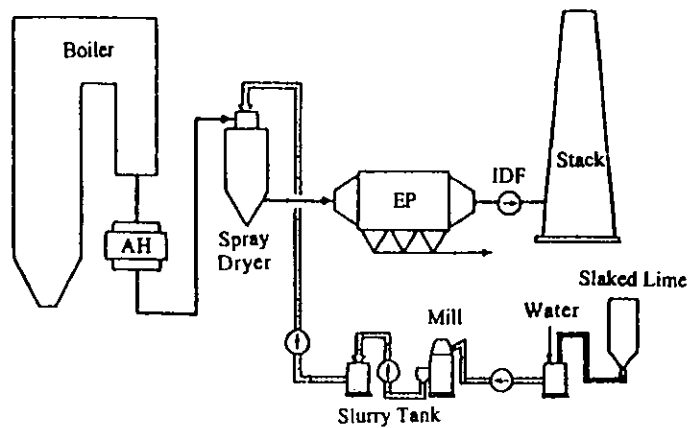


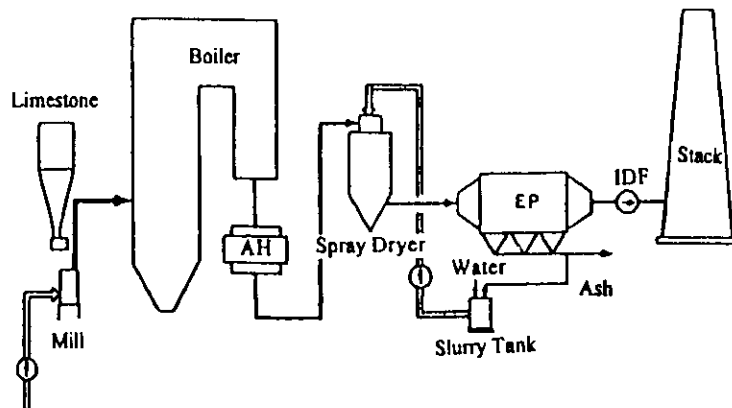
Fig.7.7.3 Magnesium Hydroxide Method Desulfurization/Denitration Process Flow Sheet<sup>4)</sup>



(a) Desulfurization within the furnace System



(b) Semi Dry Desulfurization System



(c) Dust Recirculation Semi Dry Desulfurization System

Fig.7.7.4 Simple Dry, Semi Dry Desulfurization <sup>5)</sup>

(6) Desulfurization using activated carbon

The development of this process is being furthered in Japan and Germany. This desulfurization, denitration method, splits the activated carbon in to two layers. The first layer absorbs the  $SO_2$ , while ammonia is injected into

the second layer, utilizing the activated carbon as the catalyst for denitration in order to change the NO<sub>x</sub> into nitrogen. The absorbed SO<sub>2</sub> is recovered as sulfur or sulfuric acid. The construction costs of the activated carbon process are lower in comparison with that of other wet processes but operating costs run high.

#### 7.7.2 Future Trends for Flue Gas Desulfurization

It is believed that for the time being the majority of flue gas desulfurization will be done by the lime gypsum method. In the future, there will be requests for more compact gas treatment, including wastewater treatment facilities. And, as the majority of the coal combustion in Japan utilizes coal with low sulfur content, there will also be demands for reduced emission levels, and improvements in the desulfurization efficiency. As one step in energy saving measures, methods for utilizing waste heat and reheating exhaust gas to prevent steam plume are being investigated. As a way to reduce the wastewater processing plants, the non-wastewater process is being examined. There are two methods which have been developed. One such process calls for the use of heat from exhaust gas to vaporize the wastewater, after which a dust collector is utilized to remove solid particles within the wastewater. Another method separates solids and liquids using electro dialysis on the waste water.

It is believed that there is an increase in the number of flue gas desulfurization units also in developing nations. However, there is an inclination to buy cheaper units and sacrifice the desulfurization efficiency. Facilities which produce sulfur or sulfuric acid by-products or directly produce fertilizer by-products all have high facility costs. As by-products are recovered mixed with coal; when using the dry process which uses limestone and slaked lime, and also the simple semi dry desulfurization method, the effective use of these compositions will be an important issue in the future.

## 7.8 Improving combustion as a countermeasure to NO<sub>x</sub>

### 7.8.1 Introduction

Within the technology for the prevention of the discharge of nitrogen oxides, there are technologies for NO<sub>x</sub> suppression (low NO<sub>x</sub> combustion technology) and flue gas denitrification, and they are put to practical use at most combustion facilities. A low-NO<sub>x</sub> burner, a two-stage combustion method, an exhaust gas recirculation method and a denitration method within the furnace for large boilers are the means usually adopted to improve combustion as countermeasures for NO<sub>x</sub>.

### 7.8.2 History of combustion improvement as a countermeasure against NO<sub>x</sub> in Japan

NO<sub>x</sub> is harmful by itself in addition to being one of the materials that cause photochemical oxidants which tends to occur frequently in the years since the incident at Rissho high school in the summer of 1970. Various countermeasures have been adopted for the emission source such as combustion fixation equipment and automobiles.

The Environment Agency established an environmental quality standard for NO<sub>2</sub> (a value of 0.02 ppm or less per hour for a 24-hour average) in May 1973, but, in July 1978, it was again revised upward to the 0.04 to 0.06 ppm zone per hour on a daily average basis.

The Agency, in order to achieve this environmental quality standard, established a NO<sub>x</sub> emission standard for smoke and soot emitting facilities in August 1973 (the first regulation) and, after that, expanded the reinforcement in steps in December 1975 (the second regulation), June 1977 (the third regulation), August 1979 (the fourth regulation) and September 1983 (the fifth regulation).

Along with these regulations, the development of low NO<sub>x</sub> combustion technology was energetically carried forward in every direction. At first, the two-stage combustion method was typical for low NO<sub>x</sub> combustion technology, but, then, each maker developed a low NO<sub>x</sub> burner and, afterwards, an improved low NO<sub>x</sub> burner and an in-furnace denitrification method were developed and put to practical use.

Resulting from this, at present, it was possible to decrease the NO<sub>x</sub> concentration in exhaust gas by 60-70% through improved combustion for boilers, to 200 ppm for coal combustion, 100 ppm for heavy oil combustion and to about 50 ppm for gas combustion. Expenses for reduction is about 1/10 those of flue gas denitration<sup>1)</sup>.

### 7.8.3 Low NO<sub>x</sub> combustion technology in Japan

#### (1) The generation of NO<sub>x</sub> in the combustion process

As for nitrogen oxides which are generated during combustion, some is NO<sub>2</sub>, but most is NO. NO is oxidized in the atmosphere and becomes NO<sub>2</sub>. NO and NO<sub>2</sub> are together called NO<sub>x</sub>, but, NO<sub>x</sub> is classified into, thermal NO<sub>x</sub> which is generated from N<sub>2</sub> and O<sub>2</sub> in reaction in high temperature conditions in the combustion air, and fuel NO<sub>x</sub> which is generated from oxidization of N during in fuel combustion of fuel.

More thermal NO<sub>x</sub> is produced when ① the combustion temperature is higher, ② higher oxygen

concentration in the combustion zone, and ③ combusted gas stays longer in high temperature area. Also, the more concentrated the oxygen, the more fuel NO<sub>x</sub> will be produced.

(2) Low NO<sub>x</sub> combustion technology

To decrease the discharge quantity of NO<sub>x</sub> at combustion in industrial equipment, NO<sub>x</sub> generation should be restrained according to the opposite principle mentioned above for the generating mechanism or flue gas denitrification should be conducted to remove the produced NO<sub>x</sub> from exhaust gas. Fig. 7.8.1 illustrates the case of applied NO<sub>x</sub> restraint technology and flue gas denitrification technology for boilers in the generation of electricity.

To decrease the quantity of NO<sub>x</sub> emissions, combustion should implement countermeasures for NO<sub>x</sub> reduction first. Besides the implementation of a low air fuel ratio, or preventing air from invading the furnace, it is very important not to raise the temperature in the furnace, the furnace's load or the preheated air temperature higher than needed, considering the heating process and the entire balance of the equipment.

NO<sub>x</sub> countermeasures by improved combustion include the following.

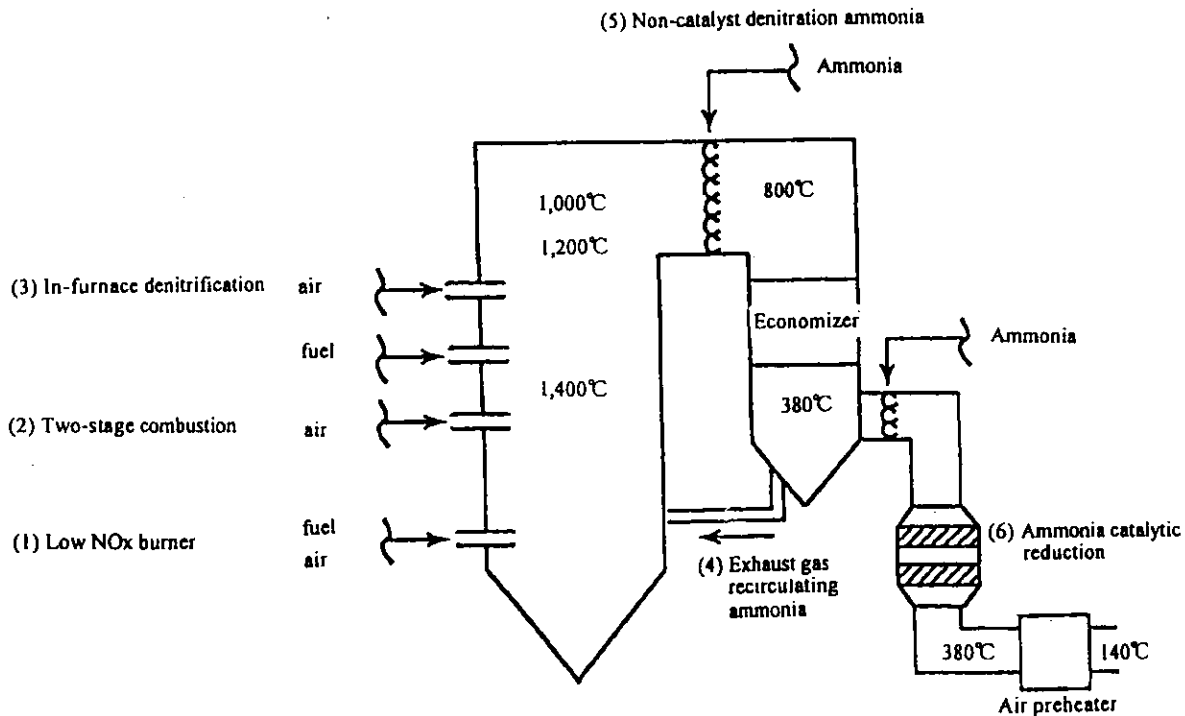


Fig. 7.8.1 NO<sub>x</sub> reduction technology for boilers for power generation

① The use of the low NO<sub>x</sub> burner

Because the cost of the equipment to use a low NO<sub>x</sub> burner is comparatively low, and the reduction of NO<sub>x</sub> is effective, it is the mainstream NO<sub>x</sub> countermeasure at present. Methods to reduce NO<sub>x</sub> have been developed through decreased oxygen concentration, a lower flame temperature, and a shorter period of residence time gas in a high temperature area. One or a combination of these methods are incorporated into various types of businesses such as (a) a rapid combustion-type, (b) a divided flame-type, (c) auto-recirculation-type, or (d) a step-by-step

combustion-type to reduce NO<sub>x</sub>. A step-by-step combustion type of burner is most popular because it is especially effective against both thermal NO<sub>x</sub> and fuel NO<sub>x</sub>, and there is a considerable reduction in the NO<sub>x</sub> ratio.

② The two-stage combustion method

Air for combustion is supplied in two stage, where, in the first stage, the supply of the air amount is limited to 80-90% of the theoretical amount of air, and, in the second stage, the shortage is supplemented with perfect combustion in the total system. By forming a deoxidization area in the first stage, the generation of NO<sub>x</sub> is restrained by the decline of the flame's temperature and the drop in the oxygen concentration. This method has been adopted for almost all large-sized boilers.

③ In-furnace denitrification method

This method is used to deoxidize NO<sub>x</sub> with hydrocarbon fuel by bypassing part of the main fuel and uses it as a fuel for denitrification and the denitrification reaction is completed entirely within the furnace. A reburning system and the three-stage system belong to this method. A reduction in NO<sub>x</sub> of 40% to 50% is possible, and, if used in conjunction with other improved combustion methods, an 80% reduction can be reached. However, Since this method requires a high (long) furnace, its application to existing furnaces is difficult in many cases<sup>1)</sup>.

④ Exhaust gas recirculating

This method mixes part of the exhaust gas in the air to be burned as shown in Fig.7.8.1 and, through combustion, NO<sub>x</sub> reduction is attempted by decreasing the flame temperature. The exhaust gas recirculating rate, restricted by the limit of stable combustion, is generally 10-20%.

(3) Recent trends in low NO<sub>x</sub> combustion technology

With the increase of pulverized coal-combustion boilers since the oil shock, many makers have energetically carried forward with development of a low NO<sub>x</sub> burner to burn pulverized coal. The PM burner divides the pulverized coal through a distributor into the pulverized coal flow of high concentration and low concentration to supply and forms a fuel surplus flame and a thin flame in the furnace. Exhaust gas is introduced in the top and bottom of the fuel surplus flame to delay the diffusion mixing, forming a strong deoxidization flame in the first combustion area of the burner in an attempt to keep production of NO<sub>x</sub> low. The burner itself reduces NO<sub>x</sub> to 200 ppm. In a combination of two-step combustion, reduction as low as 100 ppm is reported to have been achieved.

Looking at the advanced MACT method and the expanded deoxidization method (INPACT method) for denitration in the furnace, there is much residence time, between the burner and the over-fire air port (AOP). In the middle, there is an inter-stage air port (IAP) for two-stage combustion to produce namely multiple combustion stages to reduce the amount of NO<sub>x</sub>.

To counter NO<sub>x</sub>, the fuel direct injection (FDI) burner system was developed for the furnace which performs a high temperature preheated combustion. FDI is a method that combusts fuel and air in the furnace, directly injecting them separately at high speed and with certain intervals into the furnace, which has a high temperature exceeding the fuel's auto ignition temperature. Because the gas in the furnace is sufficiently mixed (the exhaust gas recirculating effect) by each jet stream, and the flow into the furnace of the fuel and air to be burned is nearly parallel, a rapid mixing does not occur (slow combustion effect). The two effects from this are that the occurrence of a peak temperature in the flame is suppressed and the amount of NO<sub>x</sub> discharged is reduced to the 1/10 level.

#### 7.8.4 Indicated problems

As compared with the flue gas denitrification method, the NO<sub>x</sub> countermeasures with the improved combustion method has an inferior NO<sub>x</sub> reduction percentage, but the cost is 1/10 that of the flue gas denitrification method. Even in the case of employing the flue gas denitrification method, the cost is favorable as it reduces the amount of ammonia, if, at first, the flue gas is processed after the decrease in NO<sub>x</sub> from improved combustion. Also, depending on the combustion equipment, sometimes there is no space to install flue gas denitrification equipment.

With the rise of the acid rain problem, as for the flue gas processing by developing countries, countermeasures against SO<sub>x</sub> have become the focus at present, but the discharge of NO<sub>x</sub> will become a big problem. Improved combustion as a countermeasure against NO<sub>x</sub> should be immediately put to practical use in developing countries and a method with little energy loss occurring is especially needed.



## 7.9 Denitrification of Exhaust Gas

### 7.9.1 Introduction

Air pollution resulting from nitrogen oxides (NO<sub>x</sub>) in Japan, has been improved through the use of the low NO<sub>x</sub> combustion and the flue gas denitrification.

### 7.9.2 History of Flue Gas Denitrification Techniques in Japan

As previously mentioned, the incident where photochemical smog plagued the Risho High School in Tokyo in 1970 placed focus on NO<sub>x</sub> as an air pollutant. Along with the low NO<sub>x</sub> combustion techniques, research in flue gas denitrification techniques were also begun.

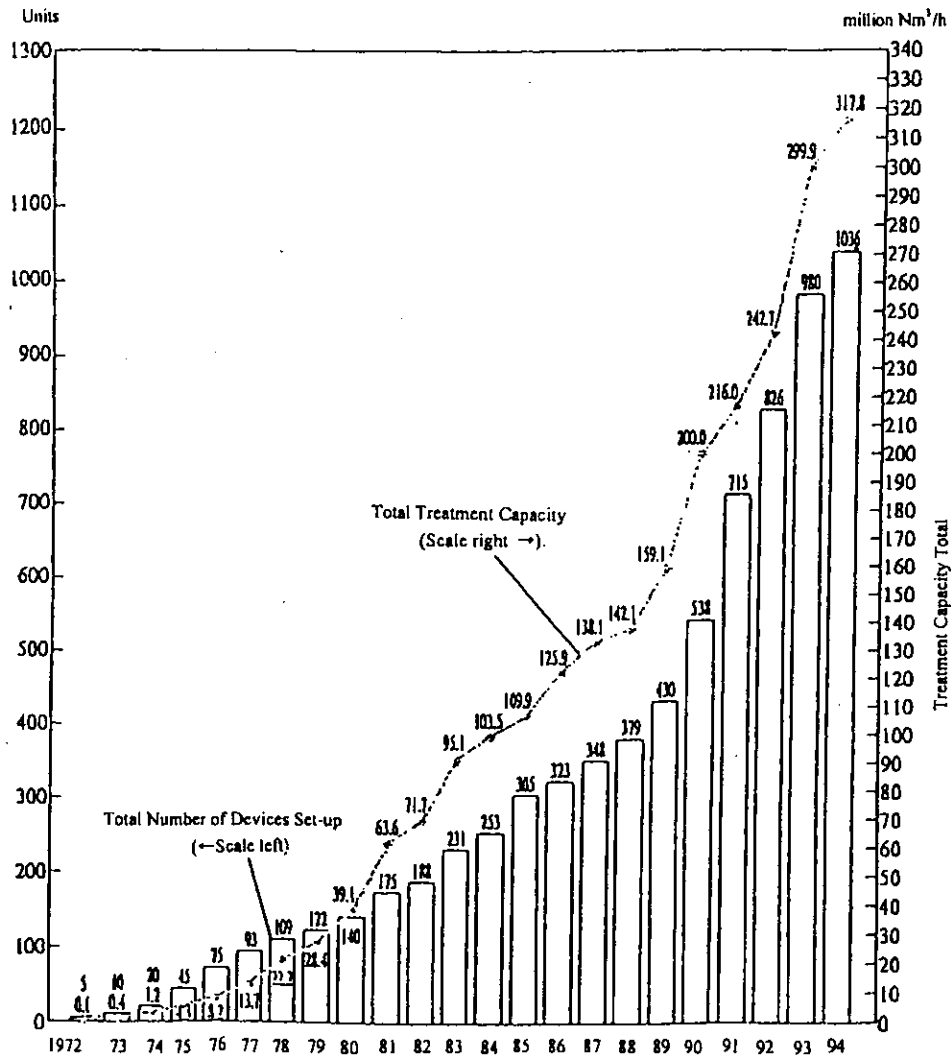
In the early stages, the catalytic cracking process, catalytic oxidation method, and non-selective catalytic reduction process were some of the dry process researched. Meanwhile, wet process such as the direct absorption method, oxidation absorption process, liquid phase absorption method, and complex salt creating absorption method were researched. However, the reaction of NO was poor and in the end these methods did not make it to the stage of practical use. The practically applied (includes substantive testing) flue gas denitrification process is illustrated in Table 7.9.1<sup>1)</sup>. For the most part, the dry process is the mainstream in practical application in Japan.

Table 7.9.1 List of Denitrification Process

		Reaction Agent	Method	Status Practical Application
Dry method	Ammonia Catalytic Reduction Process	NH <sub>3</sub> , (NH <sub>2</sub> ) <sub>2</sub> CO	Utilizes a catalyst, causes a reaction between ammonia and NO <sub>x</sub> , and separates the non-harmful nitrogen (N <sub>2</sub> ) and water vapor (H <sub>2</sub> O)	Used in boilers, gas turbine, diesel, garbage incinerator, and heating furnaces, accounts for over 90% of the denitrification devices currently in use
	Non-catalytic Reduction Method	NH <sub>3</sub> , (NH <sub>2</sub> ) <sub>2</sub> CO	Uses ammonia as a reduction agent, which is then sprayed into the exhaust gas, reduces NO <sub>x</sub> into N <sub>2</sub> using a gas phase non-catalyst	Used in petroleum heating furnaces, garbage incinerators. Denitrification rate is low and there are many non-reacting reduction agents, therefore there are only a few points of application.
	Activated Carbon (simultaneous desulfurization · denitrification)	NH <sub>3</sub>	Adsorbs SO <sub>x</sub> from the exhaust gas using activated carbon (or activated cokes), the NO <sub>x</sub> is decomposed to N <sub>2</sub> by the NH <sub>3</sub> agent utilizing activated carbon (activated cokes) as its catalyst.	Used in boilers and incinerators. It is also being used as a simultaneous desulfurization/denitrification unit in industrial fluidized bed boilers.
	Electron Beam Irradiation Method (simultaneous desulfurization · denitrification)	NH <sub>3</sub>	NH <sub>3</sub> is added to the exhaust gas and irradiated with an electron beam. The SO <sub>x</sub> is converted into ammonium sulfate and the NO <sub>x</sub> is converted into ammonium nitrate.	US and Germany have long performed testing for verification of the method. Testing also being done in Japan. Hopes for use in boilers and incinerators.
Wet method	Oxidation Reduction Method	O <sub>3</sub> , ClO <sub>2</sub> , Na <sub>2</sub> SO <sub>3</sub>	NO is oxidized using ozone or chlorine dioxide and adsorbed by a soda solution.	Several units being used in relatively small boilers and heater furnaces. Not easily used for large-scale facilities as the oxidizing agents such as ozone and chlorine dioxide are expensive.

Among dry process, the ammonia catalytic reduction process is the most widely used and accounts for about 90% of the total in treatment capacity<sup>2)</sup>. Also, in regards to the flue gas processing system which has made possible in recent years the ability to simultaneously perform both denitrification and desulfurization, substantive testing is being run and the development of this technique is continuing.

As shown in Fig.7.9.1<sup>3)</sup>, the flue gas denitrification system began to be implemented from around 1972. By the end of fiscal 1994, there were 1,036 units in operation. The total treatment capacity of exhaust gas is 317.8 million m<sup>3</sup>N/h, which exceeds that of flue gas desulfurization systems.



Note: The values shown are those current as of January 1, for the years up to 1982. For the years starting from 1983, the values shown are those which were current as of March 31.

Source: Environment Agency

Fig.7.9.1, Status Reports for Flue Gas Denitrification Devices based on Year

### 7.9.3 Recent trends in Flue Gas Denitrification Technology in Japan

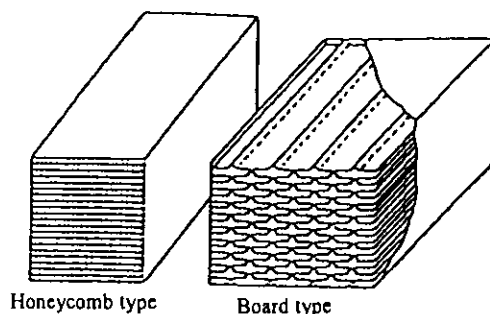
#### (1) Ammonia Catalytic Reduction Method

NH<sub>3</sub> is injected into the exhaust gas and due to the function of the catalyst, the NO<sub>x</sub> in the gas is decomposed to N<sub>2</sub> and H<sub>2</sub>O. This basic reaction is shown as follows in Eq. (1) and (2):



The catalyst which is currently being used, uses TiO<sub>2</sub>, which receives little toxicity from SO<sub>x</sub>, as a catalyst support and V<sub>2</sub>O<sub>5</sub>, WO<sub>3</sub>, etc. as active bodies. Depending upon its use, a number of substances with different component make-up are used. In order to prevent the catalyst from losing pressure to some extent, the shape of the medium is either the honeycomb type and the board shaped parallel flow type. (refer to Fig.7.9.2<sup>4)</sup>)

As a result of all the technological development that has been done up until now, denitrification systems based on the ammonia catalytic reduction process are now able to run stably on automatic pilot. They are widely used in power generator boilers, diesel engines, gas turbines, and garbage furnaces. At present, the ammonia catalytic reduction method is the most popular technology used in the denitrification units.



The ordinary dimensions of the honeycomb type are 150 mm × 150 mm, and 500-1,000 mm long. The board type comes in various sizes.

Source: Junpei Ando: Exhaust Gas Purification Technology of the World, Coal Technology Research Center, 141 (1990)

Fig.7.9.2 Typical shapes of catalysts

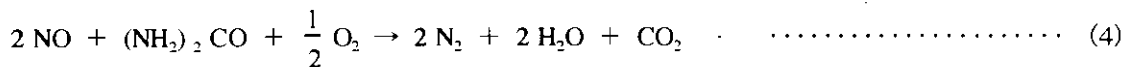
#### (2) Non-catalytic Reduction Process

This method is based on the theory that by using ammonia or urea water as a denitrification reducing agent and spraying it into exhaust combustion gas in the temperature range of 800~1,000°C where both NO<sub>x</sub> and O<sub>2</sub> exist, NO would be selectively reduced. The basic reaction is illustrated in Eq. (3) and (4).

• In the case of ammonia



• In the case of urea water



As clearly seen in the reaction equations, in order to perform the non-catalyst reduction method, the existence of oxygen in the exhaust combustion gas is indispensable. However, if the coexisting oxygen concentration exceeds 1% or more then the rate of denitrification will indicate a maximal value at a certain density as the production of the NO by-product increases due to the oxidation of the ammonia near 900°C.

This method is used in boilers, garbage incinerators, etc. However, the rate of denitrification is not very high, at around 30~40%. In particular, in the case of garbage incinerators, it is widely used as a method of reduction when NOx reaches its peak concentration.

### (3) Techniques for Simultaneous Desulfurization and Denitrification<sup>5)</sup>

As flue gas process techniques, there are already established techniques, the lime-gypsum wet process for desulfurization and the ammonia catalyst reduction process for denitrification. Both these methods are in practical use. However, focus has centered on promoting the development of a dry process which implements simultaneous desulfurization and denitrification techniques due to problems such as the need for a vast amount of water when using the lime-gypsum wet process. Or in the case of the ammonia catalyst reduction process, the need for measures to prevent the clogging of the latter portion of the equipment due to the life span of the catalyst or ammonia leaks.

#### ① Activated Carbon Method<sup>6)</sup>

This process utilizes activated carbon (or activated cokes) in order to adsorb the SOx out of the exhaust gas. Also due to the catalytic effects of the activated carbon, NH<sub>3</sub> decomposes NOx to N<sub>2</sub>, and both desulfurization and denitrification occur at the same time. Besides being used in several incinerator units, it was put into operation as the denitrification unit for the fluidized bed boiler (350 MW) at the Electric Power Development Co., Ltd., Takehara thermal power plant which began operation in 1995.

In this method, the SOx in the exhaust gas is easily adsorbed by the activated carbon in the desulfurization tower. It is captured in the form of sulfuric acid or ammonium sulfate (hereafter abbreviated as (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) (when injected with ammonia.) However, the adsorption capacity of activated carbon is limited and regeneration is necessary. Therefore, the activated carbon is used as a moving bed, shifted to a regeneration system where it is regenerated and then circulated around for use once again.

Also, activated carbon is the same as the metallic catalyst which is used in the ammonia catalyst reduction method, it works as a reduction and decomposing catalyst due to the existence of the ammonia. Also, it is believed to have the ability to remove NOx due to the denitrification reaction caused by the reducing substance on the surface of the activated carbon or the adsorption.

#### ② Electron Beam Irradiation Process<sup>7)</sup>

This technology achieves simultaneous desulfurization and denitrification by adding NH<sub>3</sub> to the exhaust gas, using an electron beam to irradiate the substance changing the SOx into (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and the NOx into ammonium

nitrate. This method has cleared the substantive testing level where it was used in boilers of coal-fired thermal power stations and is in the stages prior to actual application.

Firstly, a dry type electrostatic precipitator is used to roughly collect the dust in exhaust gas of about 130°C. After this the gas is passed through a GGH where it is cooled down to about 110°C. Furthermore it is cooled down to around 60~70°C in the cooling tower, a temperature which is suited for the desulfurization reaction. It is then irradiated by electron beam in the reaction chamber.

By submitting gas to an electron beam for irradiation, radicals with extremely strong oxidation properties such as O\*, OH\*, HO<sub>2</sub>\*, are created from the elements O<sub>2</sub> and H<sub>2</sub>O found in the exhaust gas. It is due to these radicals that SO<sub>x</sub> and NO<sub>x</sub> are oxidized down to H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub>. These strong acids react with the previously added NH<sub>3</sub> to create (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and ammonium nitrate.

#### 7.9.4 Specified Issues

Looking from the standpoint of the reexamination of worldwide and Japanese energy demand, accompanying the increase in usage of fossil fuels, especially coal combustion, there will be further calls for the reduction of total exhaust for SO<sub>x</sub> and NO<sub>x</sub>.

In addition, due to the recent global environmental issues, there is a need to make consideration toward the reduction of CO<sub>2</sub>. By putting dust collector, flue gas desulfurization units, and denitrification devices into operation at thermal power generators, this only results in a decrease in the efficient transmission of electricity. There needs to be technological development in the future which is geared to the construction of an overall optimum flue gas processing system which also takes into consideration CO<sub>2</sub> exhaust.

## 7.10 Toxic Gas Treatment

Toxic gas emissions come in diverse forms depending on the source of this emission. When selecting the collection method for polluted gas and the treatment methods, it is necessary to take into consideration the properties of the raw gas, at the same time taking into account the facility site, surrounding environment, and economical efficiency. ① The thermal combustion method, ② catalytic method, ③ condensation method, ④ adsorption method, and ⑤ absorption method are just some of the processes found when categorizing the treatment methods available. ① Combustion, ② catalytic, and ③ condensation methods are described in Chapter 7.12 under hydrocarbon control. In this chapter we will focus on the ④ adsorption and ⑤ absorption methods.

### 7.10.1 Adsorption Method

The adsorption method utilizes a porous substance with a large internal surface area, has a special affinity for the adsorbed gas, and is selective. Consequently, it is necessary to make selections based on the components of the gas to be treated.

Exhaust gas treatment using the adsorption method makes it possible to obtain a solid removal efficiency with a relatively basic equipment. It is necessary to take into consideration such factors as the treatment of used adsorbents, the recycling of the adsorbed components or their disposal. In general, in the treatment of low concentrate gases, most methods perform an exchange with a new adsorbent which does not utilize the desorption process. In cases where high concentrate gases are treated, the desorption process is incorporated and the recycle of desorbed components and regeneration of the adsorbent are performed. Also, the adsorption device is employed as an gas concentration device in conjunction with other equipment such as a combustion device, for use in treatment methods.

#### (1) Adsorbent

Fig.7.10.1<sup>1)</sup> illustrates the types of adsorbents and their main uses. Adsorbents are divided up into two different categories, physical adsorbents and chemical adsorbents. Physical adsorbents consist of such items as activated carbon and zeolite. The most commonly used adsorbent is the activated carbon. Adsorbents come in different forms, granular, fibrous, spherical, and honeycomb shaped. On the other hand, chemical adsorbents include impregnated carbon which retains acids, alkali, oxidizing agents, and catalytic substances on the surface of the activated carbon. For example, as a treatment for base substances such as ammonia and amine, sulfonated carbon, chemically treated coal, is used. And for the treatment of acidic substances, sulfonated soda carbon is used. In addition, for the treatment of sulfur compounds, there are chemical adsorbents which use iron ions as a base agent or which apply the use of ion-exchange resins.

Table 7.10.1 Types of Adsorbents and their Usage

		Type of Adsorbent	Main Use
Physical Adsorbent		Activated carbon	Recovery of deodorant hydrocarbon related solvents, desulfurization, denitrification, recovery of chlorofluorocarbon, recovery of halogenated hydrocarbons, adsorption of styrene
		Zeolite (hydrophilic)	Adsorbs ammonia, demoiseurize air, deodorize night soil treatment facility, oxygen in air, substrate for biological deodorizing method, adsorbent for methyl sulfide and methyl disulfide
		High silicazeolite (hydrophobic)	Adsorption of alcohol, aromatics, and paraffin from aqueous solution
		Molecular sieving carbon	Oxygen in air
		Silica gel	Demoisturization, deodorization of air
		Alumina	Demoisturization, deodorization of air
		Activated clay	Refine solution, decoloring deodorization of fats
Chemical adsorbent	Additive/adsorbent agents	Basic gas	Adsorption of ammonia, trimethylamine
		Acid gas	Adsorption of hydrogen sulfide, methyl mercaptan
		Metal impregnated carbon	Adsorption of carbon monoxide, hydrogen cyanide, carbonyl chloride
		W/oxidizing agent impregnated carbon	Decomposition/adsorption of nitrogen monoxide, hydrogen sulfides, amine, aldehyde, acrolein
		Ion-exchange resin	Adsorption of ammonia, trimethylamine, hydrogen sulfide, methyl mercaptan
		Iron oxide deodorant	Adsorption of hydrogen sulfide, acetic acid

(2) Adsorption equipment (Adsorber)

The methods available methods for contact between the adsorbent agent and treated gas in the adsorber are the moving bed, fixed bed (packed) and rotary honeycomb method. In most cases the fixed bed method is used due to its easy equipment structure and operation management.

The fixed bed adsorber is illustrated in Fig. 7.10.1. There are two types of packing methods into fixed bed, one where the fixed bed is immediately filled with adsorbent and another where it is exchanged using a removable cartridge.

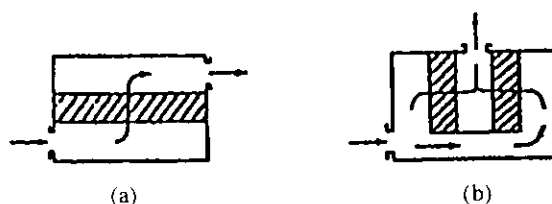


Fig.7.10.1 Fixed Bed Adsorber<sup>1)</sup>

For granular activated carbon, the adsorbent is used in a 30-80cm depth single bed or multi-layer bed. If the packed bed is too thin, then the concentration distribution within the inner layer will become disproportionate and if it is too thick then pressure loss will increase. For granular activated carbon, the basic current velocity of the raw gas is about 30 cm/s~60 cm/s. For continuous fixed bed systems, it is necessary to have at least 2 towers in order to perform adsorption and desorption alternately. This is in order to perform adsorption in one tower while doing desorption in the other. When repeatedly performing this cycle, desorption is completed prior to saturation of the adsorbent by the adsorption substance, and cooling must be done to the adsorption temperature.

The moving bed adsorption method, shifts the adsorbent during treatment, transferring the adsorbent to the recycling process, resulting in consecutive operation. It is a suitable method for the treatment of large volumes of gas. The drawbacks of the moving bed which utilizes granular activated carbon, is that activated carbon is easily worn away and the device becomes easily clogged due to dust. One type of moving bed type is the concentration equipment, which uses granular bead-shaped carbon and is shown in the Fig.7.10.2. In this method, the globular (about 0.7 mm in diameter) activated carbon is slid down a sloped porous board where it moves down successively to the lower level. The raw gas is introduced into the system from the bottom of the tower where it is absorbed. The used adsorbent is continuously sent back to the desorption tower for reuse and the treated gas components are separated and concentrated. Then they are transferred to another treatment device, such as an incinerator, for final treatment.

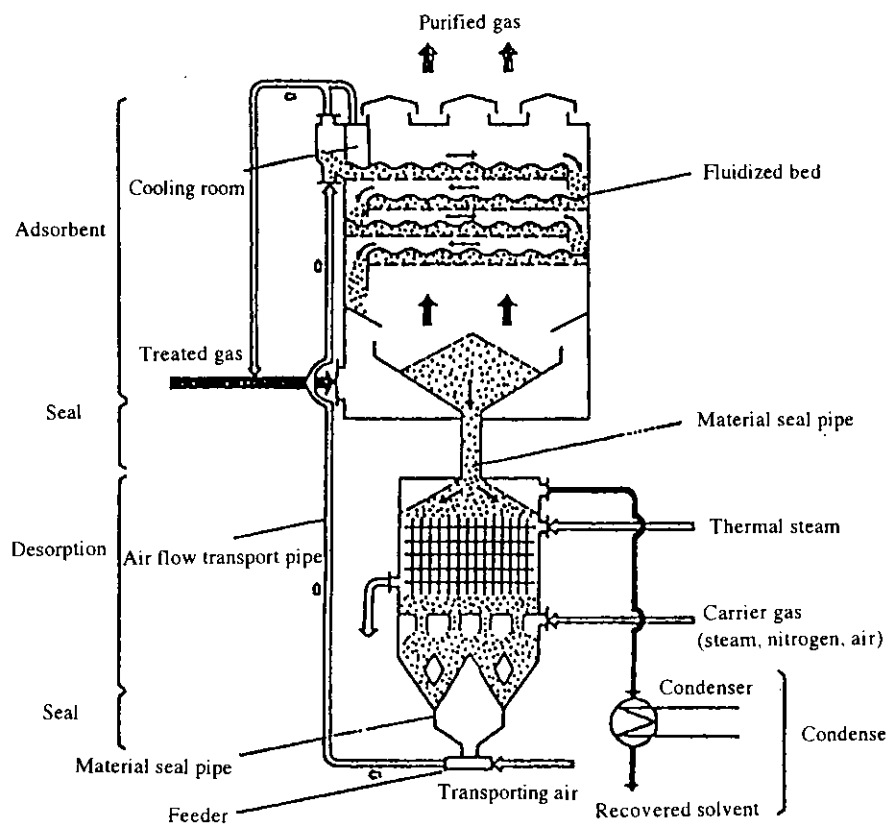


Fig.7.10.2 Continuous adsorption/desorption device using bead-shaped carbon <sup>2)</sup>

The rotary honeycomb adsorber is illustrated in Fig.7.10.3. Activated carbon in the shape of a honeycomb is formed into a cylinder shaped device. The cylinder shaped device is rotated. Adsorption and heated desorption are continually performed and is used for concentration of the treated gas. When the components of the raw gas are combustible, the volume of the gas is reduced and the concentration of the elements are increased. It is then possible to effectively treat these components using a combination of a combustion system and catalytic oxidation system.



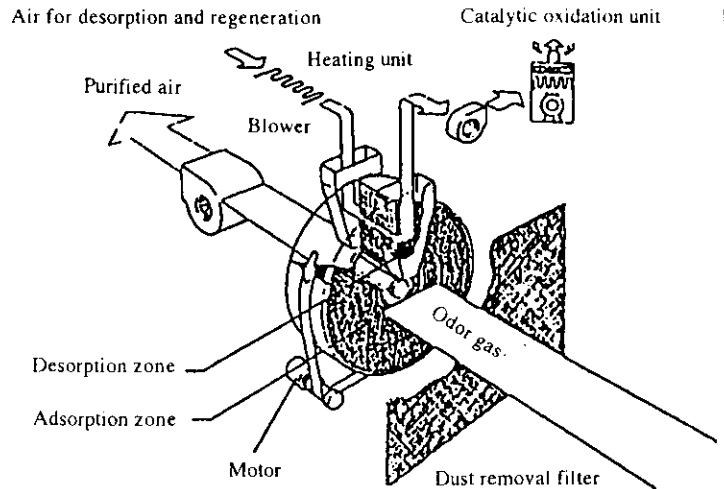


Fig.7.10.3 Rotary honeycomb type continuous condensation equipment <sup>2)</sup>

### (3) Regeneration of adsorbents

Under the desorption method, there exists the thermal swing method using heat and the pressure swing method by reduced pressure. In particular the heated desorption methods, one of thermal swing methods, is widely used. As a heating medium, heated water vapor, heated air, and heated nitrogen gas are used. There are methods performing desorption by carrying out heating and depressurizing at the same time as well. Fire and explosion must be considered when utilizing combustible organic solvents. Especially, in the recovery of ketones such as methylethyl ketone and cyclohexanone, there are instances of fire occurring due to a rise in temperature resulting from the oxidation or thermal storage in ketone. In particular, adequate desorption is necessary when the device stops operating.

In general, in the regeneration of adsorbents, a fair amount of time is required for complete desorption, therefore 100% desorption is not performed. In actuality, from the technical and economical standpoints, a suitable rate of desorption is performed for reusing the adsorbents.

### (4) Pre-treatment of Exhaust Gas

In the event that the treated gas is of high temperature and contains a large amount of dust or mist, it is not suitable for adsorption. It is also the reason for loss of adsorbents. Also, when the treated gas has a high concentration, in order to extend operations over a long period of time, it is necessary to attach appropriate facilities for pre-treatment. The water absorption method is commonly used as a pre-treatment method. Ample care must be taken that spray from the cleaning fluid is not carried into the adsorption bed in order to keep the activated carbon from becoming wet. The lower the temperature the more effective removal is. In general, it is essential to keep the temperature of the treated gas below 40°C.

## 7.10.2 Absorption Method

### (1) Neutralization Absorption Method and Oxidation Absorption Method

The purification-absorption method is most frequently used as a removal method for toxic gas. When largely categorizing the absorption treatment methods, one method calls for absorption by water, while other methods are forms of chemical absorption which utilize absorbents to cause chemical reaction with polluted gas. The latter is divided into two methods, one which neutralizes the toxic components in the gas and one which separates the toxic components from the non-toxic components through oxidation. Water absorption method must be applied for the dissolvable substance in water. Neutralization method includes the removal of basic gas such as ammonia, trimethylamine by acid washing, and other type of removal which removes acid gas such fluorine, hydrogen fluoride, chlorine, hydrogen chloride, mercaptan, and hydrogen sulfide by alkaline washing. In the oxidation absorption method, aqueous solutions such as hypochlorous acid soda, potassium permanganate, and hydrogen peroxide are used as absorbents. In particular, this is used in the decomposition treatment of odor gases.

In situations where dust coexists, it is effective in removing dust at the same time absorbing gas substances. The solubility of organic compound toxic gases in water is in general low. It is difficult to achieve the necessary amount of removal results by just using the adsorbent method. In most cases it is used in conjunction with other treatment methods.

### (2) Absorption Equipments (Absorber)

Among absorption equipments, there are those types which disperse the absorbent into the gas and those which disperse the treated gas into the liquid absorbent. For those gases which are highly soluble in absorbent solutions, the device which disperses the absorbent liquid into the gas is a suitable system. For those gases which are less soluble in the absorbent, the treatment method where the gas is dispersed into the absorbent is appropriate. In general, highly soluble agents for should be removed substances are used as the absorbent. Therefore, in most cases a packed tower or spray tower which disperses the absorbent solution is used. In the treatment of large volumes of gas, porous plate-type tower (Moretana-tower) and cross flow contact equipment are used.

### (3) Mist Removal

In general, the absorption washing method results in secondary pollution as the absorbent turns to mist and scatters into the atmosphere. Therefore it is necessary to install a mist removal device. In most cases mist removal is performed by putting packing into the upper portion of the absorption tower. However, this method has a low effect in the removal of mist. In particular, when gas temperatures are high and include condensed gas, after the mist leaves the absorption tower, the substance becomes condensed within the piping and is littered. For this reason, it is best to install the mist removal device as near as possible to the exhaust outlet.

## 7.11 Automobile Exhaust Gas Control Measures

### 7.11.1 Introduction

Among the automobile exhaust gas control measures, those which apply to gasoline fueled vehicles have nearly been perfected technologically. An important issue which still remains to be resolved is that of how to handle the NO<sub>x</sub>, black smoke and particulate matter which are emitted from diesel powered vehicles.

### 7.11.2 Measure for gasoline fueled vehicle exhaust gas control

The basic concept on the reduction of exhaust gas from gasoline fueled vehicles focuses on ① the control of pollutants through the improvement of the combustion system, ② the after treatment during emission, and ③ the sealing off of areas open to the atmosphere plus the return of evaporative emission to the intake line. However, in general the measures put out for CO, HC and NO<sub>x</sub> are conflicting. The measures being taken for the reduction of emissions from gasoline fueled vehicles have become more complex. And more systems require a combination of a variety of reduction methods. Fig.7.11.1 illustrates a system of measures used for the reduction of exhaust gas from gasoline fueled vehicles.

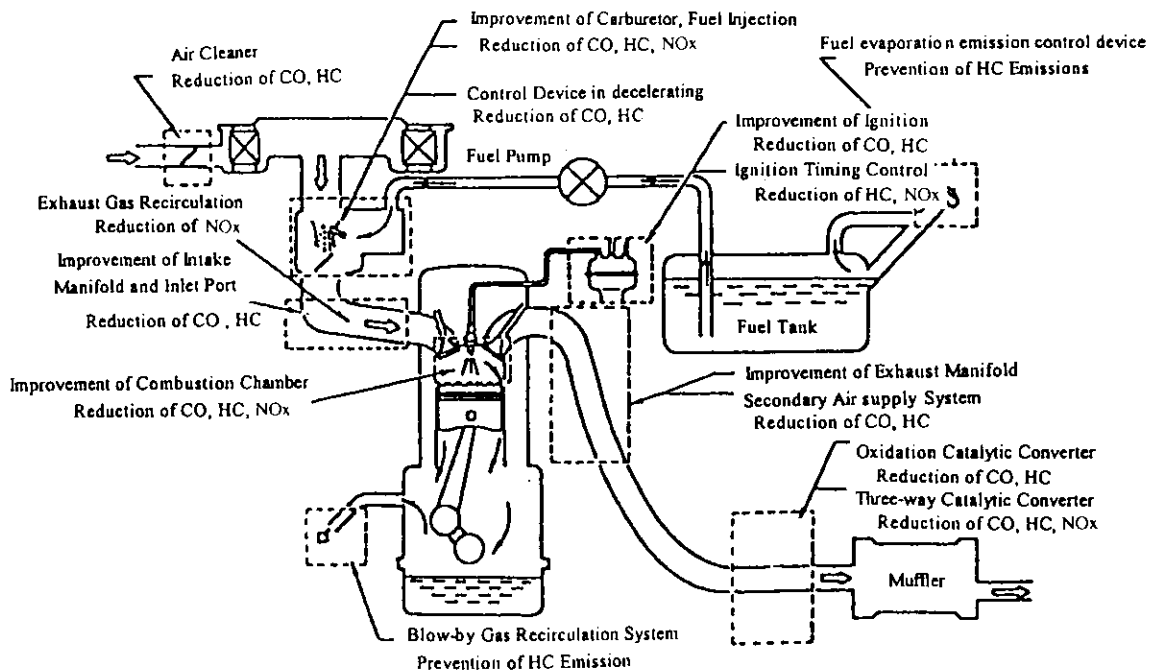


Fig.7.11.1 Example of Gasoline-Fueled Engine Meeting Exhaust Gas Standards

(1) Measures to control pollutants by improving the combustion system

① Ignition timing controlled: In general, by the retard of ignition timing NO<sub>x</sub> and HC can be reduced. However, if the retard of ignition timing is too far then there are adverse effects to the engine power and fuel efficiency.

Therefore, both driving performance and purification of emissions are controlled compatibly by computer.

② Fuel-cut in decelerating : When decelerating, the throttle valve closes, the amount of air in the combustion chamber lessens and incomplete combustion occurs. In order to prevent this from happening, the amount of fuel is cut at the time of deceleration. The vacuum in decelerating is detected and the throttle valve is forced open. The system then increases the intake air volume.

③ EGR: A part of exhaust gas returned into the intake air and the maximum temperature for combustion is lowered to control the production of NOx. The exhaust gas recirculation is 10% and the amount of NOx emitted is cut in half.

④ Lean burn: In general, CO and HC are heavily emitted when the air fuel ratio is overrich. NOx reaches its maximum concentration once it nears its stoichiometric air fuel ratio. Anything less than this level in the air fuel ratio results in a reduction in concentration of emissions. Therefore the air fuel ratio is controlled at around 22-24, controlling the amount of CO, HC, and NOx. A separate measure must be implemented in order to deal with instability in the engine which will occur along with the control of air fuel ratio, in order to obtain compatibility between the reduction in NOx and fuel efficiency.

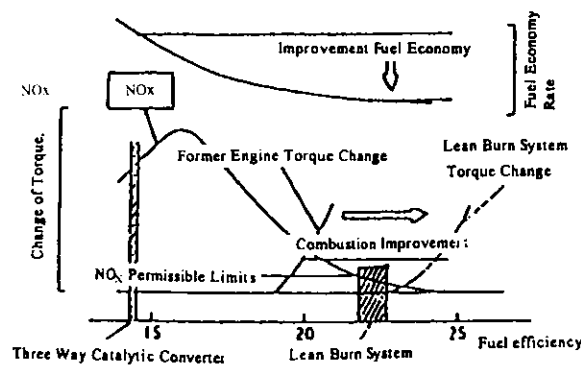


Fig.7.11.2 Lean Burn System

(2) After treatment technologies in the exhaust line

① Oxidation catalytic converter: A catalyst (platinum, palladium, etc.) are installed midway in the exhaust pipe. As the exhaust gas passes through the pipe, the CO and HC are oxidized and are changed into harmless forms, CO<sub>2</sub> and H<sub>2</sub>O. In this case, a system which supplies secondary air before the catalyst is necessary. In the secondary air supply system there is a secondary air injection system which forces air through using an air pump. There is also a system with a secondary air vent which opens into the atmosphere when exhaust gas vacuum is registered in the pulsation of the exhaust pressure from inside the engine exhaust pipe and introduces secondary air into the exhaust pipe corresponding with the exhaust gas vacuum.

② Three way catalytic converter: This device uses platinum or rhodium as its catalyst. Through an oxidation reaction with the CO and HC within the exhaust gas, and by a reduction reaction with the NOx, all three substances are simultaneously converted into non-toxic substances. This differs from an oxidation catalytic converter in that there is a need to control the air fuel ratio to its stoichiometric level. For this purpose, the air fuels ratio feedback system is indispensable, where the O<sub>2</sub> concentration in the exhaust gas is continuously detected and the data is sent

to a computer. Also, the fuel supply system uses a electronic controlled fuel injection system to deal with various types of air fuel ratio controls. (Fig.7.11.3)

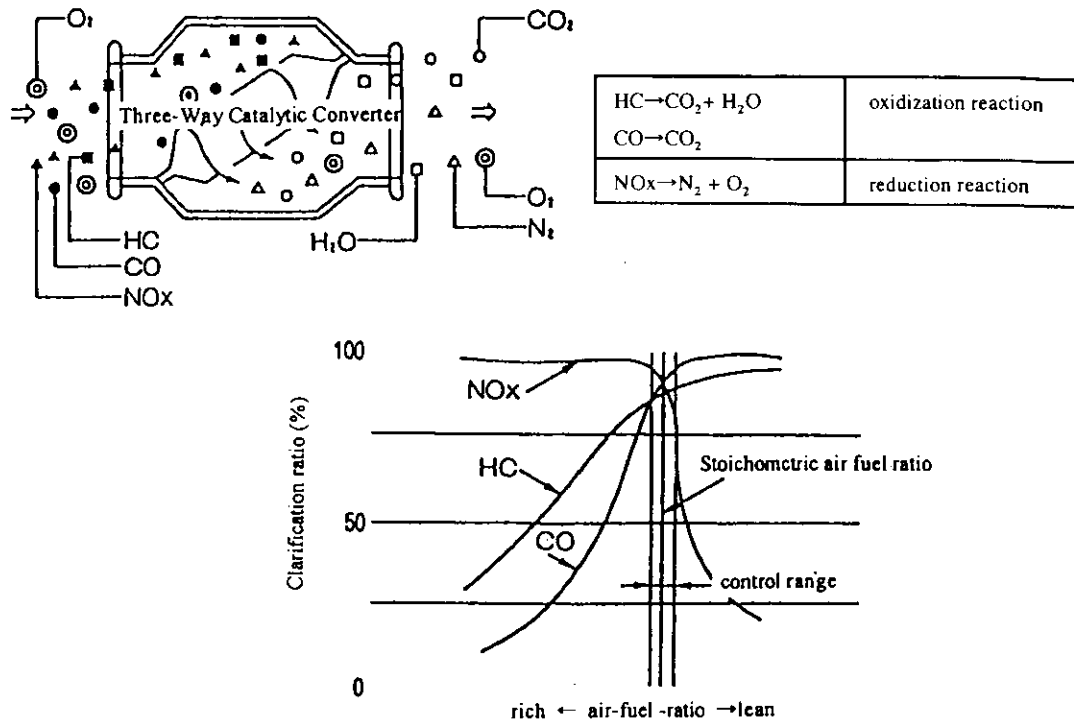


Fig.7.11.3 Principle of a three-way catalytic converter

### (3) Countermeasure of unburned fuel

① Blow-by Gas Recirculation system: The blow-by gas which is blown past the pistons and cylinders, is once again recirculated to the intake line for combustion.

② Fuel Evaporative Emission Control Device: The evaporative fuel from the fuel tank and carburetor is initially adsorbed by the activated carbon (charcoal canister) and the engines exhaust gas vacuum is used to recirculate it into the intake line once more.

### 7.11.3 Diesel powered vehicles exhaust gas emission control measures

Exhaust gas emission control for diesel powered vehicles include ① the improvement of the exhaust and intake line, ② improvement of the combustion chamber, ③ improvement of the fuel injection system, ④ after treatment, and ⑤ improvements to the fuel itself. An outline of these related technologies is shown in Fig.7.11.4.

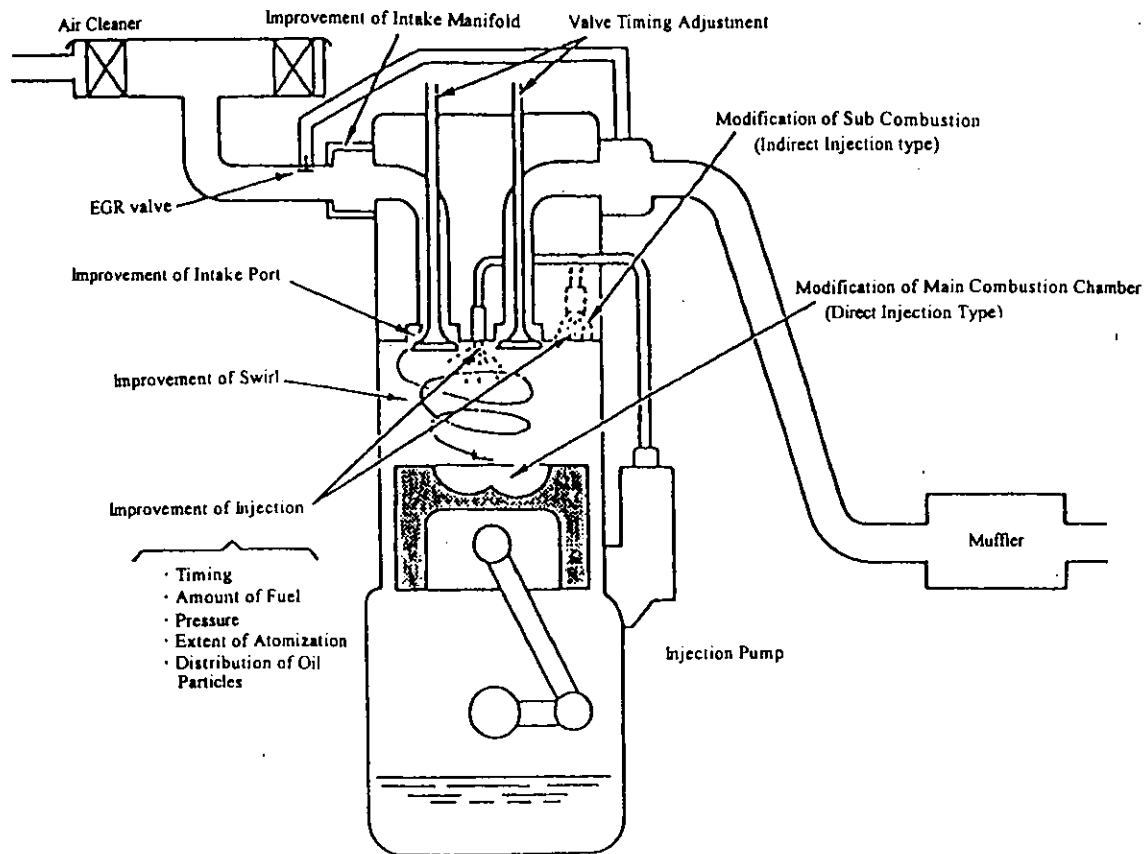


Fig.7.11.4 Example of Diesel-Powered Engine Meeting Exhaust Gas Standards

(1) Improvement of intake and exhaust line

In the intake line the aim is to improve the optimization and volumetric efficiency of the air flow. Improvements in the intake manifold, intake port, and changes of the valve timing are performed. The air intake resistance is reduced and the trapping efficiency is lifted. This not only allows for sufficient appropriation of the air necessary for combustion but also improvements in the mixing of air with fuel using a moderate swirl. This allows for proper performance in fuel combustion.

Also, by increasing valves, the passage way through which the gas passes becomes that much wider, making it capable of performing quick and a lot of gas exhaust and intake. This improves volumetric efficiency. Supercharging is the process by which air, with pressure higher than that of atmospheric pressure, is sent to the engine via a compressor which is operated by a turbine being rotated by exhaust energy emitted from the engine. This supercharging reduces the air fuel ratio resulting in a decline in black smoke and PM (particulate matters). The creation of NO<sub>x</sub> is controlled by charging air, cooled down after having been heated by super charging, to the engine.

For passenger cars, the method where a portion of the exhaust gas is recirculated to the intake line and the combustion temperature is lowered to reduce the NO<sub>x</sub> is already in use. However, for trucks and buses which have a longer lifespan and are used under high load conditions, there are numerous issues dealing with reliability and

durability such as corrosion of the inner engine due to the S content in diesel fuel, abrasion caused by dust, and deterioration of oil. The improvements in technology to control black smoke such as the reduction of the ratio of the S content in diesel fuel and the improvement of combustion, will likely see the increased application of EGR to smaller trucks in the future.

## (2) Improvements of Combustion Chamber

In order to reduce black smoke and PM, the swirl in the combustion chamber should be increased. This is the opposite when aiming to reduce the NO<sub>x</sub>; the swirl should be reduced. The conditions of the gas flow within the cylinders are largely effected by factors in the combustion chamber.

## (3) Improvements of fuel injection system

① The fuel injection timing retard: The NO<sub>x</sub> from diesel engines occurs most frequently for pre-mixed combustion. For pre-mixed combustion, the longer the time lapse between fuel injection and ignition, the more NO<sub>x</sub> emissions are created due to powerful combustion. Consequently, in order to reduce the occurrence of NO<sub>x</sub>, it is best to retard fuel injection as much as possible and shorten the pre-mixed combustion time. However, by timing retard fuel injection the combustion temperature is cut and an increase in black smoke and PM is experienced along with worsened fuel efficiency and power down. In order to overcome these demerits, improvements to the combustion chamber conditions, quicker air flow, and the promotion of a spray diffusion are just some of the improvements to combustion which must be made at the same time. Recently, the control of the fuel injection is being done in order to attain a balance between the reduction of black smoke and PM and improvement in performance. This is being done by controlling the pre-mixed combustion at the onset of combustion in order to reduce NO<sub>x</sub> and promote diffusion combustion during the latter half of fuel combustion.

② High pressure injection: Fuel is injected into the combustion chamber at high pressure and converted into fine particles in order to improve the mixture of air and fuel. By setting the injection pressure at anywhere between 400 kg/cm<sup>2</sup> and 1,000 kg/cm<sup>2</sup>, black smoke can be reduced to 1/4. However, the improvement of combustion via high pressure injection causes an increase in NO<sub>x</sub>. Therefore it is necessary to simultaneously implement NO<sub>x</sub> countermeasures such as retarded fuel injection timing.

## (4) After treatment

For diesel powered vehicles, it is difficult to utilize the oxidation catalytic converter or three-way catalytic converter used by gasoline fueled vehicles as the S content in diesel fuel causes an increase in sulfate and excess O<sub>2</sub> hinders NO<sub>x</sub> reduction treatment. However, measures to cut the ratio of the S content in diesel fuel in recent years, has led to the use of a SOF reducing oxidization catalytic converter in some types of vehicles. Also, development is progressing for the practical use of the Diesel Particulate Filter (DEF) which is a treatment which reduces black smoke by trapping dust in a honeycomb trap (material: cordierite or silicon carbide) through which the exhaust gas passes.

In regards to the reduction catalytic converter, zeolite (crystalline silicate minerals largely containing water) works in conditions where there is a high concentration of O<sub>2</sub>. However, there are many problems such as, the NO<sub>x</sub>

reduction is low (about 10%), low temperatures obtaining no results and the existence of water lowering its effects. Practical application of the catalyst is complex.

#### (5) Measures concerning fuel

The ratio of the S content in diesel fuel was reduced from 0.4% to 0.2% in 1992. Content was reduced to 0.05% starting from 1997. Also, by adding oxygenates such as polypropylene glycol alkyl ether or vegetable oil methyl ester to light oil, HC, CO, and PM can be reduced. In this case, the increase in NOx is controlled by the introduction of cetane number improver.

#### 7.11.4 Specified Issues

There have been remarkable developments in the advancement of technology for reducing exhaust gas from gasoline fueled vehicles. In the future, higher costs for applying these solutions to heavy duty gasoline fueled vehicles is a major problem. Concerning diesel powered vehicle, the reduction of black smoke via improvements in combustion are progressing but the problem of increased NOx still remains. Also, in regards to DPF systems or hybrid systems, the issues of lower costs and reliability and durability of the system still exist.



## 7.12 Hydrocarbon Species Control Measures

### 7.12.1 Introduction

In Japan health damage problems of photochemical smog began from 1970, and countermeasures were implemented to reduce the emission of the causative substances, nitrogen oxides and hydrocarbon species. In addition in recent years countermeasures not only to photochemical smog, but also the hydrocarbon species like benzene, hydrocarbon included chlorine and so on which cause health damage by themselves have become a problem and those countermeasures to these substances have covered a very broad spectrum.

### 7.12.2 Hydrocarbon Species Emission Sources

This section discusses the stationary sources among hydrocarbon species emission sources relating to air pollution are reported. Major emission sources of hydrocarbon species in Japan are combustion facilities such as boilers, the petroleum-related industry including gasoline refueling, chemical industry, printing industry, motor vehicle industry, construction industry and dry cleaning industry. Hydrocarbon species substances exist in the atmosphere as vapor or mist. They are mainly fuel types and various solvents. The hydrocarbon species which are designated as hazardous substances and also widely used substances in Japan are methanol, acetone, methyl ethylketone, benzene, toluene and xylene and hydrocarbon included chlorine, trichloroethylene and tetrachloroethylene and so on. In addition, chlorofluorocarbon, which is a causative substance in depletion of ozone layer, and dioxin which was used as a defoliant in the Vietnam War, are both also hydrocarbon species.

### 7.12.3 Countermeasures to Hydrocarbon Species in Combustion Emission Gas

Due to the wide use of light type heavy oil in Japan, hydrocarbon species in emissions gases produced by combustion facilities like boilers are normally under 10 ppm. Decreasing hydrocarbon species in combustion emission gas depends mainly on the improvement of the fuel and combustion conditions. If a lighter fuel is used, hydrocarbon species generally decrease. It is effective due to improve combustion conditions to adjust mixed air volume. Generally the best air fuel ratio for heavy oil is 1.3 but as shown Fig.7.12.1 if the air fuel ratio is decreased, the concentration of hydrocarbon species in the emission gas increases <sup>1)</sup>. On the other hand, if the air fuel ratio is increased considerably, non-burned hydrocarbon occurs and the concentration of hydrocarbon species increases <sup>2)</sup>.

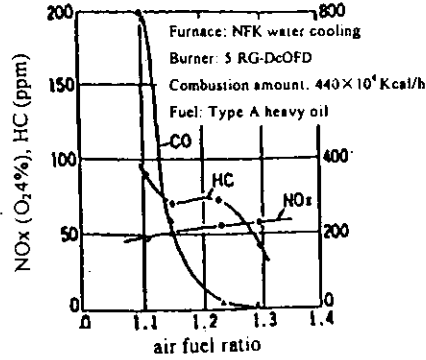


Fig.7.12.1 Relationship between NOx, CO, HC and air fuel ratio

#### 7.12.4 Gasoline Vapor Control Measures

In Japan, gasoline vapor emission during the refueling of gasoline stations by tanker lorries is one of the major causative substances of photochemical smog<sup>3)</sup>. This emission value of gasoline vapor rises according to increased air temperature, and as shown in Fig. 7.12.2<sup>4)</sup>. Therefore, if air temperature rises one degree, the volume of gasoline vapor emission increases by 10%. The main gasoline vapor countermeasure is use of the vapor return method. The equipment is shown in Fig. 7.12.3. This method is for sealing off the piping system and refueling and refluxing vapor to tank trucks during fueling. Because the equipment is relatively cheap, use of this method is mandated in major Japanese cities<sup>3) 5)</sup>.

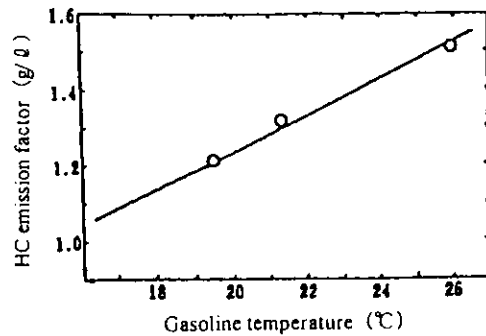


Fig.7.12.2 The temperature of gasoline and the volume of emission of hydrocarbon species during gasoline refueling.

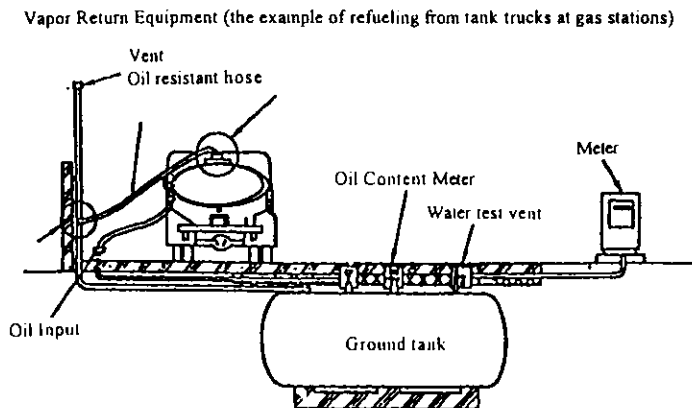


Fig.7.12.3 Vapor Return Equipment

### 7.12.5 Hydrocarbon Species Treatment Apparatus

Treatment methods of hydrocarbon species are broadly divided into the condensation method, absorber method, adsorption method, direct combustion method, and catalyser oxidation method. The adsorption method is already reported in 7.10.

#### (1) Condensation Method

This is a method of recovering vaporized hydrocarbon species by cooling and condensing at a temperature below its boiling point. So it is suitable for cases when there is a high concentration of hydrocarbon species and a small gas volume. In many cases it is rare to use this method only by itself; it is usually used as a pretreatment before using absorber or adsorption equipment. It is used for substances of low boiling points like gasoline and toluene.

#### (2) Absorption Method

This is a method using absorber which has a high solubility for hydrocarbon species gas for contact absorption. Kerosene, light oil, acetone and alcohol are generally used as absorbers. It is used when refueling to tanker lorries from main tanks in oil refineries, but in many cases gas solubility toward absorbers is influenced by temperature, so in Japan the removal ratio may be varied according to the season. When the gas has reactivity against acid or alkalinity, caustic soda or potassium permanganate solution are used in the absorber.

#### (3) Direct Burning Method

With this method, if the combustion efficiency is high then there is a more than 95% removal ratio regardless of the concentration of hydrocarbon species, and it can be used with all the hydrocarbon species except those which emit toxic gases ( $\text{Cl}_2$  etc.), such as hydrocarbon included chlorine. But because supplementary fuel is necessary, this method is often used as removing countermeasures for dust emitted from incinerator or with afterburner for vapor plume removal after the scrubber.

#### (4) Catalyser Oxidation Method

This is a method for the complete combustion of hydrocarbon species gas under combustion conditions of 250~350°C and between 0.1~0.3 seconds with catalysts such as platinum or nickel. The fuel cost with this method is 30~40% lower compared to the direct combustion method. But caution is required when using this method, as the catalyst may be contaminated by some types of emission gases and the catalyst has selectivity towards gases. Also, it is necessary to raise the temperature of low temperature gases. The relationship between gas temperature and oxidation ratio is shown in Fig.7.12.4<sup>6)</sup>.

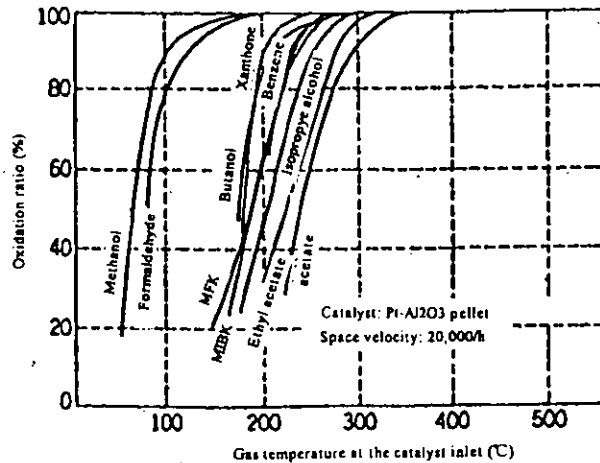


Fig.7.12.4 An Example of gas temperature and oxidation ratio in catalytic combustion method

### 7.12.6 Countermeasures of hydrocarbon included chlorine

Trichloroethylene and tetrachloroethylene, which are extensively used in the electronic and dry-cleaning industries, have become a problem in Japan. It is necessary to suppress as much as possible the emission of these two substances from rooms and tanks before emission gas treatment, and to control the loss of solvents for example, by having a large vapor cavity ratio in the vapor cleaning tank.

### 7.12.7 Chlorofluorocarbon

In chlorofluorocarbon recovery, the activated carbon adsorption method is common. With other methods, treatment of the emitted halogen group is necessary. It is also important for vehicle air conditioning units and refrigeration units which include chlorofluorocarbon not to be dismantled outdoors.

In Japan, the major dioxin emission source is thought to be waste incinerators. It has been reported that the ingestion quantity of dioxin in the atmosphere around municipal waste incinerators is 0.9~1.2 pg/kg/day<sup>7)</sup>. As a countermeasure to this, low temperature dust collection, which does not include corona discharge, is efficient. There has been a gradual conversion of municipal waste incinerators from high temperature dust collection with electrostatic precipitators to low temperature dust collection using bag-filters.

## 7.13 Offensive Odor Control Measures

### 7.13.1 Planning for Controlling Offensive Odor Deodorization Method

The basis for planning the odor control measure is to understand the source of the odor, the process of generation, and the conditions surrounding its output accurately. In the event of odor, in most cases it can not be anticipated as to where the odor is generated from. The number of sources and the amount being generated must be thoroughly investigated. And the final step of reducing these amounts are the basis of establishing control measures. In order to determine the amount of odor being emitted, the Odor Emission Rate (OER) is tested. The major sources of the odor are checked and in general, counter measures for sources with large OERs are established initially. The OER is indicated by taking the product of the odor concentration by exhaust gas volume ( $\text{m}^3/\text{min}$ ). The first step of the odor control measure is to investigate the cause of the odor from the source. Then, methods, including ideas on changing current procedures, are looked at to find a way to reduce the amount of the odor. At the same time, in order to lessen the volume of processed gas as much as possible, covers and suitable hood work are done to close off openings. Countermeasures are implemented for stopping leakage from such areas as the facilities. Reduction of the volume of processed gas is a necessity in cutting cost for deodorization units. Following this, deodorization control measures are performed for the odor that is generated. First, the major components of the odor and their concentrations must be comprehended. Then the most appropriate method for deodorization must be selected. In addition to this, should there exist numerous sources, the concentration level of the odor must first be clarified. The odors are then separated based on their concentration level and processed. In this case, it is not enough just to send the odor into a newly set up deodorization device. Treatment methods such as the use of high concentrate odor as auxiliary air for the furnace of the existing facility or for use as aeration gas in water treatment, are being implemented. Fig.7.13.1 gives an example of deodorization method at a raw sewage treatment facility <sup>1)</sup>.

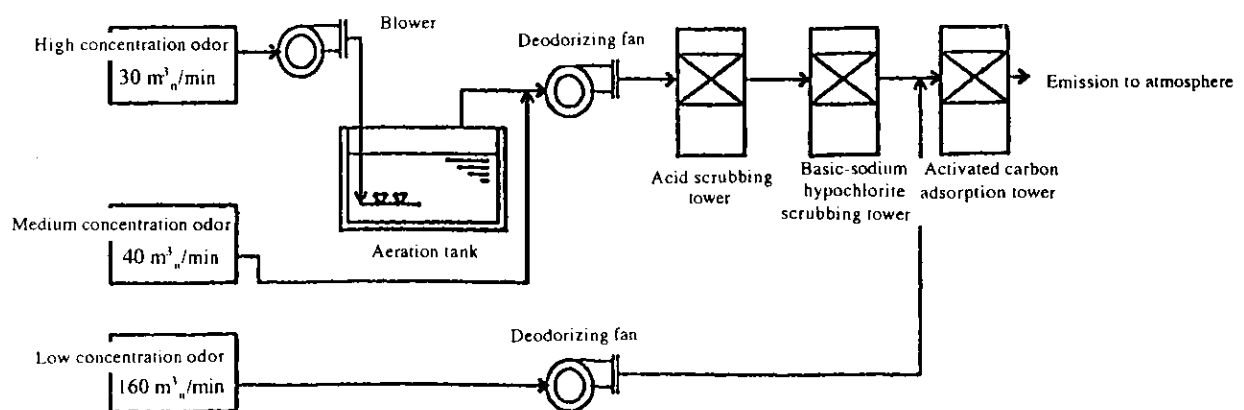


Fig.7.13.1 Examples of Deodorization Method at Night Soil Treatment Facility <sup>1)</sup>

### 7.13.2 Deodorizing technologies <sup>2) 3)</sup>

Methods used for the removal of offensive odor include the thermal combustion method, adsorption method,

wetscrubber method, biological deodorization method, ozone oxidation method, and the use of deodorants<sup>4)</sup>. These methods and their respective devices are selected based upon the physico-chemical qualities of the substance to be deodorized and the conditions of the facilities exuding the odor. The following covers the characteristics of each device and their range of application.

## (1) Combustion method

### ① Direct Burning Method

The offensive odor substance is decomposed via combustion at a high temperature (650-800°C, 0.3-0.5 second retention time) and deodorized. For the combustion process, auxiliary fuels such as city gas and kerosene are used. This is the most dependable method as long as the substance is completely eliminated by combustion. However for those substances which can not be completely oxidized, the method only results in creating toxic substances in addition to the unpleasant odor. This method can be applied for use treating a wide range of odors. As this method utilizes high temperatures, the running cost is high and it is necessary to install a heat recovery unit. It is also effective as a control measure for sources which emit high concentration odors continuously. However, it is difficult to reduce the odor concentration level of the exhaust gas to below 100 after deodorization.

### ② Catalytic Combustion Method

The method deodorizes by decomposing the odorous substances via catalytic combustion. Catalysts such as platinum, vanadium, and manganese are used. Oxidation takes place at fairly low temperatures (300-350°C). For this reason, running costs are low in comparison with the direct combustion method as less heat is used. Preliminary treatment such as with a filter, is necessary as catalyst poison such as heavy metal, silica, halogen, dust, mist, and nicotine effect performance.

### ③ Regenerative Thermal Oxidation Method<sup>5) 6)</sup>

In the regenerative thermal oxidation method, the temperature of the processed gas is maintained at high levels (800-900°C). The offensive odor substances within the processed gas are oxidized and emitted as carbon dioxide or water. The basic mechanism of this method and the direct burning method are the same. This device, after deodorization, passes the high temperature exhaust gas through the accumulator (packed with metal or ceramic thermal storage material). The heat contained by the gas is transferred to the accumulator as it passes through. This heat is used to increase the temperature of unprocessed gas. Deodorization takes place in the combustion chamber. In this case, as the reception of heat is taken directly from the accumulator and due to the fact that there is a sufficient area of contact, more than 90~95% of the heat is recovered. This method is used to deodorize solutions. In cases where the odor is of low concentration and there is a long operating time, the running cost is low.

## (2) Adsorption method

This method utilizes an adsorbent such as activated carbon to adsorb the offensive odor substance. It is a suitable method for situations where the flow of processed gas is relatively large and the concentration of the offensive odor substance is low. It is used as the last step of the deodorizing device as a processing method. The scale of the processing device changes vastly depending on the concentration, humidity, and amount of gas. Also, it is

necessary to select the proper adsorbent for the offensive odor substance. In most cases the adsorption method device is used in conjunction with other devices such as absorption method. High concentrate offensive odors are handled by a separate device, while this device treats relatively low concentrate odors. Units currently being used are ones which exchange the whole adsorbent, recycle the adsorbent it using steam, or the new honeycomb-type method.

### (3) Wet Scrubbing Method

Easily dissolvable substances in the exhaust gas are dissolved using such solutions as water, acid, alkaline, or acidic solution. These substances are absorbed in the solution in order to remove odors. In general, the absorbent solution for acid gas (hydrogen sulfide, etc.) is alkaline solution (sodium hydroxide), that for alkali gas (ammonia, amine, etc.) is acidic solution (sulfuric acid, etc.), and that for aldehyde and ketone is sodium thiosulfate. Basic sodium hypochlorite is used as a oxidizing agent for general organic substances. This method is also adaptable for use in dealing with relatively large flows of processed gas. The initial costs for setting up this system are relatively low but the running costs are effected by liquid waste disposal. Recently, activated sludge has been used as the absorbent liquid. A method is being developed where this is used in combination with the biological deodorization method.

### (4) Ozone Oxidation Method

Ozone, which is a tough oxidizing agent, is used and the offensive odor substances are decomposed by oxidation. In some cases it is used in conjunction with the absorption method or the absorption method.

### (5) Biological Deodorization Method <sup>7) 8)</sup>

Microbes found in black soil, activated sludge, and peat moss are used to decompose the odor substance through a biochemical reaction. This method is used in treating sewage, raw sewage, and organic solvents. This method has a low running cost (0.2-0.8 times that of other methods), is easy to run and manage, and there is no fear of secondary pollution. In order to increase the amount of microbes, it is necessary to obtain a nutritional source and water. For this reason the removal of hydrophilic substances is high. The biological deodorization method is categorized as shown in Fig.7.13.2.

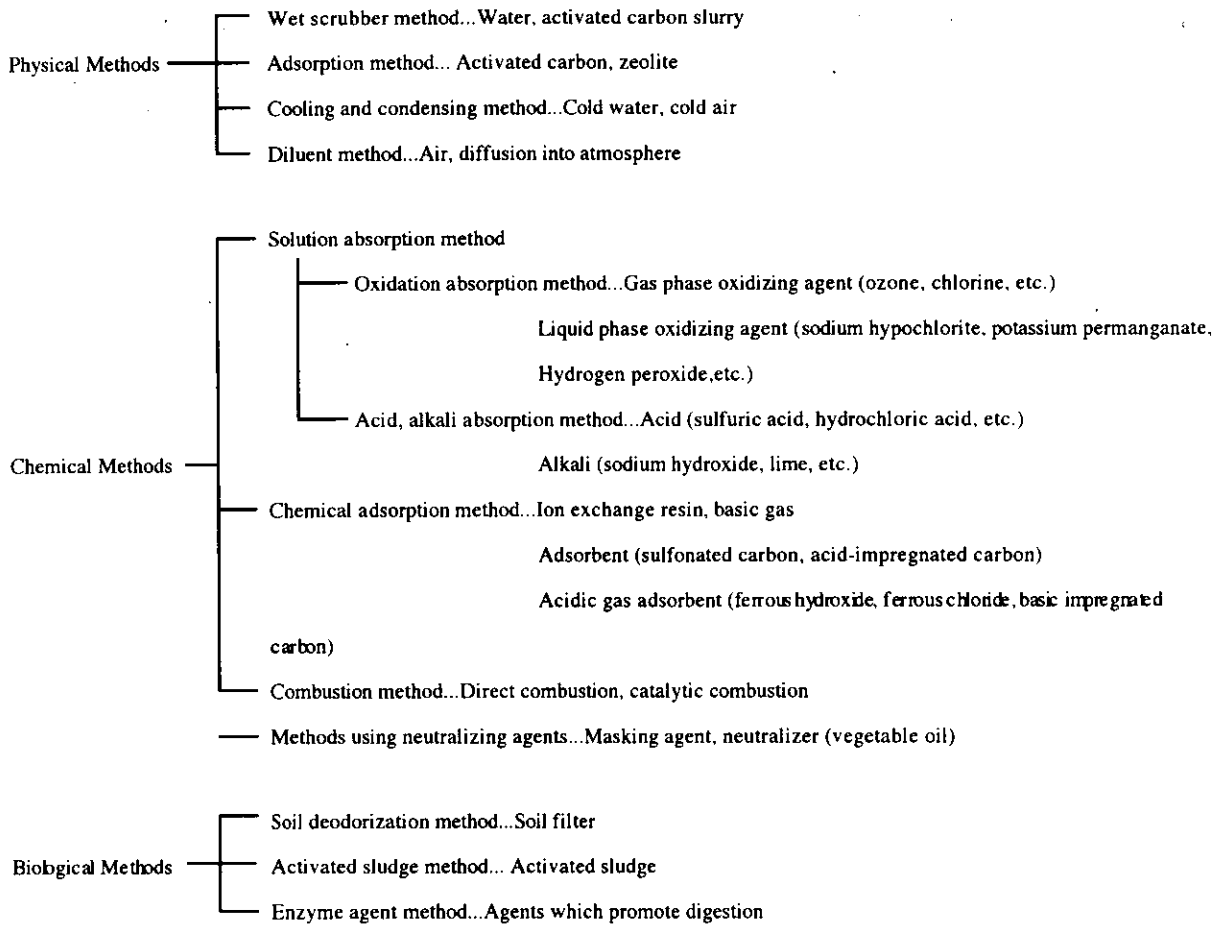


Fig.7.13.2 Categories of Deodorization Methods <sup>4)</sup>



## 7.14 Development Planning and Examples of Countermeasure

### 7.14.1 Overview

In Japan, development of industrial areas on land reclamation along sea sides is promoted and operations on coastal industrial areas are typical characteristics of Japanese industry. Air pollution in these areas and various countermeasures were instituted by stages. Industrial development planning should not be overlooked when considering air pollution problems in Japan.

### 7.14.2 The Negishi Bay Industrial Area Development and the Yokohama Type Agreement on Pollution Control

Negishi Bay is located on the coast south of Yokohama Harbor and led in the fishery industry in olden days. In 1898, Okuda Naohiro applied to the Kanagawa Prefectural governor with a sea reclamation plan for this area. However, the plan put to the Yokohama Board of Aldermen for discussion in 1910, the plan set in front of the Ministry of Home Affairs in 1940, and the International Sightseeing Base Plan in 1951 etc., were all unsuccessful. Then, the "First Stage Plan of Negishi Bay Reclamation Project" was approved in 1957 by the Yokohama Board of Aldermen and was finally begun in 1959. It was the area containing 6,105,000 square meters of land reclaimed for an industrial area gathered with many large factories, during the first and second stages of the project, that air pollution became a problem (Fig. 7.14.1<sup>1)</sup>). The problem materialized, especially, when, in 1964, Tokyo Electric Power Company requested Yokohama City to agree on the construction of a 530,000 Kw coal firing thermal power plant of the Electric Power Development Company, the project forwarded with a coal policy outline decided upon in a cabinet meeting in 1962, in part of the area to be advanced by Tokyo Electric Power Company. Yokohama citizens who had experienced air pollution from coal-firing electric thermal plants were naturally opposed to this. Yokohama mayor Ichio Asukata did not have the administrative authority to regulate air pollutants emission. But he agreed to it under his proposal conditions using two articles contained a land sale contract. The first article was that, in case of reselling or loaning to a third party, the agreement of Yokohama City was required. The second article was that in case of reselling or loaning the reclaimed land to a third party, even after the land was deliver from the city, the agreement of Yokohama City was also required. Namely, Yokohama City closed "gentlemen's agreement" with the Electric Power Development Company, which obligated the to discuss with Yokohama City and carry out pollution control measures as conditions for the Electric Power Development Company to get permission for land use from the city. The city requested Tokyo Electric Power to close a similar contract to operate the first liquefied natural gas firing thermal plant in Japan at this site. This type of agreement on environmental pollution control is known as the "Yokohama Type."<sup>2)</sup> and spread throughout the countryside. Further, Electric Power Development adopted high efficiency electrostatic precipitator with sulfuric acid adding system in flue gas to adjust the electric resistivity of dust investigated by Lodge corporation in Limited kingdom, and promoted a perfect air pollution control measure meeting the specific conditions of regulation on stack height and air pollution concentration in the flue. Also, Nippon Oil Refining Corp.(Negishi oil refinery), which occupies most of the area

of the Negishi Bay industrial area and is called the founder, executed measures such as using high quality, low sulfur fuels, adoption of low NOx burners, the electrostatic precipitator and so on<sup>4)</sup>.

#### 7.14.3 The Yokkaichi Petroleum Complex Plan and the Air Pollution Problem

In 1956, Showa Yokkaichi City Oil Company began the construction on a petroleum refinery with daily volume of 40,000 barrels at the 100 hectare site of the Navy's former No.2 Fuel Arsenal complex on the coast of Ise Bay. Then, the complex began operations by supplying raw materials to a factory group from this in 1957. Mitsubishi Petrochemical Corporation completed the Yokkaichi plant to produce 22,000 tons of ethylene annually in 1959 and Chubu Electric Power Company started operations of the first heavy oil firing thermal power system in Japan at its Mie plant (125,000 Kw) in 1961. Since then, the various factories have been established at the No.1 Industrial Zone as listed in Table 7.14.1. Responding to a plan made by the Ministry of International Trade and Industry, Yokkaichi City in Mie Prefecture reclaimed about 90 hectares of land and, in 1963, Daikyo Oil Company and Chubu Electric Company started operations on this reclaimed land in the No.2 Industrial Area. Yokkaichi City has developed into a preeminent petrochemical industry area in Japan. However, as a symbol of huge advanced economic growth, the complex encumbered with environmental pollution problems such as air pollution. In April 1960, a complaint was lodged by the council of residents of the Shiohama area which neighbors the No.1 Industrial Area of Yokkaichi, requesting to Yokkaichi City to take countermeasures stating that "It is impossible to sleep with the factory noise and gas, even at night," and of "Nausea and irritation from the odor of gas." In October of the same year, a pollution countermeasures section was founded and Yokkaichi City established a pollution control committee. Then, Professor Yoshida Katsumi of the Mie Prefectural Medical College and Professor Mizuno Hiroshi of Nagoya University took charge of the committee, and an investigation of air pollution and its influence on health damage was begun. In March of 1961, the sulfur dioxide concentration in the atmosphere was measured a value per hour beyond 1 ppm with a maximum of 1.64 ppm, and, in the summer of the same year, residents of the Isozu area complained of coughing, throat irritation and intense asthma from the wind coming from the factory. Professor Yoshida reported from his study of these symptoms and concluded that the sulfur dioxide and sulfuric acid mist affected the cilia cells and this could progress to bronchitis, pneumonia and pulmonary emphysema. An anti-pollution movement rose, and in October, the coordinated Yokkaichi council of residents decided to claim an early solution and the improvement of the control equipment. In November 1960, Mie Prefecture and Yokkaichi City began a systematic investigation on air pollution. Their report of 1962 warned that the health damage would increase if the air pollution situation did not improve early in the Isozu area. Professor Yoshida made it clear in his epidemiological survey that more damage to health was observed in the area polluted with higher amounts of sulfur dioxide. According to the starting of operation of the No.2 Industrial Area in 1963, the mayor of Yokkaichi, standing in the forefront of the residents, promised to hit on some countermeasures, but this area was not specified in the Soot and Smoke Emission Regulation Law of 1962, so it was specified by a research by Kurokawa Mission in 1966. However, this administrative action did not bring the improvement. And the construction of high stack was promoted, in order to avoid the pollution under gusty condition that was pointed out by Ito Kyoji, director of Applied Weather Development National Weather Institute member of Kurokawa Mission, but it expanded the polluted area.

In the Shiohama area where respiratory organ diseases increased from around 1961, the council of residents was burdened with the whole medical payments and requested public money. It is believed that some deaths occurred from the air pollution in 1964 and in December of the same year, the mayor of Yokkaichi expressed the intent to appropriate public money. In 1965, the “Yokkaichi Pollution-related Medical Treatment Investigation Meeting” was started and public money for medical payments was made available. After that, as the number of patients continued to increase and with no change in the scope of the occurrence except by going to trial, nine Isozu residents filed suit against six companies in the No.1 Industrial Area in September 1967. The plaintiff’s successful lawsuit in the long trial of four years and ten months produced much administrative reconsideration, and caused a sense of crisis in business circles because their positions on environmental protection were being questioned. A shift came for the Yokkaichi area with a proclamation on the regulation of total sulfur oxide emissions in 1972 called the Mie Prefecture pollution prevention ordinance, and the quantity of sulfur oxides emitted into the atmosphere which was more than 100,000 tons annually decreased phenomenally to 17,000 tons. The mortality rate of the residents from complaints of pulmonary obstruction decreased from the peak of 1975, too. Incidentally, as for the countermeasures against sulfur oxides, exhaust gas desulfurizing equipment accounted for 30% and other methods included conversion to low-sulfur fuel.

Table 7.14.1 The main businesses at the No. 1 Complex at Yokkaichi City

Company Name	Main products at the time
Showa Yokkaichi Petroleum Yokkaichi Refinery	Gasoline, kerosene, fuel oil
Mitsubishi Chemical Industry Yokkaichi factory	Carbon black
Japan Synthetic Rubber Yokkaichi factory	Synthetic rubber
Ajinomoto Tokai factory	Monosodium glutamate
Yokkaichi Synthesis Yokkaichi factory	Surface-active agents
Japanese Butanol Yokkaichi factory	Butanol
Chubu Electric Power Mie thermal power plant	Electric power
Mitsubishi Petrochemical Yokkaichi plant	Ethylene, polyethylene, polypropylene
Mitsubishi Monsanto Yokkaichi plant	Styrene family of resins
Matsushita Electric Works Yokkaichi factory	Heat hardening resins
Mitsubishi Edogawa Chemical Yokkaichi Factory	Hydrogen peroxide
Petrochemical Burdish Yokkaichi factory	Blister polystyrene
Ishihara Sangyo Kaisha Yokkaichi factory	Titanium oxide, agricultural chemicals

#### 7.14.4 The Significance and Air Pollution Counter Measures of the Ogishima Project <sup>7)</sup>

NKK, has been established to have the aim to found the first steel pipe production in Japan in 1912, started blast furnace operations in 1919 at its steel pipe manufacturing plant in Kawasaki, then began operations of the Thomas converter in 1938, prospered and became the leader in the integrated steel work.

The Ogishima project involved the company’s Keihin works on a 3,940,000 square-meter site, producing 5.5 million tons (rude steel) annually through 7 blast furnaces. The site made Kawasaki and Yokohama, Kanagawa Prefecture on the coast of Tokyo Bay the dispersion area. NKK moved the Keihin Steel Mills to a 5.5 million square-meter site on newly constructed Ogishima Artificial Island in Tokyo Bay and concentrated Steel Mills



## 7.15 The cost of air pollution control technology

### 7.15.1 Changes in plant and equipment investment capital

Fig.7.15.1 shows changes in previous years air pollution control countermeasure equipment cost (based on expenditure). During a period of about 30 years from 1966 to 1995 the cost based on expenditure of air pollution control countermeasure equipment was a total of ¥4.7 trillion. The gross total cost in 1990 was ¥6.6 trillion yen (\$46.7 billion, at \$1=¥142).

The cost breakdown was:

- dust collector: ¥2.3 trillion (\$ 16.2 billion)
- flue-gas desulfurization equipment: ¥1.4 trillion (\$ 10.2 billion),
- heavy and light oil desulfurization equipment: ¥880 billion (\$ 6.2 billion)
- flue-gas denitrification equipment: ¥440 billion (\$ 3.1 billion )

Dust collector formed the base of air pollution control countermeasures and covered about one-third of all expenditure. Historically, investment in air pollution control countermeasure equipment has increased since the mid-1960s. At first, the main emphasis was on dust collection and heavy oil desulfurization and then later on high stacks. In the 1970s flue-gas desulfurization commenced and in the mid-1970s flue-gas denitrification was introduced.

### 7.15.2 Sulfur Oxides Countermeasures

In the early 1960s, in Japan's heavily polluted areas direct combustion of crude oil with a small sulfur content was undertaken. From 1967 heavy oil desulfurization technology which removed the sulfur content in crude oil during refining was introduced. Investment in heavy oil desulfurization plant and equipment began in 1966 and in the decade 1967~1976 increased to around ¥585 billion. Through this about 45% of the sulfur content in heavy oil could be removed. With the introduction of heavy oil desulfurization, the sulfur content in heavy oil which averaged 2.6% in 1966, decreased to 1.93% in 1970, and then markedly to 1.43% in 1973.

But due to the technological limits of heavy oil desulfurization in decreasing the sulfur content in heavy oil, it was necessary to introduce flue-gas desulfurization to reach the appropriate environmental standard. Also, at the same time natural gas began to be imported in 1969.

In response, significant investment in plant and equipment for flue-gas desulfurization started in 1970. The annual expenditure was a substantial ¥146.7 billion in 1974, ¥95.2 billion in 1975, and ¥98.0 billion in 1976 and in the early 1980s annual expenditure was ¥30~40 billion. In the 1990s, expenditure increased significantly again as Japan entered a period of reequipping equipment that had been put in place in the 1970s. (refer to Fig. 7.15.1)

In addition, in the 1990s investment in plant and equipment for heavy and light oil desulfurization, which had decreased in the 1970s, began increasing.

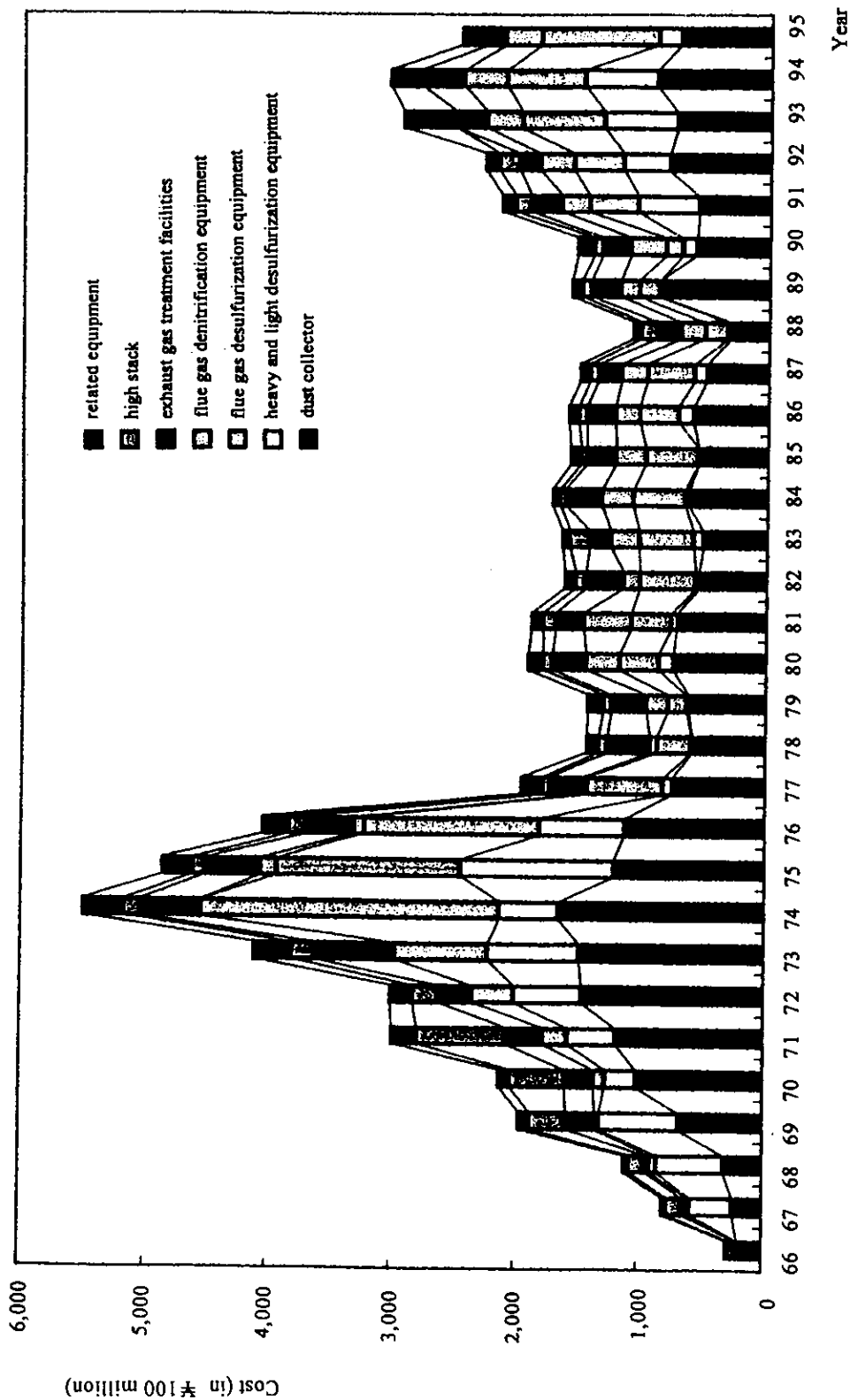


Fig.7.15.1 Changes in the cost of air pollution control countermeasure equipment (at 1990 prices)  
(made from the documents of the Japan Society of Industrial Machinery Manufacturers)

### 7.15.3 The countermeasure cost of individual investment– the example of heavy oil power plant

From the above, it is possible to get an overall understanding of equipment installation costs for sulfur oxides countermeasures from a macro-economic viewpoint. On the other hand, for a micro-economic analysis running costs must be added to construction cost (ie, the cost of plant and equipment investment). For these calculation examples I refer to the ideal model (Nishijima) of a thermal power plant (a heavy oil power plant with a power generation capacity of 300 MW, and a 1million m<sup>3</sup> volume of emitted gas ). If the SO<sub>x</sub> concentration in flue gas is desulfurized from 2,000 to 200 ppm using the lime gypsum method with heavy oil as fuel, the cost is estimated to be ¥2,600 per ton of heavy oil consumed. In 1993 the wholesale price of C-type heavy oil was around ¥19,000 per kℓ and so desulfurization costs were more than 10% of its price. And if the concentration of sulfur content in the heavy oil is set at around 1.5%, the cost of sulfur removal is a high ¥200,000 per 1 ton of sulfur content.

In the trial calculation the total cost of countermeasure costs, including not only flue gas desulfurization but also dust collection and flue gas denitrification, were ¥1.35 per KW of generated power. At current prices the cost of thermal power generation by power plant is around ¥10 per 1 KW, which means countermeasure costs are 13% of the total cost of power generation per KW. However, this is only one example of a trial calculation.

### 7.15.4 The economic influence of sulfur countermeasures

As I indicated above, in Japan when fossil fuel with a sulfur content is combusted the cost must be calculated according to the sulfur content volume and the cost is 10~20% of the fuel price. In macro-economics terms, this is the equivalent influence as an increase in the fuel price. The changes of the wholesale price of C-type heavy oil show a remarkable increase from ¥9,100 per kℓ in 1973, to ¥19,300 in 1974 and a peak of ¥61,400 in 1982. In the 1990s it has been at a lower level of around ¥20,000 per kℓ. The economic influence of the cost of sulfur countermeasures was in large part absorbed by the changes in oil price resulting from the oil crisis.

#### (7.1) Literature cited

- 1) Shinbo, Motoji; Can Production be in Harmony with the Environment pp.165~168, The Nikkan Kogyo Shinbun Ltd (1973)
- 2) Urano, Kohei; The New Environmental Safety Control System of Chemical Substances, Registration of Emissions and Movement of Environmental Contaminant Substances (PRTR), Kagaku Kogaku, 61, 50~53 (1997)
- 3) Kamaya, Shinzen et al (translators); Differentiation of AIDO Modern Chemical History: Two Professional Areas, pp.448~449, Misuzu Shoho Publishing Co, 1979
- 4) Oba, Hideki; Environmental Issues and World History, pp.191~202, Association for Environmental Pollution Control Inc. (1979)
- 5) Tsuchide, Atsushi et al; The Ideas of the Chemist Ryutaro Tsuchida, pp.99~112, Kagakudojin Publishing Company, (1979)
- 6) Himi, Yasuji; Air Pollution Countermeasures in Japan and Other Asian Countries, Inorganic Materials 1, Nov. (No. 253) 67~76 (1994)
- 7) Himi, Yasuji; What Direction should Researchers go in Thinking of the Researchers at the Research Institute of Regional Pollution Examinations From an Engineering Viewpoint., *Journal of Environmental Laboratories Association*, 11, 109~123 (1986)

#### (7.4) Literature cited

- 1) Under the Supervision of the Ministry of International Trade and Industry, Environmental Protection and Industrial Location Bureau. Technology and the Laws and Regulations of Pollution Control (Atmosphere Edition) p.86 (1990)
- 2) Edited by the Energy Conservation Center, Japan.; Energy Conservation Combustion Technique, p.208 (1984)
- 3) Edited by the Energy Conservation Center, Japan.; Energy Conservation Combustion Technique, p.220 (1984)
- 4) Edited by the Energy Conservation Center, Japan.; Energy Conservation Combustion Technique, p.2 (1984)
- 5) Under the supervision of Norio, Arai.; The Emission of Combustion Products and Control Technology, p.417 (1997)

#### (7.5) Literature cited

- 1) Kanagawa Prefecture Government, Environmental Department, 1979 Report on Research; Report on the Emissions of Hydrocarbon Species Substances, pp.6~44 (1980)
- 2) US Department of Health, Education, and Welfare, Public Health Service; Atmospheric Emissions from Petroleum Refineries, A Guide for Measurement and Control (1960)
- 3) Himi, Yasuji; The Current State of Pollution and Countermeasures, (VI), *Gypsum and Lime*, No.125, 31~37 (1973)
- 4) Kanagawa Prefectural Secretariat for Environmental Pollution Countermeasures; Research Report Relating to



## the Vaporization Control of Hydrocarbon Species Substances in Kanagawa Prefecture (1975)

### (7.6) Literature cited

- 1) Under the Supervision of the Ministry of International Trade and Industry, Environmental Protection and Industrial Location Bureau; Technology and the Laws and Regulations of Pollution Control (Atmosphere Edition, 4th edition)
- 2) Industrial Research Center of Japan Inc, Production Goods Work Station; Encyclopedia of Air Pollution Control (1995)
- 3) Iitani, Koichi; New Trends in Filter Dust Collection Technology, *Industrial Pollution Control*, 18, (4), 304~310 (1982)
- 4) Toray Research Center Inc.; New Developments in Gas Filters (1994)
- 5) NTS; The New Technology System for Energy, edited by the Heat Transfer Society of Japan, (1996)
- 6) JISB9909; Ways of Expressing in Manuals for Dust collector
- 7) The Association of Powder Processing Industry and Engineering, Japan (Dust Collection Branch), The Society of Powder Technology Japan (High Heat Dust Collection Group Meeting) 1993 Regular Meeting
- 8) The Society of Powder Technology Japan (High Heat Dust Collection Group Meeting) 1994 1st Regular Meeting
- 9) Kaneoka, Chizuo and Tamori, Ikuo; The Development and Movement of High Temperature Ceramic Filters, *Solids Handling and Processing Industry*, 20, (2), 57~64 (1988)
- 10) Oya Yoshio and Emi Jun and Mori; Early Stage Collection Efficiency of Electric Filters and the Stability of Dust Collecting Operations, Kagaku Kogaku Ronbunshu Collection of Chemical Engineering Essays, 18, (1) 29~36 (1992)
- 11) Nagata, Masayuki; High Speed Electric Dust Collection Equipment, *Industrial Machinery*, 517, 27~30 (1993)

### (7.7) References

- 1) Edited by the Environment Agency; The 1996 Environmental White paper (all theses) p.49 (1997)
- 2) Arai, Mitsubishi; Heavy Industries Technical Review, 27, (4) 303 (1990)
- 3) Ogawa; *Sulfuric Acid and Industry* (9), 161 (1991)
- 4) Ikeno et al.; Ishikawajima- Harima Engineering Review, 30, (3), 183 (1990)
- 5) Yoshii et al., Mitsubishi Heavy Industries Technical Review, 29 (1) 60 (1990)

### (7.8) Literature cited

- 1) Ando, Junpei; Global Flue Gas Purification Technology, The Coal Mining Research Center, Japan, p.170 (1990)
- 2) Takahashi, Yoshiro et al, Mitsubishi Heavy Industries Technical Review, 17 (6) 929 (1980)
- 3) Nakamachi, Ichiro; *Combustion Research*, no.98, 51 (1994)

#### (7.9) Literature cited

- 1) Kido, Nobuo; The Basics of Gas Purification Technology; New Energy Technology for the Environment and Energy Conservation, NTS, pp.395~401 (1996)
- 2) Environment Agency Official Air Pollution Control Division, Air Quality Bureau; The Actual Conditions for setting up Smoke and Soot Treatment Equipment, *Environment and Measurement*, 22, (2) 4~7 (1995)
- 3) Edited by the Environment Agency; The 1996 Environmental White paper (all theses) p.35 (1997)
- 4) Ando, Junpei; World Flue Gas Purification Technology, The Coal Mining Research Center, Japan, p.141 (1990)
- 5) Kido, Nobuo; The Current State of Air Pollution Control Technology in Japan, *Industry and Environment*, (285), 31~34 (1996)
- 6) Electric Power Development Co.; The Dry Process Activated Carbon Method and Simultaneous Desulfurization and Denitrification, Environmental Technology to Protect the Earth, #94 of a 100 piece selection, pp.414~417 (1994)
- 7) Doi, Yoshitaka et al.; A Corroborative Evidence Test of Coal Generated Power Station Flue Gas Treatment by Electron Beam, *Ebara Engineering Review*, (166), 56~64 (1995)

#### (7.10) References

- 1) Kato, Seitaro; General Summary of Pollution Control Countermeasures (Atmosphere Edition) Japan Environmental Management Association for Industry
- 2) Mizushima, Kiyoshi; *Chemical Engineering* (7), 81 (1994)
- 3) Daikin Document, EDX-10b
- 4) Igumo, Masanori; *Pollution and Countermeasures*, 21, (12), 46 (1985)

#### (7.11) References

- 1) Environment Agency, Air Quality Bureau; Environmental Measures towards Automobiles in Japan, July 1996 (1996)
- 2) Japan Automobile Service Promotion Association.; Automobile Exhaust Gas Countermeasures, 1993 Edition (1993)

#### (7.12) Literature cited

- 1) Hirose, Yasuo; Combustion Emission Substances, *Industrial Heating*, 18, (3) 65~71 (1981)
- 2) Tsuji, Masaichi; A General Outline of Pollution Control Combustion Technology, Association of Japan Heat Energy Technology (1973)
- 3) Ando, Tamotsu; Current HC Control in Yokohama City, *Journal of Japan Society for Atmospheric Environment*, 13, 382~386 (1978)
- 4) Kashimura, Hiroaki and Suyama, Yoshiaki and Saiki, Yoshio and Yamamoto, Akio and Nagami, Yasuo; The Emission Volume of Hydrocarbon Species during Automobile Gasoline Refuelling, *Journal of Japan Society for Atmospheric Environment*, 18, 432~438, 1983

- 5) Environment Agency, Air Quality Bureau; Manual for Hydrocarbon Species Emission Control (1981)
- 6) Japan Chemical Industry Association, Emission Control Technology of Offensive Odors and Hydrocarbon Species (2) (1977)
- 7) Environment Agency, Internet Homepage (1997)

#### (7.13) Literature cited

- 1) Kawasaki, Shinichiro and Takishima, Mitsuru; Treatment by Activated Sludge Aerated Deodorisation in Sewerage Facilities, *Journal of Odor Research and Engineering*, 24, 141~145 (1993)
- 2) Ishiguro, Tatsuyoshi; The Basics and Implementation of Odor Countermeasures, Ohmsha Publishing Co, (1997)
- 3) Editorial Committee for the Encyclopedia of Environmental Pollution Control; Encyclopedia of Air Pollution Control, Industrial Research Center of Japan Inc (1996)
- 4) Offensive Odor Law Study Group; New Handbook of the Offensive Odor Control Law, Gyosei Corporation (1996)
- 5) Okada, Hisayuki; Research into the Treatment of Offensive Odor from Emission Gas including VOC by Adtech Thermal Storage-Type Deodorization Equipment, *Journal of Odor Research and Engineering*, 27, 189~191 (1996)
- 6) Yamada, Hideo; The Thermal Storage-type Combustion Deodorization Method, *Journal of Odor Research and Engineering*, 27, 192~199 (1996)
- 7) Osako, Masaharu and Higuchi, Takeshi and Tanaka, Masaru; Technical Appraisal of the Biological Deodorization Method based on Survey Research, (the 3rd), *Journal of Odor Research and Engineering*, 25, 310~313 (1996)
- 8) Morita, Yoichi; Emission Gas Treatment using Microscopic Organisms, General Research Center for Resource and Environmental Technology, the 10th Research Conference Document, 48~53 (1996)

#### (7.14) Literature cited

- 1) Yokohama City; Outline for the Construction of a Coastal Industrial Area in Negishi Harbor, Yokohama City (1965)
- 2) Yokohama City Port and Harbor Bureau Southern Coastal Area Development Department; Reclamation of Yokohama City, pp.40~101, Gihodo Shuppan Co. Ltd (1992)
- 3) Himi, Yasuji; Environmental Technology Transfer to Developing Countries, (1) *Journal of Life and the Environment*, 38, (6) 72~76 (1993)
- 4) Nippon Oil Co. Ltd, Nippon Petroleum Refining Co. Ltd History Editorial Department; A Thirty Year History of Nippon Petroleum Refining, Nippon Petroleum Refining Co. Ltd (1982)
- 5) International Center for Environmental Technology Transfer; Pollution in Yokkaichi City and Steps Towards its Environmental Improvement (1992)
- 6) Yoshida, Katsuki; Air Pollution in Yokkaichi City and Particulars, The 24th Air Pollution Conference Summary, pp.63~73 (1983)

7) Nippon Kokan 70 Year History Editorial Department; A 70 Year History of Nippon Kokan K.K.

**(7.15) References**

- 1) Nishijima, Yoichi; Engineering the Earth's Environment, Japan Productivity Center for Social Economic Development
- 2) Japan Society of Industrial Machinery Manufacturers; A Record of the Actual Sales of Environmental Protection Equipment

## Chapter 8 Measuring method of air pollutants (emission sources)

### 8.1 Measuring and analytical method of air pollutants in exhaust gas

#### 8.1.1 Overview

Measurement of the air pollutants concentration in the exhaust gas from air pollutants sources, contained fine particles such as soot, fume and dust and toxic gasses such as sulfur oxides and nitrogen oxides is extremely important, to obey the air pollution regulation law and ordinance, for the establishment of the air pollution control measures and for the testing on the performance of air pollution control equipments. Moreover, the calculation of air pollutant emission amounts from the air pollutants concentration and exhaust gas flow rate is necessary. The measurement technique of the air pollutants concentration in exhaust gas consist of suction (sampling) of exhaust gas, collection of air pollutants in suction gas, determination of collected air pollutant and measurement of suction gas volume. So, this technique is called “stack sampling” frequently.

The exhaust gasses suction method differs basically between particles floating with inertia in exhaust gas and gaseous air pollutant with no inertia. The reason being that if the flow of suction gas at the tip of suction nozzle of sampling tube have turbulence and appreciable change of flow direction, the particles concentration in suction gas is led to different concentration from actual in exhaust gas. In this regard, JIS-Z8808 on this measuring method prescribe that the measuring position shall be located at a place where the flue gas is under a comparatively uniform flow, avoiding the place where a duct bends or its sectional shape is sharply changed in flue, stack or duct. Because this is also important to measure the exhaust gas flow rate based on measuring results of exhaust gas velocity, JIS-B9905 on measuring method of gasses air pollutant concentration in exhaust gas prescribe in the same way. The particle concentration in exhaust gas is different even within the same cross-section of flue, stack or duct, so it is essential to select multiple sampling points within the same cross-section selected as the measuring position, as shown in Fig.8.1.1, for example. Further, collected particles sample by stack sampling are also used to analyze the heavy metal and polycyclic aromatic hydrocarbons. Measuring methods for the temperature and the static pressure of exhaust gas are prescribed in JIS-Z8808.

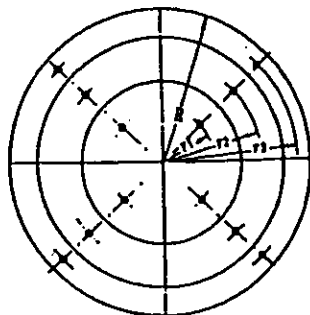


Fig.8.1.1 Measurement points in a circular cross-section of a smoke funnel (in the case of 12 points)

## 8.1.2 Particle concentration measuring method

### (1) Sampling method of particles

In order to archive the quantitative particles sampling in exhaust gas, a sampling tube with suction nozzle whose tip has been completed to an acute angle of maximum  $30^\circ$  shall be prepared, and this is inserted into the flue through the sampling holes at selected sampling position in flue. Furthermore, stack sampling has to be prompted, holding equally the suction gas velocity at tip of suction nozzle of sampling tube and the exhaust gas velocity in sampling point, in order to avoid the inertia effect of the particles in exhaust gas. This operation is called isokinetic sampling, and in not considered necessary when sampling gaseous air pollutants.

There are two methods for isokinetic sampling. One uses an ordinary-type sampling tube that measure the exhaust gas velocity at sampling points by Pitot tube, calculate the suction gas flow rate realized the isokinetic sampling previously, and suck the exhaust gas with this gas flow rate. And another method uses a balanced-type sampling tube that has function to realize the isokinetic sampling, holding equally the static gas pressures of suction gas in suction nozzle and in exhaust gas at sampling point, or the dynamic gas pressure of suction gas in suction nozzle and the static gas pressures in the gas meter of particle sampling apparatus. Furthermore, the particle sampling apparatus consists of the sampling tube with suction nozzle, the particle collector with dust filter, suction gas flow rate measuring apparatus (gas meter) and gas suction pump.

In addition the dust (particle) collector is classified as either type 1 which is inserted into flue, and type 2 which is placed outside of the flue connecting behind suction nozzle.

The type 1 dust collector as shown in Fig.8.1.2 which has cylindrical heat-resistant silica fiber filter inside a glass filter holder to protect the filter inside a glass filter holder to protect the filter is used generally.

The exhaust gas velocity at sampling points is essential to realize the isokinetic sampling, and as a rule, an L-type Pitot tube as shown in Fig.8.1.3 is used frequently.

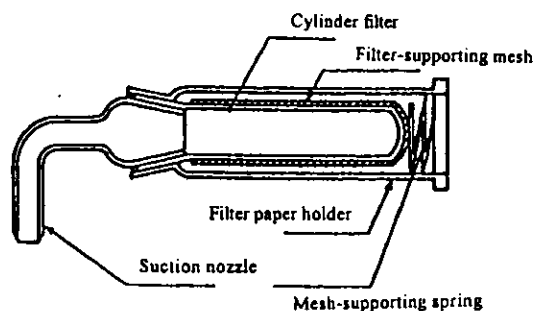


Fig. 8.1.2 Example of dust collector using type I cylinder filter paper

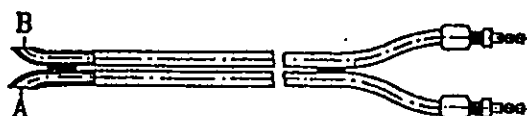


Fig. 8.1.3 Special Pitot tube

In this case, the Pitot tube coefficient has to be calculated previously. The gas velocity can calculate using Eq. (1) to measure the pressure difference between 2 pressure measuring holes center (A), (B) and direction of exhaust gas flow at sampling point as shown in Fig.8.1.4.

$$v = c \times \left[ \frac{2gh}{\gamma} \right]^{0.5} \dots\dots\dots (1)$$

- Where, V: gas velocity (m/s)  
 c: Pitot tube coefficient (-)  
 g: acceleration of gravity = 9.8 (m/s<sup>2</sup>)  
 h: value of pressure difference measured by inclined-tube manometer (mmaq)  
 r: density of exhaust gas (kg/m<sup>3</sup>)

For stack sampling to measure the particle concentration in exhaust gas, the particle sampling apparatus is assembled at sampling position, the sampling tube is inserted into flue, the tip of suction nozzle is turned towards direction of exhaust gas flow and is matched to the location of sampling point, suction nozzle is held parallel to direction of exhaust gas flow, and the stack sampling is started holding the isokinetic sampling operating the particle sampling apparatus. In this case, the filter of dust collector is set for a constant weight and has to be weighed by balance.

During the sampling, the isokinetic sampling operation has to be maintained, and the temperature and pressure in suction gas flow rate measuring apparatus has be measured.

Moreover, for dust sampling, in the “each point sampling method”, the dust filter for particle collection is replaced at every sampling point that has been selected multiple ones, the particle concentrations at each point are determined, and the mean concentration in cross-section of flue is calculated, and in “traverse sampling method”, particle sample is collected during equal time at each sampling point by same dust filter for particle collection, and mean concentration only is obtained.

With later method, it is not possible to obtain a particle concentration distribution for a cross-section of the sampling position of flue, and therefore has no link to the simple sampling method that use the sampling point of representative particle concentration for a cross-section of flue.

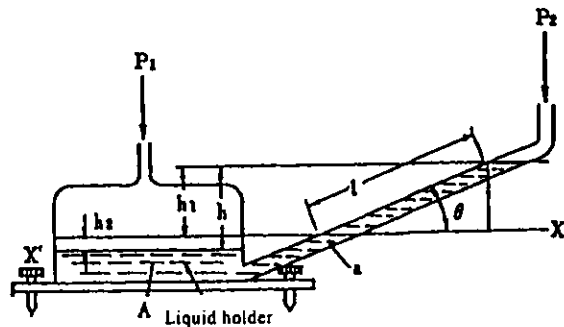


Fig.8.1.4 Inclined-tube monometer

## (2) Particle concentration measurement and calculation method

Following the dust stack sampling operation, dust filter used for particle collection is set for a constant weight again and is determined, and weight of collected particle is determined.

The particle concentration in exhaust (flue) gas shall be expressed by the mass of particle contained in  $1 \text{ m}_N^3$  of dried flue gas ( $\text{g}/\text{m}_N^3$ ) which has been converted into standard condition ( $0^\circ\text{C}$  in temperature,  $101.3 \text{ Kpa}$  { $760 \text{ mmHg}$ } in atmospheric pressure), and it can be obtained from the mass of collected particle, the suction gas volume measured by suction gas flow rate measuring apparatus (gas meter), and the temperature and the pressure at gas meter in stack sampling operation. See JIS-Z8808 for details.

In addition, the particle size distribution in flue gas and the concentration of each particle size ranges can measure using the classifying dust collector as shown in Fig.8.1.5.

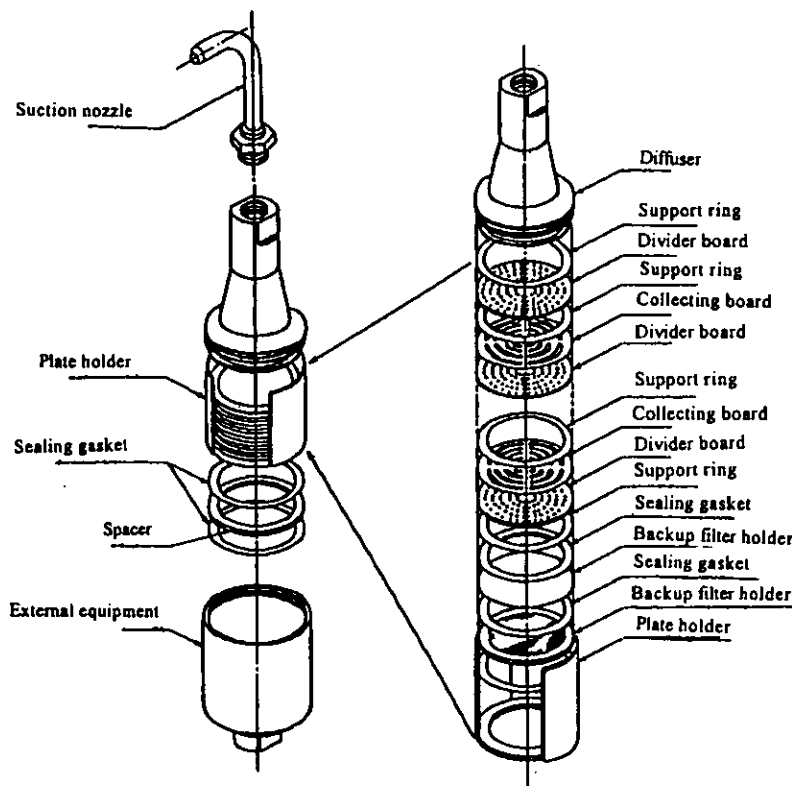


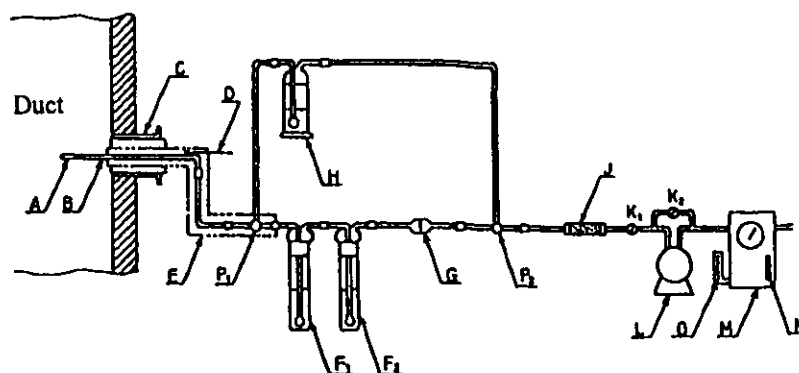
Fig.8.1.5 An example of classifying collector

### 8.1.3 Gaseous pollutants concentration measuring methods

The sampling apparatus used for measuring the concentrations of gaseous air pollutants consists of a test gasses collecting tube, the collecting part for sample gas such as gas absorption bottle, a collection bottle, the section gas flow meter (gas meter), and a suction pump, as shown in Fig.8.1.6. In addition, a filter is fitted into the test gasses collecting tube in order to prevent contamination by particle, into condensates water. Further, in order to prevent the component gas is absorbed, collecting tube is either heated or kept at a high temperature. Moreover, designated quantity absorbing liquid to use absorption of gaseous air pollutants that ought to be analyzed, is added to



in the absorption bottle or collection bottle. For collecting the gaseous air pollutants, a test gasses collecting tube is inserted into the flue, a fixed quantity of sample gasses are sucked using a suction pump, and the target pollutant gas components are absorbing by the absorbing liquid. In this event, if an impinger or other gas absorption bottle is used, the total quantity of suction gas is measured by the gas meter, and both its temperature and pressure, are determined. Further, if a collection bottle for which the contents in a vacuum are known is used, its temperature and pressure are determined. Generally, the test gasses collection tube is inserted into the center of the flue and there is no need for isokinetic sampling. When the collection operation is completed, the gaseous air pollutants absorbed in absorbing liquid in gas absorption bottle or collection bottle are determined, according to prescribed analytical method. And the concentration of gaseous air pollutants can calculate because volume of determined air pollutants can be obtained. In this case, because the determined air pollutant volume under the standard condition can be calculated, it is necessary to convert the suction gas volume of sampling operation to standard condition using the measurement results of temperature and pressing of suction gas in gas meter. See JISKO1303, JISKO104, and JISKO095 for details.



- |                                 |   |                                 |                        |
|---------------------------------|---|---------------------------------|------------------------|
| A                               | : Filter substance                                | J                               | : Dry tube             |
| B                               | : Test gasses collecting tube                     | K <sub>1</sub> , K <sub>2</sub> | : Flow adjustment cock |
| C                               | : Temperature-preserving substance                | L                               | : Suction pump         |
| D                               | : Thermometer                                     | M                               | : Gas meter            |
| E                               | : Heater  | N                               | : Thermometer          |
| F <sub>1</sub> , F <sub>2</sub> | : Absorption bottle (capacity 250 ml or 100 ml)   | O                               | : Manometer            |
| G                               | : Glass filter                                    | P <sub>1</sub> , P <sub>2</sub> | : Three-way flow cock  |
| H                               | : Cleaning bottle (Holds absorption liquid 50 ml) |                                 |                        |

Fig.8.1.6 Example of test gasses sampling apparatus

#### 8.1.4 Calculation of quantities of air pollutant emissions

The quantity of air pollutant emissions is calculated using Eq.(2) from the exhaust gas flow rate and the concentration of pollutants in the exhaust gas. In this case, the concentration of pollutants in the exhaust gas is expressed as the value in dry gas, excluding the water (moisture) content, and the exhaust gas flow rate is determined for wet gas conditions, which includes the water (moisture) content, so that it is necessary to know the flow rate of the dry exhaust gas. For this reason, the water (moisture) content of the wet exhaust gas is calculated using regulation JISZ8808. Moreover, value of moisture content in flue gas is necessary to determine the suction flow

rate to realize isokinetic sampling of the particulate substance in an ordinary type sampling tube.

$$S = 0.06C \times \left(1 - \frac{X_w}{100}\right) \times Q \quad \dots\dots\dots (2)$$

Here,  $S$  = Quantity of air pollutant emissions (kg/h)

$C$  = Concentration of air pollutants in the exhaust gas ( $g/m_N^3$ )

$X_w$  = Water (moisture) content of the exhaust gas (volume %)

$Q$  = Wet exhaust gas flow rate ( $m_N^3/min$ )

Moreover,  $Q$  is determined from fuel a cross-sectional area in sampling position, and the exhaust gas flow velocity at sampling point regulation of JISZ8808 and JISB8222, or calculated from the fuel composition and the main exhaust gas composition.

### 8.1.5 Easy measuring methods of black smoke emission using Ringleman's smoke chart

Ringleman's smoke chart is the oldest measuring method of smoke concentrations emitted from chimneys, and was widely used by heat management engineers. Further, it was also used in the Osaka Prefectural Pollution Control Ordinance, proclaimed in 1932, to prevent air pollution by ban of black smoke emissions exceeding a level 3 on Ringelman's chart over 6 minutes per 1 hour.

As shown in Fig.8.1.7, Ringleman's smoke chart depicts black grids of 0 mm, 1.0mm, 2.3 mm, 3.7 mm, 5.5 mm and complete black on intervals that are complete white, 9.0 mm, 7.7 mm, 6.3 mm, 4.5 mm, and 0 mm on a sheet of white paper 21 cm × 14 cm, creating a Ringleman's smoke chart in which the black sections occupy the white paper in proportions of 0, 20, 40, 60, 80, and 100%, and a visual comparison is then made between the smoke chart and the blackness of smoke emitted from a chimney, which smoke is then classified in six stages from 0 to 5.

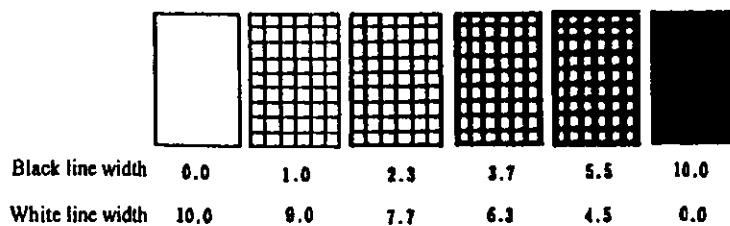


Fig.8.1.7 Ringleman's smoke chart

When measuring, the observer must stand to keep a distance of approximately 40 m from the chimney, and the blackness of the smoke at a distance 30 to 45 cm above the chimney vent is compared with the naked eye, with the chart standing observer's eye level at a location 16 m away. In this case, the measuring position and the flow of the smoke are kept at right angles without ever facing into the sun, and care must be taken that there are no impediments such as mountains, buildings, or trees in the background. Moreover, this method use for grading the concentration of black smoke, although there are various problems in, anybody can make this type of measurement easily.

## 8.2 Methods for measuring and analyzing the main components of combustion exhaust gas (flue gas)

### 8.2.1 Overview

The main components of combination exhaust gas (flue gas) emitted by fuel firing are by carbon dioxide and water vapor, and carbon monoxide, which is formed from the inefficient burning of fuels and the nitrogen and excess oxygen in the air for combination. Of these, the measurement and analysis of carbon dioxide, oxygen, and carbon monoxide have long been considered important for thermal energy management. Further, they are also necessary for calculating the flow rate of flue gas. In addition, in case that the air pollutant emission standards such as dust and soot and nitrogen oxides are prescribed in these concentration base, these are regulated under constant oxygen concentration condition in order to avoid dilution by air. So measurement of oxygen concentration in conduction combination exhaust gas is also indispensable for the judgment of the absence on the air pollutants emission standards according to law and ordinances. The most important items are explained in Table 8.2.1.

Table 8.2.1 Types and principles of gas analyzers

	Measuring method	Name of analyzer	Composition measured
Chemical gas analyzer	Solution absorption reaction	Hempel gas analyzer	CO <sub>2</sub> , O <sub>2</sub> , CO, CmHn
		Orsat gas analyzer	CO <sub>2</sub> , O <sub>2</sub> , CO
Physical gas analyzer	Gas thermal conductivity rate	Electrical CO <sub>2</sub> analyzer	CO <sub>2</sub> , H <sub>2</sub>
	Gas specific gravity	Specific gravity type CO <sub>2</sub> meter	CO <sub>2</sub>
	Infra-red absorption	Infra-red gas analyzer	CO <sub>2</sub> , CO, CH <sub>4</sub> , SO <sub>2</sub>
	Solution conductivity rate	Automatic SO <sub>2</sub> recorder	SO <sub>2</sub>
	Power generated	Zirconium O <sub>2</sub> meter	O <sub>2</sub>
	Current generated	Galvanic cell O <sub>2</sub> meter	O <sub>2</sub>
	Thermal fuel heat	Thermal fuel O <sub>2</sub> meter Inflammable gas meter	O <sub>2</sub> CO+H <sub>2</sub>
	Magnetic type	Magnetic type O <sub>2</sub> meter	O <sub>2</sub>
Separation by column	Gas chromatograph	CO <sub>2</sub> , O <sub>2</sub> , H <sub>2</sub> , CH <sub>4</sub> , SO <sub>2</sub> , NO <sub>2</sub>	

### 8.2.2 Sampling methods of sample gas

There are many cases where flue gas is negative pressure, and it is deemed necessary to use a suction pump in sampling test gasses and, moreover, to ensure that no air becomes mixed into sampling tube. Further, because there are times when the water vapor in the flue gas condense within the sampling tube, sampling tube has to slant and attach the drain eliminator to prevent clogging. Moreover, in order to prevent the contamination of sampling gas with soot or ash contained in flue gas, a filter is attached to the sampling tube.

### 8.2.3 Orsat analyzer

The Orsat analyzer is used widely in the analysis of carbon dioxide, oxygen, and carbon monoxide in flue gas. In the operation of this apparatus, flue gas sample is filled in 100 ml (some analyzer use 50 ml) gas burette containing sodium chloride solution, and transferred to an absorption bottle containing potassium hydroxide solution to absorb  $\text{CO}_2$ . And then sample gas is returned to gas burette once again and its volume reduction due to  $\text{CO}_2$  absorption is read to determine the  $\text{CO}_2$  volume concentration in flue gas. Next, sample gas in gas burette is transferred to an absorption bottle containing pyrogallol/potassium hydroxide solution which absorb Carbon monoxide, in order, and some operations are repeated in order to determine the  $\text{O}_2$  and  $\text{CO}$  volume concentration in flue gas. An outline of which is shown in Fig.8.2.1. Moreover, results obtained from the Orsat analyzer are the volume concentration (%) of the gas components in the dry flue gas.

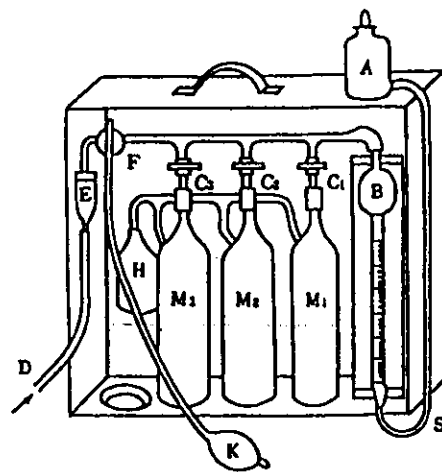


Fig.8.2.1 Orsat analyzer

### 8.2.4 Methods using measuring equipment

Besides the manual methods such as using the Orsat analyzer, continuous analyzers for determining the main component gasses in flue gas are also employed.

#### (1) Electric analyzer for Carbon dioxide

Each gas has its own unique thermal conductivity. And if the assumption that the thermal conductivity of air is 100, can be concluded, or the thermal conductivity of flue gas main component, water vapor is 100, oxygen is 101, nitrogen is 100, and carbon dioxide is 59. Like this, the thermal conductivity of the main component gasses excluding  $\text{CO}_2$  is approximately 100, so  $\text{CO}_2$  concentration in flue gas can analyze by the measurement of flue gas thermal conductivity. Fig.8.2.2 is principle of  $\text{CO}_2$  analyzer used this principle. The air that is standard gas is filled up in cell  $S_1$ ,  $S_2$  and flue gas is flown in cell  $G_1$ ,  $G_2$ . And platinum filament heated by constant electric current is set in each cells and bridge is wired as shown Fig.8.2.2. Because the electrical conductivity of the filament changes depending on the thermal conductivity of the gas in each cells, in this instance the conductivity of the gas

indicator, that is to say, the electrical current, which is related to the concentration of carbon dioxide, is shown, so that the  $\text{CO}_2$  in the fuel can be continually analyzed by this circuit.

(2) Mechanical carbon dioxide analyzer

This analyzer shows the density of gas mechanically. In other word, this  $\text{CO}_2$  analyzer is instrument sown in Fig.8.2.3, adopting the measuring principle that the density of  $\text{CO}_2$  is longer than air. As the figure shows, this device houses two identical winged wheels, one is the air chamber, and another is the chamber passed into flue gas, and wheel turn at the same speed, and the wind pressure due to this is encountered by the winged wheels, which can rotate freely against the pressure. Because a torque proportional to the density of the gas in each chamber is generated against the opposite side winged wheels that encounter the wind pressure, a device that displays the torque proportion makes a record, thereby analyzing the gas density or, in other words, the  $\text{CO}_2$ .

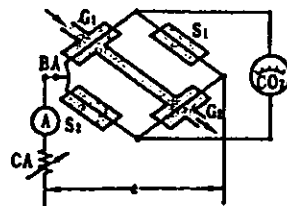


Fig.8.2.2 Principles of  $\text{CO}_2$  analyzer

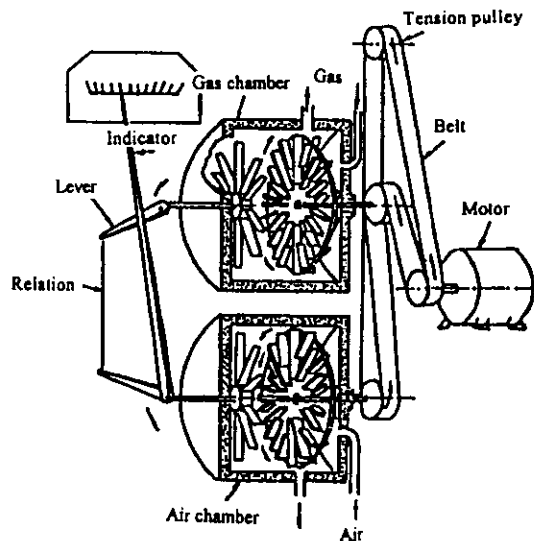


Fig.8.2.3 Principles of mechanical  $\text{CO}_2$  analyzer

(3) Magnetic oxygen analyzer

The magnetic oxygen analyzer uses the attraction force generated when oxygen molecules, which are paramagnetic, are magnetized in a magnetic field to measure the oxygen concentration, which can be classified by both the magnetic wind method and the magnetic strength method. The former heats part of the  $\text{O}_2$  molecules that have been attracted by the magnetic field and detects the strength of the magnetic winds generated by the loss of

magnetism, and the latter classifies the pressure detected as changes in the background pressure of the supporting gasses that continually pour suction in fixed amounts into the magnetic field to work continuously on the oxygen molecules that cycle the dumbbell, to detect the level of deviation in the dumbbell generated by differences in the strength of the magnetization of the oxygen in the test gas.

(4) Electrochemical oxygen analyzer

The electrochemical oxygen analyzer uses an electrochemical oxidation-reduction to continuously measure the oxygen concentration. A pole is fixed at each tip of a zirconium element that has been heated to a high temperature, the test gasses are passed along one end, and air along the other, and by means of a gas permeable membrane and the zirconium, which can detect electrical impulses at both ends, the electrical current of the oxygen, which has been absorbed and diffused in the electrolytic cell is reaction (redox) by the surfaces of the solid electrode.

(5) Infra-red absorption carbon monoxide analyzer

Using an infra-red gas analyzer, the concentration of carbon monoxide in the flue gasses is measured continuously.

## 8.3 Automatic exhaust gas air pollutant measurer

### 8.3.1 Overview

The continuous measurement of the concentration of air pollutants in flue gases is important to obey of the air pollution control law and ordinances, management of air pollutants emission sources. Furthermore, in the regions on regulation of total emission, some factories regulated by this rule have the obligation to measure continuously their air pollutants emission, and there are also occasions where continuous measurement takes place under the Agreement on Environmental Pollution Control.

### 8.3.2 Composition of the Continuous measuring system

The continuous analyzer for air pollutants in flue gas uses a flue gas sampling tube, which is inserted into a flue through which flue gasses flow, as shown in Fig.8.3.1, for example, the sample gasses are introduced to the main unit of the automatic analyzer (the intake part), and the electrical signal corresponding to the concentration of air pollutants that is detected by the main unit's detector part (the analyzer) is recorded by the indicating recorder. And the zero gas and standard gas are used for calibration.

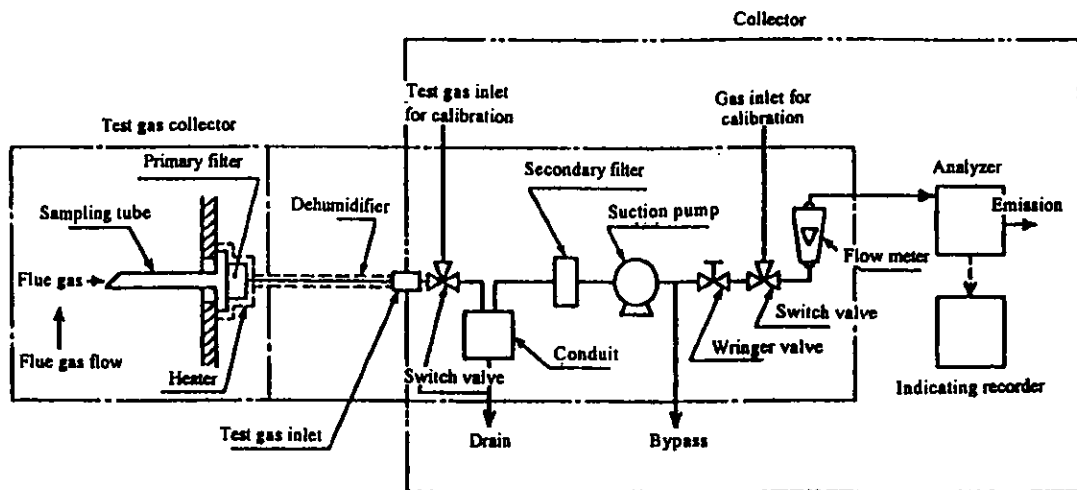


Fig.8.3.1 Example of measurer composition

### 8.3.3 Continuous sulfur oxides analyzer in flue gas

The types of continuous sulfur oxides analyzer in flue gas as shown in Table 8.3.1 are prescribed by JISB7981. Of these, the solution conductivity analyzer is made up of reference electrodes, a gas absorber, an absorbing port solution pump, an absorbing solution tank, an amplifier, a waste solution tank, and so forth, and records the changes of the conductivity due to sulfur oxides that are absorbed by the hydrogen peroxide absorbing solution. Further, the

infra-red absorption system (non-diffusion infra-red analyzer) uses a heated light source which passes a electric current through resistors such as Nichrome wire or silicon carbide, then passes the light through a reference cell that is held in either argon or nitrogen, and a measuring cell through both a revolving sector and an optical filter, converts the differences in the infra-red absorbed into an electrical signal, and records the results. This construction is shown in Fig.8.3.2. Moreover, the ultra-violet absorption system uses either a deuterium discharge tube or a medium pressure mercury lamp, collects the ultra-violet using a spectroscope, and continuously records the absorption of emitted flue gasses.

Table 8.3.1 Types of sulfur dioxide measurers

Types of measurers	Range (1) vol ppm	Notes
Solution conductivity system	0 to 25 } 0 to 2,000	To be applied when the effect of coexisting carbon dioxide, ammonia, hydrogen chloride and nitrogen dioxide is negligible or removable.
Infra-red absorption system	0 to 25 } 0 to 2,000	To be applied when the effects of coexisting moisture, carbon dioxide, or hydrocarbons is negligible or removable.
Ultra-violet absorption system	0 to 25 } 0 to 2,000	To be applied when the effects of coexisting nitrogen dioxide is negligible or removable.
Ultra-violet fluorometry system	0 to 10 } 0 to 1,000	To be applied when the effects of coexisting hydrocarbons is negligible or removable.

Note (1): The values within this range are divided appropriately according to the measurement objectives.

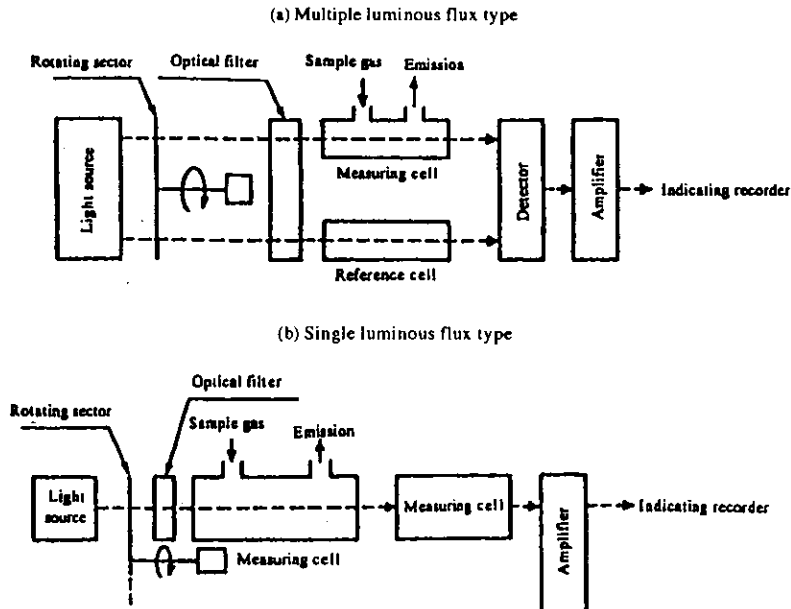


Fig.8.3.2 Examples of continuous infra-red gas analyzer composition

#### 8.3.4 Continuous nitrogen oxides analyzer in flue gas

The systems of continuous nitrogen oxides analyzer in flue gas as shown in Table 8.3.2 are prescribed in



JISB7982. Of these, the chemiluminescence is a practical application of chemiluminescence which ozone reacts with nitrogen oxide in the flue gasses. In other words, this instrument uses the phenomenon that when nitrogen dioxide excited by the electrons generated by this reaction return to ground state, the light across a spectrum 600 to 3000  $\mu\text{m}$  in proportion to the concentration of nitrogen oxides in flue gas is emitted. This construction is shown in Fig.8.3.3.

Table 8.3.2 Types of nitrogen oxides measurers

Types of analyzer	Range <sup>(1)</sup>	Target substances for measurement	Applicable conditions
Principle	vol ppm		
chemiluminescence system	0 to 10 } 0 to 2,000	Nitrogen monoxide Nitrogen oxides <sup>(2)</sup>	To be applied when the effect of the coexisting CO <sub>2</sub> is negligible or removable.
Infra-red absorption system	0 to 10 } 0 to 2,000	Nitrogen monoxide Nitrogen oxides <sup>(2)</sup>	To be applied when the effects of the coexisting CO <sub>2</sub> , sulfur dioxide, moisture, or hydrocarbons can be ignored, or when those effects is negligible or removable.
Ultra-violet absorption system	0 to 50 } 0 to 2,000	Nitrogen monoxide Nitrogen dioxide Nitrogen oxides <sup>(3)</sup>	To be applied when the effects of the coexisting sulfur dioxide and hydrocarbons is negligible or removable.

Note <sup>(1)</sup>: The values within this range are divided appropriately according to the measurement objectives.

<sup>(2)</sup>: Nitrogen dioxide shall be measured by converting it into nitrogen monoxide.

<sup>(3)</sup>: The values given as take value of nitrogen dioxide and nitrogen oxide.

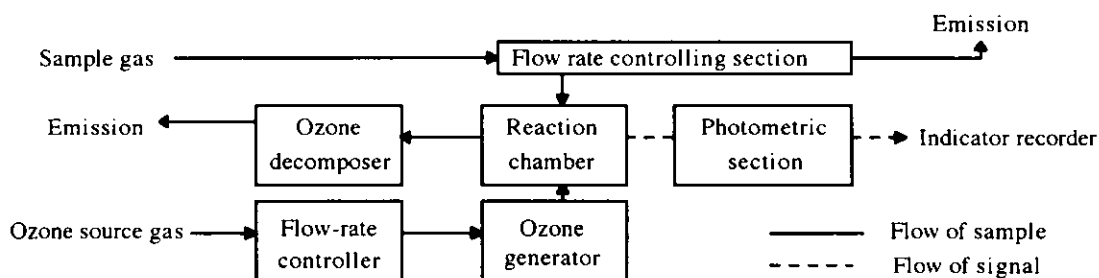


Fig.8.3.3 Example of chemiluminescence analyzer

## 8.4 Fuel testing methods

### 8.4.1 Overview

Fuel experiments present the data that can be used in air pollution control, and are essential in evaluating fuel and in combustion computation. In particular, the analytical results of sulfur in fuel, when firing liquid fuel, are essential in ensuring that emission standards in law and ordinances are being adhered to, because they are directly connected with sulfur oxides emissions from fuel firing facilities. The main points are explained below.

### 8.4.2 Analytical method of sulfur in fuel

As analytical method of sulfur in fuel, dimethylsulfonyl III titration method, microcoulometric titration type oxidation method, neutralization titration method, fluorescent X-ray method and bomb method are used.

In dimethylsulfonyl III titration method, the petroleum or petroleum based fuel sample is burned with oxygen flame, and produced sulfur oxides are absorbed by hydrogen peroxide solution, and the sulfuric acid is determined by titration with barium perchlorate standard solution using dimethylsulfonyl III indicator. The microcoulometric titration type oxidation method is analytical method which conduct the sample fuel into heated combustion tube, and burn it in an oxygen and inert gas flow, absorb the sulfur dioxide generated by combustion in electrolyte, titrate coulometrically and obtain the sulfur content from the quantity of electricity consumed at that time.

In neutralization titration method, the fuel sample is burned with air flow in heated quartz combustion tube, and generated sulfur dioxide is converted to sulfuric acid and it is titrated by sodium hydroxide standard solution.

Fluorescent X-ray method used the no dispersion type fluorescent X-ray apparatus, and in bomb method, the fuel sample is burned with sodium carbonate solution under the atmosphere of high pressure oxygen in bomb, and generated sulfate is determined gravimetrically as barium chloride.

### 8.4.3 Analytical method of nitrogen in fuel

As analytical method of nitrogen in fuel, there is a macro kjeldahl method in which fuel sample is heated and decomposed in concentrated sulfuric acid to which a catalyst has been added, and the nitrogen in fuel compose ammonium sulfate. An alkali compound is added this ammonium sulfate and then ammonia generated by steam distillation operation. Further, it is absorbed in boric acid and is titrated by the sulfuric acid standard.

In micro kjeldahl method, the same operation is repeated, the ammonia generated by steam distillation is absorbed in dilute sulfuric acid, and 1-naphthol solution and sodium hypochlorite is added to induce coloration, then nitrogen is determined colorimetrically at 730  $\mu\text{m}$ .

And microcoulometric titration method, and chemiluminescence method are used.

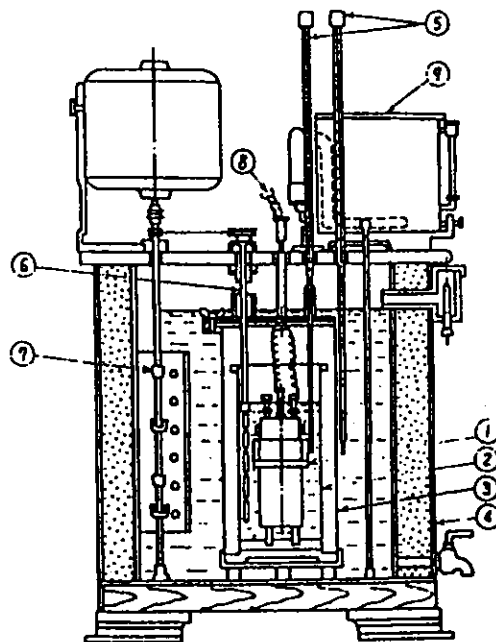
#### 8.4.4 Analytical method of carbon and hydrogen in fuel

The sample fuel is gradually heated and burned at low temperature in an oxygen flow, and the water vapor and carbon dioxide are generated absorbed into both an absorption tube packed with magnesium perchlorate and an absorption tube packed with a powder mixture made by addition of magnesium silicate to molten hydroxide a carrier, and the increased mass of these volume is hereby weighed.

#### 8.4.5 Testing method on Calorific value of fuel

For example, testing method that the fuel sample is burned in bomb of Calorimeter as shown in Fig.8.4.1, Compressed pure oxygen in high pressure, and from the scale reaching of Beckman thermometer, the Calorific value of fuel is calculated is used.

In this case, thermal equivalent of Calorimeter is measured by using the benzoic acid for calibration or international calorific value standard.



- |                         |                                |
|-------------------------|--------------------------------|
| ① Bomb                  | ⑥ Stirrer for use inside pipes |
| ② Inside cylinder       | ⑦ Stirrer for use externally   |
| ③ Intermediate cylinder | ⑧ Ignition fuse                |
| ④ External wrapping     | ⑨ Heated water tank            |
| ⑤ Beckman thermometer   |                                |

Fig.8.4.1 Example of a calorimeter

## 8.5 Air Pollutant Control Equipment

### 8.5.1 Overview

Testing methods of air pollution control equipment such as dust collector, desulfurization equipment, and denitrification equipment, and so forth, requires the technology to test for the design, maintenance, and capability guarantees, as well as being indispensable for research and development<sup>1)</sup>. This explanation centers on JISB99909<sup>2)</sup>, which stipulates expression of specification for dust collector, and JISB9910<sup>3)</sup>, which stipulates method of measuring performance of dust collector.

### 8.5.2 Dust collector testing methods<sup>1) 2) 3)</sup>

#### (1) Expression of the Specification for dust collector<sup>2)</sup>

Dust collector specifications are expressed, named, and modeled by 17 common items, and as specific items are cited as two items of gravity dust collector, four items of centrifugal force dust collector, five items of inertia dust collector, cleaning dust collector, and filtration dust collector each, and seven kinds of electrical dust collector. The examples most commonly cited are the common items and the specific items or centrifugal force (cyclone), cleaning (somer), filtration (bag filter), and electrical dust collector (electrostatic precipitator), which are in popular use.

##### (I) Common items

- ① Classification, ② Name and model, ③ Application, ④ Flow of dust collecting system, ⑤ Raw gas flow,
- ⑥ Raw gas temperature, ⑦ Raw gas pressure, ⑧ Raw gas nature, ⑨ Nature of dust, ⑩ Dust concentration in inlet duct of dust collector, ⑪ Dust concentration in outlet dust, ⑫ Dust collecting rate (efficiency), ⑬ Pressure loss, ⑭ Dimensions, ⑮ Dust emission type, ⑯ Thermal insulation, ⑰ Other.

##### (II) Specific items

- ① Cyclone force dust collector-i. Diameter that is basic for centrifugal force and fundamental gas velocity, ii. Cyclone step rate and number, iii. Presence or absence of a dust (particle) box.
- ② Scrubber-i. Kind of fluid (fresh water or sea water, or another type of fluid), ii. Fluid quantity (washing fluid quantity and pressure, additional fluid quantity and pressure, fluid holding quantity), iii. Fundamental gas velocity, iv. Gas/fluid separation method, v. Other.
- ③ Bag filter-i. Filter surface area and fundamental gas velocity, ii. Filter material (type, model, dimensions, number of bag cylinders, individual number, operating temperature range), iii. Shaking device, iv. Other
- ④ Electrical precipitator-i. Arrangement of collecting chambers, ii. Cross-sectional shape of collecting electrode, dimensions, number of ducts, duct pitch, iii. Cross-sectional shape of discharge electrodes, dimensions, number of electrodes, iv. Capacity of changing, number of equipment and changing system, rectification method, control method, hammering device, etc.

#### (2) Dust collector testing methods<sup>1) 2) 3)</sup>

##### (I) Test items

The test items of performance of dust collector are selected appropriately from the following items, having considered the test objectives.

① Raw gas temperature, pressure and composition in inlet and outlet ducts of dust collector, ② Raw gas flow, rate in inlet and outlet duct, ③ Pressure loss, ④ Dust concentration in the inlet and outlet ducts, ⑤ Dust flow rate in the inlet and outlet ducts, ⑥ Dust collection rate (efficiency) or penetration rate, ⑦ Specific gravity and particle diameter distribution of collected dust and dust in raw gasses within both the inlet and outlet ducts, ⑧ Apparent specific electric resistivity of same, ⑨ Water quantity used for dust collector and liquid gas ratio, ⑩ Waste water discharge quantity and quality, ⑪ Dust collector motive power consumption, ⑫ Noise of dust collector operation.

#### (II) Measuring methods and principles

The measurements for each performance test item are, as a rule, implemented during a stable period of operating conditions for both the dust collector itself and the air pollutant source installed dust collector. In this case, if there is a periodicity, the measurements are made across a lengthy period of time corresponding to one cycle. Moreover, as was described previously in (I) ①~⑦, subjects measured at the inlet and outlet ducts of dust collector have to be measured simultaneously, and the measuring positions within the duct and measuring points within the ducts cross-section are selected according to JISZ8808 regulations. In this case, the measuring position are selected as close to the dust collector unit itself as possible. Generally, the measuring position aims to be a position through which the gas flow within the duct can be regulated, and the principle is to avoid changes to the shape of duct cross section, but in maintaining the dust collecting efficiency such as in the bag filter and electrostatic precipitator, where it is necessary to regulate the flow of gas within the unit, it is possible to select a measuring position close to the unit itself. Further, in equipment that raw gas spins within the duct, such as cyclone, the ideal situation is to take measurements having installed a flow regulating grille within the duct. Moreover, details of these measuring methods are to be found in Chapter 8.1 and the related industrial standards (JISZ8808, JISB9910).

#### (III) Measuring methods of the temperature, static pressure and composition of raw gas in inlet and outlet ducts of dust collector

The temperature is measured using either an electrical thermometer or a glass envelope thermometer at each measuring point as described in Chapter 8.1, and prescribed by JISZ8808<sup>4)</sup>. Static pressure uses either a Pitot tube or a static pressure tube at each measuring point, and measurements conform either to JISZ8808, or to the methods described in Chapter 8.1. Moreover, in the case that static pressure measurement results in wall of duct obtained at static pressure pores installed right and left side on duct wall are equal, there is no objection to using the mean value as this measurement result. The Orsat analyzer is used for the gas composition, and the measurements conform either to JISZ8808, or to the methods described in Chapter 8.2.

#### (IV) Measuring methods of raw gas flow rate in the inlet and outlet ducts

The raw gas flow rate is measured by JISZ8808 or description in Chapter 8.1, using Pitot tube, or using an orifice, which is regulated by JISB8330. In this case, the gas flow rate can be expressed by wet gas flow rate at temperature and pressure within the inlet and outlet ducts, dry gas flow rate according to same condition, wet gas flow rate according to, temperature 0°C, pressure 1 atmosphere, dry gas flow rate according to same condition, wet gas flow rate according to specified condition, and dry gas flow rate according to same condition.

(V) Calculating methods of pressure loss

Pressure loss is expressed as the difference between the total mean pressure of the raw gas in the dust collector inlet and outlet ducts, and is calculated according to Eq. (1).

$$\Delta p = (p_{ii}) - (p_{io})$$

$$(p_{ii}) = \frac{p_{ii1} v_{ii1} + p_{ii2} v_{ii2} + \dots + p_{iim} v_{iim}}{v_{ii1} + v_{ii2} + \dots + v_{iim}} \dots \dots \dots (1)$$

$$(p_{io}) = \frac{p_{io1} v_{io1} + p_{io2} v_{io2} + \dots + p_{ion} v_{ion}}{v_{io1} + v_{io2} + \dots + v_{ion}}$$

Here,  $\Delta p$  : Pressure loss (mmAq or kgf/m<sup>2</sup>)

$(p_{ii})$  : Total mean gas pressure in the inlet duct (mmAq or kgf/m<sup>2</sup>)

$(p_{io})$  : Total mean gas pressure in the outlet duct (mmAq or kgf/m<sup>2</sup>)

$p_{ii1}, p_{ii2}, \dots, p_{iim}$  : Total gas pressure at each measuring point in the inlet duct (mmAq or kgf/m<sup>2</sup>)

$v_{ii1}, v_{ii2}, \dots, v_{iim}$  : Gas velocity each measuring point in the inlet duct (m/sec)

$p_{io1}, p_{io2}, \dots, p_{ion}$  : Total gas pressure at each measuring point in the outlet duct (mmAq or kgf/m<sup>2</sup>)

$v_{io1}, v_{io2}, \dots, v_{ion}$  : Gas velocity each measuring point in the outlet duct (m/sec)

(VI) Measuring methods of dust collection and dust flow rate inlet and outlet ducts of dust collector

These measurements are prescribed by JISZ8808 or described in Chapter 8.1. In this case, the dust concentration, in the bag filter outlet, is measured using a high volume air sampler in equipment that is not equipped with an outlet duct, such as an open model bag filter. In this case, the raw gas flow rate necessary for calculating the dust flow rate determines the blown up area measured using a portable fan anemometer, which measures the mean gas flow speed of the area blown up within the equipment.

(VII) Calculation methods of dust collection rate (efficiency) or dust penetration ratio of dust collector

① Determining method from dust flow rate within the inlet and outlet ducts

The dust collection rate (efficiency) is calculated using Eq. (2). Moreover, when  $Q_o = Q_i$ , Eq. (3) can be used.

$$\eta = \left(1 - \frac{S_o}{S_i}\right) \times 100 = \left(1 - \frac{C_o Q_o}{C_i Q_i}\right) \times 100 \dots \dots \dots (2)$$

$$\eta = \left(1 - \frac{C_o}{C_i}\right) \times 100 \dots \dots \dots (3)$$

② Determining method from dust flow rate within the inlet duct and collecting dust quantities per unit time in the dust box

In this case, the dust collection rate (efficiency) is calculated using Eq. (4).

$$\eta = \left(1 - \frac{S_e}{S_i}\right) \times 100 \quad \dots\dots\dots (4)$$

③ Determining method from dust flow rate within the outlet duct and collecting dust quantities per unit time  
 In this case, the dust collection rate (efficiency) is calculated using Eq. (5).

$$\eta = \left(1 - \frac{S_c}{[S_c + S_o]}\right) \times 100 \quad \dots\dots\dots (5)$$

④ Penetration rate of dust collector is calculated using Eq. (6).

$$p = 100 - \eta \quad \dots\dots\dots (6)$$

Here,  $\eta$ ; Dust collection rate (efficiency) (%)

$S_o$ ; Dust flow rate within the outlet duct (kg/h)

$S_i$ ; Dust flow rate within the inlet duct (kg/h)

$S_c$ ; Quantity of collecting dust per hour in the dust box (kg/h)

$C_o$ ; Dust concentration in the outlet duct ( $\text{g}/\text{m}^3$ ,  $\text{g}/\text{m}_N^3$ )

$C_i$ ; Dust concentration in the inlet duct ( $\text{g}/\text{m}^3$ ,  $\text{g}/\text{m}_N^3$ )

$Q_o$ ; Gas flow rate within the outlet duct ( $\text{m}^3/\text{h}$ ,  $\text{m}_N^3/\text{h}$ )

$Q_i$ ; Gas flow rate within the inlet duct ( $\text{m}^3/\text{h}$ ,  $\text{m}_N^3/\text{h}$ )

$p$ ; Penetration rate (%)

(VIII) Measuring method of quantity of collecting dust per hour in dust box

The quantity of collecting dust and the collecting time are measured, and calculated using Eq.(7).

$$S_c = \frac{M}{t} \quad \dots\dots\dots (7)$$

Here,  $M$  =Amount of collecting dust by dust collector during whole time of dust sampling in the duct (kg).

$t$  = Operating time of dust collector during whole sampling time in duct (h).

(IX) Measuring method of the apparent specific electric resistivity, particle diameter distribution, and specific gravity of the dust sample in the dust collector

The specific gravity for the collecting dust and the dust within the inlet and outlet ducts of the dust collector is measured using either JISZ8807 specifications, or a air comparison type pycnometer. And the particle diameter distribution is measured using either a sieriing method prescribed by JISZ8801, or a sedimentation method or microscope method prescribed by JISZ8901 and JISA1204, and is expressed as a mass basis. Moreover, the apparent electric resistivity is measured using either a paralleled disk electrode, needle-disk electrode, coaxial-cylinder

electrode and pectinated electrode method.

(X) Measuring method of quantity of used water and discharged water

The used water quantity and the discharged water quantity are both measured using either a float type area flow meter as prescribed by JISZ8761, a throttling flow meter prescribed by JISZ8762, a Venturi tube type flow meter as prescribed by JISZ8763, or by Wein, as prescribed by JISK0102.

And liquid gas ratio is calculated by Eq.(8)

Water quality is analyzed by JISK0102.

$$L = \frac{q_w}{Q_i} \dots\dots\dots (8)$$

Here,  $L$  = Liquid gas ratio ( $\ell / m^3$ )

$q_w$  = Used water flow rate ( $\ell / h$ )

$Q_i$  = Wet gas flow rate in the inlet duct of dust collector ( $m^3/h$ )

(XI) Measuring method of power consumption of dust collector

① Power consumption due to pressure loss can be obtained using Eq.(9)

$$P = 0.273 \times 10^5 \Delta p Q_i \dots\dots\dots (9)$$

Here,  $P$  = Power consumption (kw)

② Power consumption of fan motors, motors equipped in dust collector and so forth is measured by integrated. The dust collector noise is measured using JISZ8731 regulations. Moreover, the measurement method of fan noise is prescribed by JISB8330.

8.5.3 Desulfurization and denitrification equipment for flue gas testing method

These are the same as for the dust collector test methods cited previously. In this case, the concentration of dust, including within both the inlet and outlet ducts, are designated as sulfur oxides concentrations and nitrogen oxides concentrations respectively. And in the case of the desulfurization equipment, the collecting dust quantity can regard as the quantity of by-product gypsum. Further, an equipment flow sheet based on the dust collecting system is also necessary.



## 8.6 Exhaust Gas Measurement for Motor Vehicles

### 8.6.1 Introduction

Concomitant with the introduction of weight regulation for motor vehicle exhaust gasses in 1973, a 10 mode exhaust gas measurement was established and, moreover, in 1994, regulations on the particulate matter (PM) from automobiles was added to this, and methods of measuring hazardous substances from automobiles were formalized.

### 8.6.2 Measuring equipment overview

Equipment essential for measuring motor vehicle exhaust gasses are equipment that recreates motor vehicle road running conditions (power absorption and control), exhaust gas sampling system, gas analyzers, and data processor.

#### (1) Power absorption and control equipment

Having set the conditions for the test vehicle or engine, it is driven, and the equipment that absorbs the motive power generated is called a dynamometer. The dynamometer is a system comprising a combination of various control equipment and peripherals. The power absorption system under actual vehicle conditions is called a chassis dynamometer, and a power absorption system for the engine alone is called an engine dynamometer.

With the chassis dynamometer, a roller that rotates in place of the road is taken to be an infinite level road, and a motor wheel is set on the roller, enabling motive experiments to be conducted. An example of a typical system is shown in Fig.8.6.1. In this case, running resistance when actually driving on the road is added to the roller axle from the dynamometer, and the inertial resistance is added by flywheels, closely resembling actual driving conditions on the road. Normal running resistance is added to the fixed level road running resistance. Running resistance at low speed is fixed regardless of the speed, but is magnified at high speeds. Air resistance increases proportionately with the speed of the vehicle. Power generated by the test vehicle on the chassis dynamometer is for the most part absorbed by the dynamometer, but during both acceleration and deceleration, is also absorbed by the flywheel, or else partly as mechaloss (mechanical loss.)

The engine dynamometer also absorbs power from the engine, and as a control mechanism is functionally the same as the chassis dynamometer. The basic concept of the engine dynamometer is an engine mounted on a test-bench, and to transmit to the dynamometer for absorption either directly or via a propeller shaft the power generated when the engine is running, by driving the actuator through commands from the operators console and the control board. Depending on the measurement objectives, various devices can be added, but for transient operation such as in urban test drive mode, flywheel and brake apparatuses are attached, on-the-road driving conditions are recreated in the same way as the chassis dynamometer, with road resistance being provided by the dynamometer, and inertial resistance during both acceleration and deceleration being provided by the flywheel. With the engine dynamometer, because the output from the engine is transmitted directly to the dynamometer, compared with the chassis dynamometer, there is no mechanical loss or tire slippage, and as a means of measuring engine out put,

highly accurate results can be expected. Further, because the settings conditions allow for high mechanical accuracy, recreation is also good, and continuous running for long periods of time is also possible, therefore being optimum for examination of the measuring methods, endurance tests for exhaust gas reduction systems, and so forth. The engine dynamometer does not even need to be in the test room with the vehicle, enabling a compact system to be designed, and making effective experiments possible.

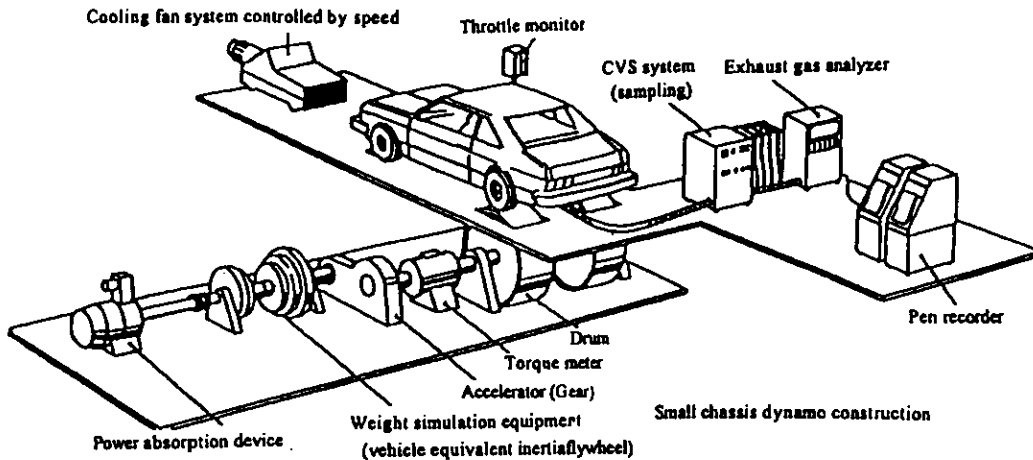


Fig.8.6.1 Gas emission measuring equipment.

(2) Exhaust gas sampling equipment.

A. Direct gas sampling

Problems with the direct sampling of automobile exhaust gasses are that pollution of the sampling process route by oil and black smoke, control of the water content included in large quantities of thermal exhaust gasses, and the dissolving of water which is either condensed or adhering to the tube walls of the hazardous substances content tube. Oil and so forth, which cause the pollution of the sampling process route are caught by an oil catcher, and the black smoke is extracted by a heated filter, after which the gasses are introduced into the analyzing system. As for the water content in the exhaust gasses, because there is a danger of measurement of the hazardous substances being sabotaged, measures are taken to condense and extract the water using an electron cooler. For the measurement of adhesive pollutants, flow pipe must be heated. The HC heated induction tube is heated to 190°C, and NOx heated induction tube is heated to 60°C. In addition, as a problem specific to direct gas sampling, there is the issue of “hangup” (the phenomenon whereby HC in the exhaust gas is first adsorbed by the measuring path, and then later becomes unstuck.) In order to prevent this kind of phenomenon, the tube is made from non-adhesive Teflon, and in addition to selecting a sampling system in which the part that comes into direct contact with the gas has a small surface area, backflush (air is flushed from the secondary filter backwards to the exhaust gas inlet) is also carried out, with the intention of dislodging any HC which has been adsorbed.

B. Dilute gas sampling

As a method of solving the problems of direct sampling and accurately obtaining the concentration and quantity of hazardous substances in driving, where the transient operation are subject to violent change, the CVS (Constant

Volume Sampling) method, which is a method of dilute gas sampling, has been created. It has been used in America since 1972 as the official method for regulating motor vehicle exhaust gasses, and Japan has also adopted the 10-mode regulation as the official method since 1973. In 1983, the European Community (EC) reviewed its total back method, which has been used up till then, in favor of the CVS method.

The CVS method collects all the exhaust gasses and dilutes the clean air (the dust are extracted by micro-glass fiber, and the HC by charcoal filter) (10 to  $10^2$  times), by means of which, the total quantity of exhaust gas and diluted air is sucked by a blower as though it were always the same fixed amount. There are two blower sucking type, PDP (Positive Displacement Pump) and CFV (Critical Flow Venturi). With the PDP method, by operating a Roots Blower at fixed rotations, a combined quantity of constantly fixed exhaust gas and dilution air is sucked. The CFV method uses a principle whereby a fixed flow is achieved when the gas is pushed to the speed of sound, by which means, it is possible to control the flow by selecting and setting the Venturi to correspond the flow required.

In this way, in the CVS method, the total flow of the exhaust gas and the dilution air becomes constantly fixed, and the concentration of hazardous substances within that total flow is proportionate to the weight of the exhaust. Consequently, the gas to be measured is collected in a sample bag as a fixed proportion of the flow from the diluted exhaust gasses, and after the driving mode has been completed, when it is measured in an analyzer, the quantity of hazardous substance emissions from the weight base of the test vehicle can easily be determined. However, because there are many times when pollution of the dilution air becomes a real problem, the dilution air also needs to be collected for analysis in a separate sampling bag, and then subtracted as the background value.

The substance of the bags used in this method is TEDLAR (fluoride vinyl resin PUF50  $\mu$ ), with a capacity of 150 to 200  $\ell$ , and a sampling quantity of 5 to 10  $\ell$ /min. The CVS method dilutes the exhaust gasses with air immediately after they emerge from the muffler, and this system instantly heats the gasses to room temperature, so that because there is no need to install a low temperature trap to eliminate water content in the collection system, there are the advantages that the loss of water-soluble gasses can be avoided, there is little change in the composition due to mutual reaction with highly active chemical substances because of the dilution, and no HC hang-up need be considered either. However, when measuring HC in diesel exhaust gasses, because there is a risk of adsorption by the HC bag, the diluted exhaust gasses are continuously introduced to, and measured by, the analyzer as they are, without being replenished in the bag, and the concentration quantified, by which means the amount of HC exhaust in the test mode can be determined.

### C. Dilution tunnel

The construction of a dilution tunnel used in sampling particulate matter in motor vehicle exhaust gasses is shown in Fig.8.6.2. As the figure shows, the dilution tunnel system can be largely divided into four: the main dilution tunnel, the second tunnel, the PM sampling equipment, and the CVS equipment.

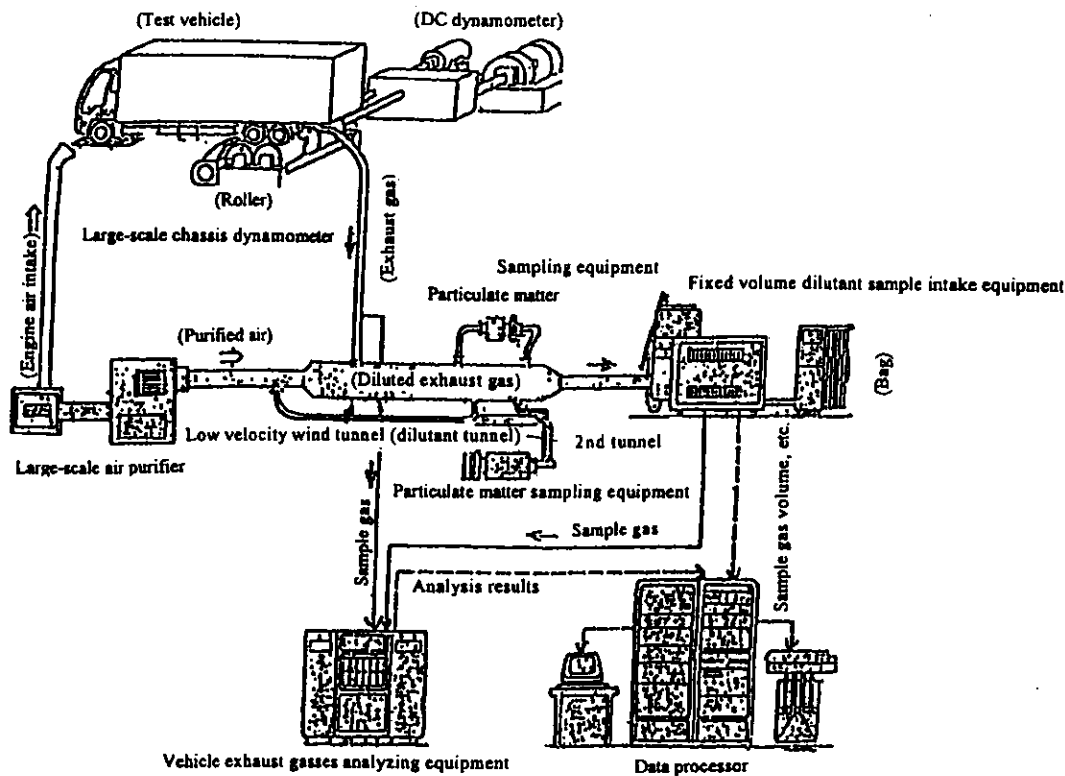


Fig.8.6.2 Dilution tunnel construction

### (3) Exhaust gas analyzer

When black smoke and PM are separated from the motor vehicle exhaust gas components, the composition of the gas that is targeted for analysis are NO<sub>x</sub>, HC, CO, which are the targets of regulation in many countries, and O<sub>2</sub>, CO<sub>2</sub>, and so forth, which are necessary the fuel burning condition and the fuel consumption of an engine. When the component gasses of motor vehicle exhaust are analyzed in this way, the basic functions to be determined are sensitivity, linearity, the response time, interference effects, and stability (drift), and depending on the measurement objectives for each, the method of analysis has to be selected. Table 8.6.1 shows at a glance the measurement principles for each measurement item as determined by Japanese law <sup>2) 3) 4) 5)</sup>.

Depending on the motor vehicle exhaust gas measurement targets (inspection, certification, research, etc.), and the target vehicle (separated by gasoline and diesel), the analyzer measuring items that ought to be used, the measuring principles, the sampling methods, and so forth, are selected, between two and ten analyzers are assembled, and a continuous motor vehicle exhaust gasses measuring device is thereby built. Table 8.6.2 shows the measuring range, accuracy, response time, etc., at a glance for each measurement item. In this device, there is a temperature display for each point on both the heated and maintained temperature paths, and a warning circuit operates whenever there is a cooling unit malfunction, water circulation malfunction, heated line temperature malfunction, or a hydrogen flame ignition or drain level malfunction.

On the other hand, the exhaust gas measuring equipment used in spot checks for automobile repairs, which check that the exhaust standards in the vehicle processes are being adhered to during use, require different

specifications for the continuous analyzer. At times, such conditions as lightweight compactness, cheap running costs, and simple operation with short heater time are required for use in on-road inspections. The CO/HC meter, which uses the NDIR method, is a device that fulfills these conditions.

There are also emission standards for the concentrations of black smoke from diesel vehicles, but the laws on measuring black smoke concentrations in Japan use reflective smoke meters. In this method, part of the exhaust gasses from the exhaust pipe are sampled using a suction pump, and those gasses are then passed through a filter paper and a fixed beam of light trained on one black smoke spot generated on the filter paper, and the light reflected back is collected by a photoelectric element and, as the filter paper that determines the smoke concentration uses a tape system, when one measurement is completed, it is transferred automatically.

Table 8.6.1 Measuring principles determined by Japanese law

Type of vehicle		Mode name	NOx	CO, CO <sub>2</sub>	HC
Vehicles under GVW 2.5 t	Gasoline vehicles	10·15 Mode	CLD	NDIR	FID
	Diesel vehicles	10·15 Mode	CLD	NDIR	HFID
Vehicles GVW 2.5 t or over	Gasoline vehicles	Gasoline 13 mode	CLD, NDIR	NDIR	FID
	Diesel vehicles	Diesel 13 mode	HCLD, CLD, NDIR	NDIR	HFID

Note: CLD: Chemiluminescence detector, NDIR: Non-dispersive infra-red detector, FID: Hydrogen flame ionization detector, HCLD: Heated Chemiluminescence detector, HFID: Heated hydrogen flame ionization detector.

Table 8.6.2 Specifications for each analyzer

Item	CO (Hi)	CO (Low)	CO <sub>2</sub>	HC	NO	THC	NOx	O <sub>2</sub>
Measuring principles	NDIR	⇔	⇔	⇔	⇔	FID	CLD	Magnetic
Measuring range	5,000 ppm	10 ppm	1%	100 ppm	500 ppm	10 ppm	10 ppm	5%
	↓ 10%	↓ 3,000	↓ 20%	↓ 1,000	↓ 6,000	↓ 5,000	↓ 5,000	↓ 25%
Range no.	4	4	4	4	4	9	9	3
Reproducibility	Within ±1% of full scale							
Zero drift	Within ±1% of full scale/8 hours (at temperature range ±5°C)							
Span drift	As above (as above)							
Linearity	Within ±1% of full scale							
90% responses	1.5 to 3.0 sec.					1.5 sec.	1.5 sec.	2.0 sec.

#### (4) Data processor

This is a computer that calculates the measurement results and controls the analytical and measuring equipment. Depending on the test conditions, it can instruct the analyzer and the CVS equipment according to its preprogramming, and in addition to controlling the measuring system as a whole and operational management of the overall measurement system, it can record measurement data for the emitted gas concentrations and engine operation state, and convert units and calculate concentrations and weight and, as need arises, output its recordings either on paper or on a CRT screen.

### 8.6.3 Procedure for testing motor vehicle exhaust gas

#### (1) Preparations for exhaust gas testing

- A. Selecting a vehicle: Selected by considering measures against exhaust gas taken, market share, distance traveled, etc.
- B. Collecting on-road running resistance data: Implements on-road running resistance data measurements in order to set running resistance loads in the chassis dynamometer. Typical measurement methods include methods for determining a standard test vehicle (measuring air intake pressure corresponding to the speed of the test vehicle), and methods for determining a standard chassis dynamometer (measuring coasting data during on-road driving.)
- C. Fixing the test vehicle: Affixing a motor wheel to the center of the roller, and fixing the vertical wheel with a Y-shaped clamp, a wire, etc. By connecting the test vehicle muffler to the CVS gas emissions collecting tube, all the measurement devices (boost meter, fuel consumption meter, air intake flow meter, engine tachometer) can be attached.
- D. Adjusting the measuring equipment: Set the load and chassis dynamometer warmed up, set the analyzer and CVS equipment warmed up, and calibrate the analyzer.

#### (2) Setting the start conditions

Features of the exhaust gas of vehicles considerably vary at the onset of driving and compared with the driving state after ward. Consequently, exhaust gas tests also yield differences in the measurement results between the hot start method, in which the experiment is begun after the engine has been sufficiently heated, and the cold start method, in which the experiment begins immediately after the engine is started. In the case of the cold start, because the quenching zone widens due to the effects of the cold on the cylinder wall, when compared with the hot start method, the emissions of HC and CO are far greater. In order to bring the catalytic converter and its functions into play requires engine warm up time, and no matter what, immediately after beginning, the reduced effects of hazardous substances is weakened. However, in hot start experiments, it is possible to achieve a stable test condition in a short space of time (speed 60 km/h in 15 mins.), but in the cold start experiments, a long time is required to achieve a stable condition in all parts such as the test vehicle's cooling water, lubricating oil, transmission, and differential gear, if left at room temperature. The cold start experiment soak time is prescribed as being a minimum of 12 hours in America, and six hours in Japan.

#### (3) Test vehicle operation

##### A. Unsteady driving

If the test driver is driving under unsteady conditions, the vehicle can be operates as if following the test drive chart that is displayed in real time on the driver's aid CRT screen. However, the allow able error of time and speed in this case is determined as being speed  $\pm 2$  km/h, time within  $\pm 1$  second, and therefore, retesting is necessary for measurements that are repeated many times outside of the tolerance ranges. Public laws that determine driving modes in small vehicles such as gasoline-fueled passenger cars, use unsteady mode experiments in many countries. The urban driving pattern that is used as basic data for setting the emission factor for calculating total emissions of

hazardous substances in the region also use unsteady driving factors.

#### B. Steady driving

Standard test mode of gross vehicle weight of 2.5 t or higher combine the load and the engine revolutions speed to measure exhaust gas at each point. In this case, because engine control using the chassis dynamometer is technically difficult, generally, tests are conducted using an engine dynamometer. Engine control under steady conditions can either be manual (by dial settings) or commands can be entered from the control board. In either case, the vehicle driving mode is step shape, and exhaust gas measurements are taken in line with this mode. In the case of steady driving, Japan continues to use the hot start, but America uses the cold start. 13 Mode, which is suitable for heavy vehicles of GVW 2.5 t or more is also considered in Japan to be steady driving.

#### (4) Exhaust gas measurements

Exhaust gas measurements are either diluted gas measurements or direct gas measurements. In unsteady test conditions, generally, the diluted gas method is used. Part of the exhaust gas are diluted using CVS equipment, and used to fill a bag, then through a mixing process in the bag, the test modes are averaged out, and the concentration of harmful substances in the bag, and the emissions of hazardous substances from the quantity of diluted gas absorbed by the CVS equipment are calculated. The sampling quantity in the bag is set at 5 to 10 l/min., but in urban driving pattern, the sampling quantity must be adjusted depending on the length of the measurement time. The chain of measuring operations such as the filling, analyzing, exhaust, and purging of diluted gas emissions is automatically controlled by computer. If exhaust gas measurements under steady conditions are carried out using the direct gas method, fuel consumption and air intake must also be measured at the same time. When PM is collected with an exhaust gas temperature within the main dilution tunnel at 51.7°C or more, the operators switch to using the second tunnel, but in addition to the condition of exhaust gas temperature, in exhaust gas tests for vehicles in which the concentration of PM within the exhaust gas is high, the requirements may exceed the suction pump's capabilities, and there may be no choice but to use the second tunnel. Filters that have finished collecting PM are measured on a pair of scales after soaking within the range of 1 to 80 hours, and the PM emissions determined.

#### (5) Black smoke measurements

Black smoke standards for new vehicles in Japan target the black smoke concentrations at revolutions speed of 40%, 60%, 100% at maximum output under full load conditions. Consequently, when conducting tests, either a chassis dynamometer or an engine dynamometer must be used. Black smoke measurements when the vehicle is being used are adapted to suit the loadless rapid acceleration mode.

#### 8.6.4 Indicated problems

Motor vehicle exhaust gas emission measurements require major equipment, and it is not the case that just any organization can carry them out. When the fact that measurement data is as yet insufficient is considered, it is hoped that measurement facilities will be quickly built and completed. In particular, there is an urgent need to establish methods for measuring and understanding hazardous chemical species from motor vehicle.

#### (8.1) References

- 1) Japanese Standards Association; JIS Handbook 10, Environmental Technology, JIS (1996)
- 2) Environmental Agency, Planning and Co-ordination Bureau, Research and Co-ordination Division supervised; Notes on Environmental Measuring and Analytical Methods, Japan Environmental Measuring and Analysis Society (1984)
- 3) Japan Society of Air Pollution ed.; Air Pollution Handbook (1) Measurement Collection, Corona Ltd. (1975)
- 4) Himi, Yasuji; Practical Dust and Soot Measurements, Japan Thermal Energy Technology Association (1974)
- 5) JISB8330
- 6) JISB8222

#### (8.2) References

- 1) Japan Standard Association; JIS Handbook 10, Environmental Technology, JIS (1996)
- 2) Japan Society of Air Pollution; Air Pollution Handbook (4) Combustion, Corona Ltd. (1979)

#### (8.3) References

- 1) Japan Standard Association; JIS Handbook 10 Environmental Technology, JIS (1996)
- 2) Standard Association.; Air Pollution Handbook (1) Thermal Fuel edition, Corona Ltd. (1975)

#### (8.4) References

- 1) Japan Standard Association; JIS Handbook 10, Environmental Technology, JIS (1996)
- 2) Japan Standard Association; JIS Handbook 12, Petroleum, JIS (1983)
- 3) Japan Society of Air Pollution; Air Pollution Handbook (4) Combustion edition, Corona Ltd. (1979)

#### (8.5) Literature cited

- 1) Himi, Yasuji; Practical Dust and Soot Measurements, Japan Thermal Energy Technology Association (1974)
- 2) JISB99909
- 3) JISB9910
- 4) JISZ8808

#### (8.6) Literature cited

- 1) Air Quality Bureau, Environment Agency; Motor vehicle Environmental Pollution Control in Japan, Sep. 1996.
- 2) Horiba CO., LTD Zero Emission (Challenge to the Zero Emission), 1993
- 3) Road Transport Bureau, Ministry of Transport; Technical Standard for 13 Mode Exhaust Emission Measurement for Diesel Powered Motor Vehicles, Nov. 1993.
- 4) Road Transport Bureau, Ministry of Transport; Technical Standard for 10-15 Mode Exhaust Emission Measurement for Gasoline Fueled Motor Vehicles, July 1991.
- 5) Road Transport Bureau, Ministry of Transport; Technical Standard for 10-15 Mode Exhaust Emission



Measurement for Diesel Powered Motor Vehicles, July 1991.

- 6) Road Transport Bureau, Ministry of Transport; Technical Standard for 13Mode Exhaust Emission Measurement for Gasoline Fueled Motor Vehicles, Sept. 1992.

## Chapter 9 Air pollution measuring methods (atmosphere)

### 9.1 Basis of air pollution measurement

#### 9.1.1 Introduction

The first step in air pollutant concentration is to define clearly the objectives of measuring concentrations of air pollutants, and to determine the target substances for measurement. The objectives that are usually intended are as follows. ① To thoroughly understand the pollution levels and concentration fluctuations when the sources and pollutants are clearly known; ② When neither the source nor the pollutants are clearly known, to examine the causes of the pollution; ③ When the source is identified, to thoroughly understand the levels of pollution and the types of pollutants emitted; ④ When the pollutant has been identified, to specify the source and to determine the level of its contribution, ⑤ To easily understand border area concentrations and exposure sites for voluntary periods of time; ⑥ To thoroughly understand the broad range pollution level and its fluctuations.

Measurements are taken using collection-analysis (manual analysis), concentration meters, continuous (automatic) analyzers, and the other simple measuring equipment. Manual analysis is applicable to most of compounds that can be used with ordinary equipment such as spectrophotometers and gas chromatographs, because once has been collected at fields, it has to be carried back to the laboratory for analysis. The concentration meter obtains on-site values in a portable form for the target gas. The simple measuring equipment is inexpensive and easy to operate, and aims to be able to take measurements on-site. The continuous analyzer is used for fixed point observations, and records both the time course and the mean concentrations. In recent years, on-site analytical values have been obtained, and portable continuous analyzers have also been on the market, but the basic patterns of air pollution measurement are these four.

Of the above objectives, ② and ③ require accurate analysis manually, while ① and ⑥ require continuous analysis, and ⑤ uses simple measuring equipment. For the ④ objectives, both manual and continuous analysis are used contiguously. Even with expensive measuring equipment, if the selection of the equipment and the operation employed are wrong, accurate environmental information will not be obtained, whereas even with inexpensive equipment, the above objectives can be attained, depending on the approach. If a specific measuring pattern is selected appropriately, rational measurements are entirely possible.

#### 9.1.2 Measurement methods classification and sampling

There are many different analysis and detection principles that can be used in the above four measuring patterns<sup>1)2)</sup>. Fig.9.1.1 shows them ranged in order. These selections are made while considering the sensitivity of the target substance, the selectivity (interference), accuracy, economic viability, ease of use, and so forth. Each measurement pattern is explained with reference to the next paragraph. In addition, there is the laser remote sensing method, which can measure both airborne aerosols and gas across a wide area directly, without the need for sampling.

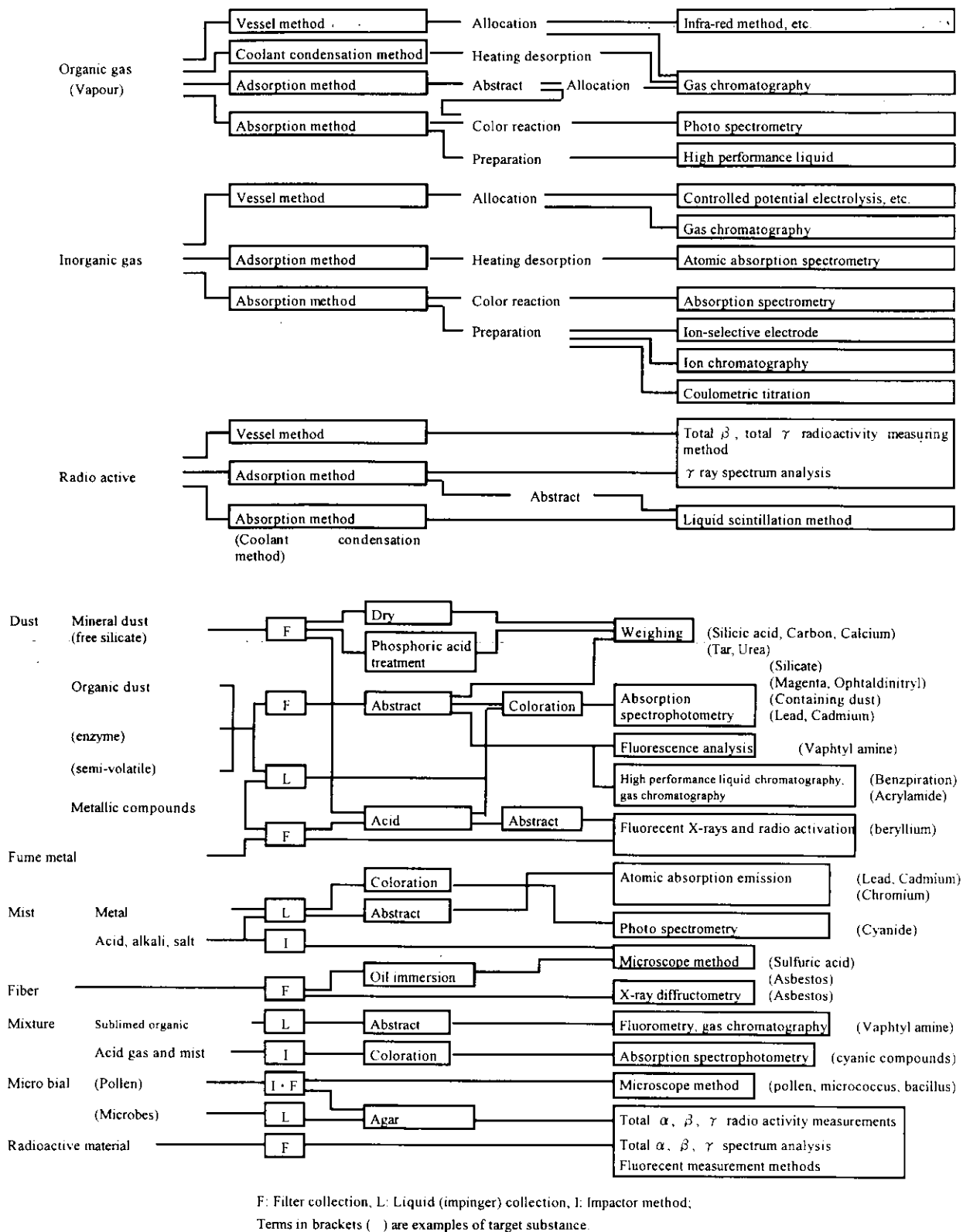


Fig.9.1.1 Measuring pattern of various kinds of pollutants using manual methods<sup>3)</sup>

In environmental measuring, except for the aforementioned remote sensing, because only part of the air is taken as a sample, sampling is essential so that both spatial and temporal representative values might be obtained. In

order to learn the pollution level for a given region, samples are taken at points so as to obtain mean concentrations and sites for which the highest concentrations can be expected: ground level of noxious substances emitted from specific chimneys and the side of major roads. On the other hand, because the concentration is governed by wind speed, climate, day of the week, and season, sampling is performed with reference to conditions that indicate both the highest and the lowest concentration in order to obtain an overview of the pollution level with only a small sample, or else measurements are taken continuously throughout the year. Moreover, in order to determine the annual mean concentration, it is necessary to take samples twice a month, each sample being over a 24 hour period.

9.1.3 Manual analysis methods

This pattern is used for nearly all pollutants (Fig.9.1.1), and the state in which they are introduced to the analyzing equipment is nearly always either liquid or gas. At this point, the concentrations of the samples in the air are determined by Eq. (1) (except for vessel collection), so that the quantitative concentration range is determined not only by the sensitivity of the analyzers, but also by the sample air volume.

$$C = \frac{M}{V} \dots\dots\dots (1)$$

Where, *C*: Concentration, *M* : Target substance amount (Detection amount × Collection quantity / Injection quantity), *V*: Sample air volume (Aeration flow rate × Time)

Consequently, in order to measure lower concentrations with the same equipment, it is necessary to increase the sample air volume within a range that does not affect accuracy. Measurement accuracy [in addition to operational errors and analytical instrument accuracy] are governed by collection efficiency and the measurement accuracy of sample air flow rate. For this reason, a balance between them is important, and if reasonable collection conditions are unclear, it is necessary to verify the collection efficiency and then, correctly connect a integrated flow meter, and start the collection. The collection efficiency by both the concentration and the flow (or gas meter) rate. This can be verified by connecting up the two collector in series. It is important to be careful here, as the flow meter (including the rota meter and hot wire types) can be easily affected by contamination in the measuring line.

When analyzing air pollutants manually, both spectrometry (absorption spectrophotometry, atomic absorption spectrometry, and ICP emission method), which is widely used, and chromatography (gas chromatography and liquid chromatography) are given in detail as examples as follows.

(1) Spectrometry

The responses (relative values of absorption and emission strength) for the sample solutions and the standard solutions are obtained using a spectrophotometer. The calibration curve is usually prepared for the determination. The responses are plotted as the Y-axis and concentrations of five types or more of the standard solution as X-axis. Which are prepared by diluting the proto standard solution (standard substances dissolved in solvents, and sometimes

available on the market). Using the usually linear relationship, the amount of the target substance in the samples determined from the sample solution responses, parallel with which, the blank values (the smaller the better) are determined from responses to the unused absorption solution and the extract from the filter paper, the difference between the being the amount of the target substance. The causes of error of the process are thought to be ① operating error (injection of the sample solution into the analyzer and transfer of the specimen of the collected target substance to the sample solution) and the analytical instrument and ② error due to the reproducibility of the equipment itself and ③ interference from coexisting substances. Against the ③ causes of error, we are to use specific collecting solutions, to select the spectrometer conditions (wave length of the spectrometer and atomization atmosphere, emission temperature, etc.), and to pretreat the sample appropriately, but these are not perfect to avoid the interference. On the other hand, the standard addition method is effective in correcting the negative and systematic error.

## (2) Chromatography

Chromatography, representative differential analytical method, is to identify over a retention time of the chromatogram peak, and then determine over the peak area. Both gas chromatograph and liquid (ion) chromatograph analyzers are composed of a separating column, a detector and a carrier introducer, and a thermostat. In order to use one, it is essential to set the temperature, the length of the column, internal diameter, packing, species of stationary liquid, type of detector, flow rate and species of carrier gas, (composition of carrier solution), sample injection volume, and other analysis conditions. In particular, the column conditions govern the degree of separation, and the detector the sensitivity and selectivity. The standard substance is injected under the set conditions, and the peak retention time is measured. Unknown samples of many kind of volatile organic compound should be identified in parallel with a mass spectrometer because the peak positions may well overlap. The atmospheric concentration is normally low, so that by collecting as shown in Fig. 9.1.1 and infusing either the whole sample collected or part thereof, the response of the volume of the control substance and the sample can be compared and calculated as per Eq. (2).

$$C = \frac{\left(\frac{\ell}{\ell_0}\right) \times \left(\frac{P_0}{P}\right) \times m}{V} \dots\dots\dots (2)$$

Where,  $C$  = Concentration;  $v$  = Sample volume,  $v_0$  = Injection volume,  $P$  = Response for standard substance,  $P_0$  = Response for sample;  $m$  = Amount of standard substance,  $V$  = Volume of air sample.

The linearity of the calibration curve must be confirmed once in the concentration range used. Moreover, the blank values are measured in determination with the pretreatment, such as the concentration operation. The error factors, such as collection conditions, including the collection rate, and the injection operation, the reproducibility of the equipment and so forth, are the same as for the spectrometry.

Computer software with data logger, or built-in computer data processors are usually used for chromatogram data processing, but manual calculation with a pen recorder is essentially the same. Rather, it is essential to

monitor such analogue data as the peak shape and so forth, and never be remiss in checking and evaluating the analytical results.

The reproducibility of the peak position (retention time) is characteristic in the chromatography to effect on errors of the identification. Because the carrier flow rate and the temperature govern the peak position and area, they must be checked after setting a stable condition. Further, over long periods, the column suffers from deterioration due to the flow out of the stationary phase liquid (gas chromatograph) and the contamination (liquid chromatograph), so that the retention time gradually changes. The retention time and area of the standard must be checked once every few days during continuous use, as well as when the equipment is first to be used each time. Moreover, the air sample collected in the vessel can be used to check the preservability, and the active gas should be collected to be changed into stable components.

#### 9.1.4 Concentration meter

Both gas concentration meters and total suspended dust concentration meters are used. Gas concentration meters introduce the air sample to the measuring equipment, and thereby measure the concentration of the target substance directly. For coexisting substances, there is a sufficient selectivity factor (response for the target gas/response for the coexisting gas), and if the detection limit is less than 1/10 of the environmental standard, the detection is very efficient. However, the concentration meter has limits to its selectivity and accuracy, therefore especially in measuring the approximate concentration level of the coexisting gases, which interfere positively with the measurement, so it is essential to check in advance the extent of the interference due to an accumulation in the concentration and reciprocal selection factors.

The meter is set to zero passing zero gas that does not contain the target component, and sets the sensitivity controlling tweezers so that the index value fits in that of a standard gas. The frequency of adjustment depends on the principles involved. For example if the principle is photometric, regular checks by the manufacturer are few and far between, and if the principle is electrochemical, the checks must be carried out each time the equipment is used. The precision is expressed as the coefficient of variation and the zero drift per unit time. Dust concentration meters are also relative concentration meters, which introduce the standard dust flow to the concentration meter, and thereby adjust the sensitivity, and neither principle experiences much in the way of fluctuation in sensitivity during normal operation, so that regular checks by the manufacturer alone are sufficient to used it.

#### 9.1.5 Continuous analyzers

Continuous analyzers are automatic devices that continuously introduce the air sample, and record the output either continuously or with intervals. Such devices have standard gas generators built-in, and introduce the standard gas in place of the air sample on average once a day, check the sensitivity, and adjust it as necessary, so that the equipment involved usually tends to be rather large. The accuracy is expressed in the same way as for the concentration meters.

## 9.1.6 Measurement accuracy and measurement management

### (1) Measurement accuracy <sup>4)</sup>

This section recounts detection limits and determination limits which depend upon measurement accuracy. Taking the detection limit ( $C_d$  in Fig.9.1.2) to be the threefold value of the standard deviation ( $S_b$ ) of the response (mean value  $R_b$ ) of the blank sample is also recommended in a IUPAC report (Fig.9.1.2.) The  $R_b$  and  $S_b$  in Fig.9.1.2 are determined by the blank tests of 10 to 20 times. Around the detection limit, the coefficient of variation is extremely huge. Fig.9.1.2 also shows the relation between the "coefficient of variation" and the concentration of Zn in ICP emission analysis <sup>5)</sup>. If the "minimum determination limit" ( $C_t$  in Fig.9.1.2) is taken as a concentration equivalent to  $R_b + 10S_b$ , an accuracy of about 10% can be expected. On the other hand, generally, because accuracy is also reduced at high concentrations, is also important to review the fact that there is a maximum determination limit. When only measurement accuracy is researched from literature, the determination limit is viewed as being in the range of 10 to 100 times the measurement accuracy.

The base line (zero air) noise band (peak to peak for 100 noise units) can be corresponded to in the recorder charts and concentration meter responses and the detection limits and determination limits defined as previously. Moreover, if the environmental concentration has no specific objectives, an effective figure for the mean value of two digits alone is sufficient.

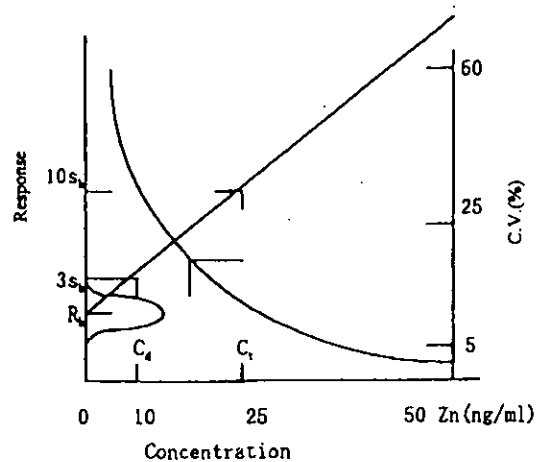


Fig. 9.1.2 Determination limits ( $C_t$ ), detection limits ( $C_d$ ) (Y-axis on the left) and standard deviation of blank ( $S_b$ ) in the calibration curve, and example of the relation between coefficient of variation (C.V.) and concentration of zinc in ICP emission spectrometry.

### (2) Accuracy guarantees and management: Tracability

No matter which measurement of the three given above which needs collection of air pollutants requires a standard for guaranteeing the accuracy. With both the liquid and solid standard substances, it is essential to consider purity of the standard substance (minimum 99.9%, though 99% alone is sufficient upon occasion.) and the stability of the prepared standard solution. On the other hand, with the concentration meters or the continuous analyzers, a standard gas cylinder is often used, and the manufacturer is requested to manage the accuracy of the cylinder concentration. Further, there are also concentration meters for which the regular inspection for the management is obligatory, and because for example, a part of the NO can change to  $NO_2$  while it is stored, or substances such as

formaldehyde may be adsorbed on the inside wall of the cylinder, these cylinders must be handled with care in the user's position.

#### 9.1.7 Laboratory management

When using a laboratory to analyze hazardous substances, it is important to consider both the environment and safety. Environment means hygiene in the work environment for the analysts, and countermeasures to contamination of air and water. Basically, it is best to choose processes that use as small amount of hazardous chemicals as possible. It is essential, for example, to use a dry process and to reduce the amount of preparation of standard solutions used to the minimum.

The first and foremost problem in the work environment is exposure of analysts to toxic vapor and suspended particles. Local ventilation must be installed in the rooms and the rooms must be kept well ventilated. Secondly, waste of analytical solution that has been used must be treated. Organic solvents can be burned, and metallic ion waste solution is treated by both adsorption and sedimentation, and if even small amount of the waste is extremely toxic, it must be put in a safe container, and labeled as such, and stored appropriately.

For safety, it is important to manage flammable gas and solvents. Large amount of hydrogen gas may leak or organic solvents could be volatilize, which when mixed with the air in the room, even to a concentration of only a few percent, once they meet a flammable source, detonation may start in the room. Care must be taken so that the storage shelves do not collapse, and that ventilation is good. Moreover, there is no danger in an ordinary room (not tightly closed) when the hydrogen leaks out of an FID gas chromatograph because it is diluted. Secondly is anoxia deficient room environment: This can be caused by major leaks of odorless nitrogen and argon. Thirdly, misoperation of the equipment should be prevented.

When setting out to analyze hazardous chemical substances such as dioxin, which has recently become a serious problem, it is needed to provide the hardware aspect such as buildings and other facilities, high quality analyzers. And so forth, analysis must be carried out in such facility as maintains rigorous safety management, such as control of waste substances, and a security system for people coming and going, and the work state. Also essential are a Quality Assurance/Quality Control (QA/QC) system, the introduction of recording systems, and a thorough comprehension of the history of a sample, from income of sample, up to the final treatment and disposal.

Because dioxin is 5,000 times more toxic than potassium cyanide, it must be kept in a room with reduced pressure to prevent its escape into the surrounding environment. In order to achieve sufficient pressure reduction, a special work box and compartment must be used. Dioxin in emission gasses and waste water from the laboratory must be treated by means of activated carbon, preventing its escape to the outside. Further, laboratory waste materials such as wipe clothes and used activated carbon must be stored in perpetuity, so appropriate facilities are also required.

Dioxin normally has a determination limit of approximately 1pg (pico gram, or  $10^{-12}$  g), so that solvents, reagents, and glass wears must be of the best quality so as to prevent contamination, and used only for dioxide analysis. Further, a blank test is combined, and the blank value from contamination during experiments is checked to be below the determination limit.



## 9.2 Continuous analysis methods

### 9.2.1 Introduction

Because there are many occasions when measurement results obtained from continuous analyzers are used by government, it is essential to organize the data in a form that can be evaluated. Using measurement methods compatible with environmental standards, it is essential to establish a system that makes possible suitable data management such as data processing, evaluation methods, and so forth. In the case of analyzers, if the equipment in question conforms to both domestic and international standards, there will be no disparity between function and capability, and it is more important to implement appropriate installation of measuring stations and their systematic support management, rather than selecting that particular type of equipment.

Measuring method of monitoring the ambient air comprises the domestic laws and/or standards of each country and also international standards. At present, the bringing into line of standards internationally is being promoted. When building an atmospheric monitoring system, the typical standards that become both the basis and the standard reference are as follows:

ISO: International Organization for Standardization (international standards)

CFR: Cord of Federal registration (US standards)

ASTM: American Society for Testing and Materials (US standards)

JIS: Japan Industrial Standards (Japanese standards), Ambient Air Continuous Monitoring Manual  
(Environment Agency)

### 9.2.2 Continuous analyzer

Table 9.2.1 shows data concerning the continuous atmospheric analyzers that are used as standards.

Table 9.2.1 (1) Standard continuous analyzers

Measurement targets		
SO <sub>2</sub> Sulfur dioxide	Principle specifications	Ultraviolet fluorescent method, standard measuring range 0 to 0.05/ 0.1/0.2 /0.5/ 1.0ppm.
	Points to note	Sufficient precautions are required for the following: When the indicated values have incurred direct influence from aromatic hydrocarbons such as toluene and xylene, when a scrubber is needed to remove hydrocarbons, sites where motor vehicle exhaust gas levels are high, when the amount of scrubbing has reached high levels for hydrocarbons generated on building sites, etc., and when replacement frequency has risen.
NO <sub>x</sub> Oxides of nitrogen	Principle specifications	Chemiluminescence method, standard measuring range 0 to 0.1/ 0.2/ 0.5/ 1.0ppm.
	Points to note	With the switch method, the NO <sub>2</sub> can be scattered if the NO concentration is high, or fluctuates severely. Check dehumidifying measures, because the indicated values are interfered with moisture. High concentrations of NH <sub>3</sub> in the air cause dirtying within the reacting cell due to O <sub>3</sub> from the gas reactions, for which a scrubber is effective in removing the hydrocarbons. Further, there are also occasions when organic nitrogen compounds are measured as NO by the converter.
O <sub>3</sub>	Principle specifications	Ultraviolet absorption method, standard measuring range 0 to 0.1/ 0.2/ 0.5/ 1.0ppm.

Table 9.2.1 (2) Standard continuous analyzers

Measurement targets		
Ozone	Points to note	Because this is the switch method, support and management of the ozone decomposer is important. Because the chemiluminescence method requires a supply of ethylene as a reactant gas, the ultraviolet absorption method is most widely used.
CO Carbon monoxide	Principle specifications	Infrared absorption method, standard measuring range 0 to 5/10/20/50/100ppm.
	Points to note	There are two methods, the switch NDIR method and the gas correlation NDIR method. The switch method is a measuring method unique to Japan.
NMHC Non-methane hydrocarbons	Principle specifications	Gas chromatograph method, standard measuring range 0 to 5/10/50ppm.
	Points to note	There are many cases of the measuring equipment being damaged by inadvertently sucking in dirty air when starting Regular maintenance work such as cleaning the switching valves, columns, and pipes is needed. The measured values apply CH <sub>4</sub> conversion and sensitivity checks are done by C <sub>3</sub> H <sub>8</sub> is needed. (Water supplied to the hydrogen generator) pure water that has been specially refined.
THC Total hydrocarbons	Principle specifications	Hydrogen flame ionization method, standard measurement range 0 to 5/ 10 /20 /50 /100ppm.
	Points to note	In the various countries of South east Asia, THC are often measured. This is a continuous measuring method, which is different from the gas chromatograph method.
SPM Suspended particulate matter	Principle specifications	$\beta$ ray absorption method, standard measurement range 0 to 1/5/10mg/m <sup>3</sup>
	Points to note	It is important to ensure that a hood or screen is attached to the tip of the sampling tube to prevent rainwater, insects, or the like from being sucked in. Because measurements in places with high concentrations of automobile exhaust gas (in particular, diesel exhaust gas) suffers from the filter paper becoming clogged, a very powerful suction pump is used. Be careful when handling $\beta$ ray-emitting isotopes.
CO <sub>2</sub> Carbon dioxides	Principle specifications	Infra red absorption method, standard measurement range 0 to 500ppm, 350 to 450ppm
	Points to note	Highly sensitive measuring equipment in the range of 350 to 450ppm, responsive to global warming, requires various important maintenance measures against ambient temperatures and dehumidification of sample air around -80°C dew point.

9.2.3 Objectives and types of ambient air measurements

There are different types of measuring stations depending on the atmospheric measurement objectives, as shown in Table 9.2.2.

Table 9.2.2 Measuring station objectives and types

General ambient air measuring stations	These are standard atmospheric measuring stations that generally monitor the state of air pollution in cities, residential areas, and industrial zones. They use standard analyzers.
Automobile exhaust gas measuring stations	These are the measuring stations that, continuously monitor the pollution state in the area around junctions and at roadsides where the air pollution is particularly severe due to automobile exhaust gas. They use analyzers that possess highly concentrated measuring ranges. It is important to ensure that dirt and particles do not interfere with the analyzers.
Ambient air measuring stations	These stations were established with the objective of obtaining basic samples from monitoring a wide range of ambient environment (background) conditions in unpolluted areas such as mountains, fields, and beaches, and because they measure low concentrations, the analyzers need to be highly sensitive with few fluctuations from zero. Further, there are occasions when analyzers are also installed to measure H <sub>2</sub> S, O <sub>3</sub> , CO <sub>2</sub> , and acid rain as well.

9.2.4 Construction and facilities of measuring stations

The measuring stations must be constructed so that the analyzers that have been installed can be protected easily

and they are easy to be maintained and managed. The points to note as for the measuring station facilities are shown in Table 9.2.3.

Table 9.2.3 Measuring station facilities

Power source Equipment	There must be sufficient surplus power, so that even when large pumps or the like are being powered, there are no fluctuations in the voltage, and this must also be the same as when an automatic voltage regulator is used. If the voltage fluctuations are severe, ideally an uninterruptable power source such as UPS should be used.
Air conditioner	The air conditioning should ideally maintain the air temperature at about -5°C, and never exceed 30°C, so that the air does not become too cold and have dew form. Also, ensure that the sampling tubes are not exposed to the chill from the air conditioner directly.

Further, the air sampling tubes should be as shown in Table 9.2.4.

Table 9.2.4 Sampling tube laying at measuring stations.

Collective sampling	The air is collected as one large flow in a manifold inside the measuring station building, where the air specimens are distributed through Teflon tubes into the various different measuring equipments. Metal tubes with Teflon lining, or reinforced glass are used. The PVC use materials which are noto charged with electricity such as PVC tubes or stainless tubes,etc..
Individual sampling	The air is absorbed by individual Teflon sampling tubes of 6 to 8mm internal diameter from the outside of the measuring station building, and the SPM uses a PVC tube with an internal diameter of approximately 20mm.

## 9.2.5 State of international standards

### (1) Introduction

Measurement and analysis technology for the ambient air is applied to a wide range of actions, centering on continuous ambient air monitoring, and the target fields are also enormously diverse. It is essential to improve the quality of measuring technology in order to more accurately measure concentrations of hazardous substances in the air than at present. Further, there is much interest in the measure and analysis of greenhouse gasses, which is in itself a problem on a global scale and new air pollutants such as hazardous chemical products, also called HAP, which have recently become as major a topic. The important point in regard to such measurement and analysis is the consistency of their results, and they should have the recreatability and reliability in a broad sense. Recently, overseas technical support has also become very active, and the international consistency on measurement results is being greatly needed. There is a necessity for standardization of the methods of measurement and analysis, and it is necessary to review them constantly to keep them up-to-date.. In Japan, the measurement and analysis of the ambient air is carried out either in accordance basically with JIS, or using methods, directions and manuals in accordance with regulations, notifications, and notices based on the Air Pollution Control Law.

Other countries also have many regulations, and the basic ideas on the ambient air are contained in "Selected Methods for the Measurement of Air Pollutants," which were determined by the U.S. Department of Health, Education, and Welfare in 1965, and it is no exaggeration to say that it forms the basis for the various standards explained below.

In the U.S., the establishment and management of environmental quality standards are laid out by the Code of Federal Regulation, which are continually revised year after year, and which are also often quoted in other standards. In the U.S., besides the CFR there are various well-known standards, such as ASTM, NIST, IEC, and ANSI. In Europe, there is the DIN in Germany and the BS in Britain and, moreover, internationally, there used to be the renowned WHO Selected Methods, all these standards have been unified internationally as the International Standard, ISO, by the International Organization for Standardization.

A summary of the contents of the CFR, ASTM, and ISO, which may also be called the typical standards, is given below. Because each standard is continually revised every year, there is a great need for research into accordance with the various Japanese standards. Further, the statements from the international standards cited below are highly detailed, and they refer to a lot of literature. They are also highly reliable as standards because their contents are revised, and they have greater value as literature than they do as standards.

## (2) Japanese standards

The measuring methods for environmental quality standards in Japan are shown in notices and notifications based on the Air Pollution Control Law, the guideline on support and management of ambient air measuring equipment, and the Ambient Air continuous monitoring manual. The main points of which are as follows:

Sulfur dioxide	Solution electrical conductivity method	JISB7952 solution electrical conductivity method
Sulfur dioxide	Ultraviolet fluorescent method	JISB 79520 ultraviolet fluorescent method
Carbon monoxide	Methods using non-diffuse infrared analyzers	JISB 7951 infrared absorption methods
Suspended particulate matter	light diffusion method, piezo-balance method, $\beta$ ray absorption method	JISB 7954 light diffusion method, piezo-balance method, $\beta$ ray absorption method
Photochemical oxidant	absorption spectrophotometry, coulometric method	JISB 7957 absorption spectrophotometry, coulometric method
Nitrogen dioxide	absorption spectrophotometry using Saltzman agent	JISB 7953 absorption spectrophotometry
Nitrogen dioxide	Chemiluminescence method	JISB 7953 chemiluminescence method
Hydrocarbons	Direct measuring method	JISB 7956 hydrogen flame ionization detection method

## (3) Code of Federal Register (CFR)

This law, which falls under the jurisdiction of the United States government's EPA, is the most influential law for environmental measuring. The 40 CFR 50 National Primary and Secondary Ambient Air Quality Standard not only indicates the national standards for air quality, but also regulates the measuring methods in detail for individual substances, as shown in Appendix A to K.

The 40 CFR 53 Ambient Air Monitoring Reference and Equivalent Methods regulates the definitions and methods of experiment for each measurement method. There are two types of measurement methods, manual and automated, and it is extremely interesting that SO<sub>2</sub> is defined to be measured by manual method.

#### Manual Reference Method

TSP High Volume Sampler

LED High Volume Sampler with Atomic Absorption Analysis

SO<sub>2</sub> Pararosaniline Method

#### Automated Reference Method

O<sub>3</sub> MP Chemiluminescence with Ethylene. CP Ultraviolet Photometry

NMHC MP Gas Chromatography with Flame Ionization Detector CP Calibration Gases

NO<sub>2</sub> MP Chemiluminescence with Ozone. CP Gas Phase Titration of on NO Standard with Ozone or NO<sub>2</sub> Permeation

CO MP Nondispersive Infrared Spectrometry Device. CP Calibration Gases

Titles for APPENDIX A~K are introduced below.

APPENDIX A - Reference Method for the Determination of Sulfur Dioxide in the Atmosphere (Pararosaniline Method)

APPENDIX B - Reference Method for the Determination of Suspended Particulate Matter in the Atmosphere (High Volume Method)

APPENDIX C - Measurement Principle and Calibration Procedure for the Carbon Monoxide in the Atmosphere (Non-Dispersive Infrared Photometry)

APPENDIX D - Measurement Principle and Calibration Procedure for the Measurement of Ozone in the Atmosphere

APPENDIX E - Reference Method for the Determination of Hydrocarbons Corrected for Methane

APPENDIX F - Measurement Principle and Calibration Procedure for the Measurement of Nitrogen Dioxide in the Atmosphere (Gas Phase Chemiluminescence)

APPENDIX G - Reference Method for the Determination of Lead in Suspended Particulate Matter Collected from Ambient Air

APPENDIX H - Interpretation of the National Ambient Air Quality Standards for Ozone

APPENDIX I - Reserved

APPENDIX J - Reference Method for the Determination of Particulate Matter as PM<sub>10</sub> in the Atmosphere

APPENDIX K - Interpretation of the National Ambient Air Quality Standards for Particulate Matter

#### (4) American Society for Testing and Materials (ASTM)

This is a broad range of standards equivalent to Japanese Industrial Standards (JIS), and it divided into 15 sections, which regulate a vast number of standards concerning all manner of materials, experiments, analyses, measurements, electricity, medical treatment, and so forth. Many standards are quoted by JIS.

Measurement and analysis of the ambient air is given in,

Section 11 Water and environmental Technology

Volume 11.03 Atmospheric Analysis

The following titles are cited for items relating to ambient air continuous monitoring in Japan:

- D- 1607- 91 Standard Test Method for Nitrogen Dioxide Content of the Atmosphere (Griess-Saltzman Reaction)
- D- 1704- 95 Standard Test Method for Determination the Amount of Particulate Matter in the Atmosphere by Measurement of the Light Absorbance of a Filtered Sample
- D- 2914- 95 Standard Test Method for Sulfur Dioxide Content of the Atmosphere (West - Gaeke Method)
- D- 3249- 95 Standard Practices for General Ambient Air Analyzer Procedures
- D- 3608- 91 Standard Test Method for Nitrogen Oxides (Combined) Content in the Atmosphere by the Griess-Saltzman Reaction
- D- 3609- 91 Standard Practices for Calibration Techniques Using Permeation Tubes
- D- 3824- 95 Standard Test Method for Continuous Measurement of Oxides of Nitrogen in the Ambient or Workplace Atmosphere by the Chemiluminescent Method
- D- 4298- 95 Standard Guide for Intercomparing Permeation Tubes to Establish Traceability
- D- 4536- 95 Standard Test Method for High- Volume Sampling for Solid Particulate Matter and Determination of Particulate Emissions
- D- 5011- 92 Standard Practices for Calibration of Ozone Monitors Using Transfer Standards
- D- 5110- 94 Standard Practices for Calibration of Ozone Monitors and Certification of zone Transfer Standards Using Ultraviolet Photometry
- D- 5149- 90 Standard Test Method for Ozone in the Atmosphere: Continuous Measurement by Ethylene Chemiluminescent
- D- 5015- 95 Standard Test Method for pH of Atmospheric Wet Deposition Samples by Electrometric Determination
- D- 5280- 94 Standard Practice for Evaluation of Performance Characteristics of Air Quality Measurement Methods with Linear Calibration Function

(5) International Organization for Standardization (ISO)

The ISO International Standard is the international standard for quality control. The ISO standards are attracting attentions by the debut of the ISO9000 and ISO14000 series. ISO holds an international position as the global standards across a wide range of materials, processing, motor vehicle, plastics, foodstuffs, medical treatments, and measuring and analysis.

Items concerning the measurement and analysis of the ambient air are basically as below, but both CFR and ASTM are often quoted with regard to their content.

- 13. Environment and Health Protection. Safety
- 13. 040 Air Quality
- 13. 040. 10 General Aspects
- ISO 4225 : 1994 Air Quality - Vocabulary - Biling Edition
- ISO 4226 : 1993 Air Quality - Unit of measurement
- ISO 6879 : 1983 Air Quality - Performance characteristics and related concepts for air quality measuring methods
- ISO 7708 : 1983 Air Quality - Particle size fraction definition for health - related sampling

- ISO 8756 : 1994 Air Quality - Handling of temperature, pressure and humidity data
- ISO 9169 : 1994 Air Quality - Determination of performance characteristics of measurement methods
13. 040. 20 Ambient atmospheres
- ISO 4219 : 1979 Air quality - Determination of gaseous sulfur compounds in ambient air Sampling equipment
- ISO 4220 : 1983 Ambient air - Determination of a gaseous acid pollution index - Titrimetric method with indicator potentiometric end-point detection
- ISO 4221 : 1980 Air quality - Determination of mass concentration of sulfur dioxide in ambient air - Thorin spectrophotometric method
- ISO 4227 : 1989 Planing of ambient air quality monitoring
- ISO 6767 : 1990 Ambient air - Determination of the mass concentration of sulfur dioxide - Tetrachloromercurate (TMC) /Pararosaniline method
- ISO 6768 : 1985 Ambient air - Determination of the mass concentration of nitrogen dioxide - Modified Griess-Saltzman method
- ISO 7168 : 1985 Air quality - Presentation of ambient air quality data in alphanumerical form
- ISO 7996 : 1985 Ambient air - Determination of the mass concentration of nitrogen oxides - Chemiluminescence method
- ISO 8186 : 1986 Ambient air - Determination of the mass concentration of carbon monoxide - Gas chromatographic method
- ISO 9359 : 1989 Air quality - Stratified sampling method for assessment of ambient air quality
- ISO 9835 : 1993 Ambient air - Determination of a black smoke index
- ISO 9855 : 1993 Ambient air - Determination of the particulate lead content of aerosols collected on filters - Atomic absorption spectrometric method
- ISO 10313 : 1993 Ambient air - Determination of the mass concentration of ozone - Chemiluminescence method

## 9.3 Planning and management of air pollution monitoring systems

### 9.3.1 Introduction

There are two kinds of air pollution monitoring: Mobile monitoring, which is used for settlement of complaint and environmental assessments, and continuous monitoring conducted in the general ambient air measuring stations, which measure the general ambient air representative of a region, and also conducted in the automobile exhaust gas monitoring stations, which measure automobile exhaust gasses along roadsides.

An air pollution monitoring system has such aims as to accurately measure, evaluate, and use fluctuations in the state of air pollution. It consists of continuous air pollutant analyzers, data processors, a data transmission system, a central data processor, terminal equipment, and so forth.

The continuous air pollution monitoring system is required to have such functions as ① on-line processing of the measured values in a short space of time at the central monitoring station, thorough comprehension of the state of pollution in real time and reacting in an emergency, ② alliance with the surrounding local governments and the ability to thoroughly comprehend in a short space of time the state of pollution over a wide area, ③ the establishment of a base to instruct the air pollution source to reduce of the emissions of air pollutants, ④ Giving information to citizens through the use of streetfront displays, ⑤ rapid detection of system abnormalities and pinpointing the location of a breakdown, and so on.

### 9.3.2 Planned disposition of the monitoring system

Notice is sent out from the Environment Agency for the suitable disposition of the monitoring system. A summary is shown in Table 9.3.1.

Table 9.3.1 (1) Summary of Environment Agency notice concerning the disposition of ambient air measuring stations.

Subject	Contents
<p>“Enforcing the law to revise part of the Air Pollution Control Law” Article 5 of the Japanese environmental atmospheric plan, 25 August 1971, promulgated by the Director-General of the Air Quality Bureau</p>	<p>Monitoring and measuring sites for air pollution</p> <p>In considering the characteristics of air pollution, it is essential to site the measuring points effectively in locations that pay attention to air pollution.</p> <p>A. Sulfur oxides: Install at one site per 25 km<sup>2</sup> as standard. Increase as need arises in regions where the air pollution is particularly severe.</p> <p>B. Carbon monoxide: Install at sites where people normally live or are active, and which are close to roads of frequent traffic or junctions.</p> <p>C. Suspended particulate matter: Combined measuring site for both sulfur oxides and carbon monoxide.</p> <p>D. Carbon dioxide: As above.</p> <p>E. Oxidant: Install in consideration of the fact that the impact of pollution due to oxides, etc., has extended into the suburbs, where pollution has hitherto not been severe.</p>



Table 9.3.1 (2) Summary of Environment Agency notice concerning the disposition of ambient air measuring stations.

Subject	Contents
Regional representations of measurement values obtained in general ambient air measuring station (investigation committee concerning regional representations of measurement values. Environmental Agency, report submitted March 1986)	Send the "Report by the investigative committee into regional representations of measurements values" to all local governments as reference material for their discussion. [Summary] Representations of NO <sub>2</sub> , SO <sub>2</sub> Differences in the mean values for a given period of time between sites is expressed as a single measurement value being representative of a region in which the tolerance has been suppressed to within a given range (5ppb), and this value is called the "regional representation." When the total value for the target area is divided up into a suitable shape for the area, the measuring station is said to have regional representation, and in this way the distribution of the measuring stations is called the "appropriate distribution of measuring station."

It is essential to thoroughly comprehend the regional representation encountered in each local government area, while referring to this notice. Here, at the local government level, regions are broken down into town and village units by surface area, the mobile measurement vehicles are employed at this juncture and the state of air pollution measured, the suitable distribution of the measuring station is discussed from these results, and the regional representative point are decided.

Further, in regions where there is an abundance of measurements and a plan for reducing the total quantity of both SO<sub>2</sub> and NO<sub>2</sub> has been established (using air diffusion simulation), it is again necessary to employ these results. Table 9.3.2 shows the points for consideration listed in the ambient air continuous monitoring manual.

Table 9.3.2 Points for ambient air measuring station consideration

Name	Contents
"Ambient Air Continuous Monitoring Manual" (revised edition) (Air Quality Bureau, Environment Agency (ed.), pub. December 1990.)	Published as a reference manual for local government continuous monitoring operations (revised from the 1986 edition.) (Extract from essential points for measuring station) <u>Items to consider when siting general ambient air measuring station</u> ① Site in consideration of the characteristics of pollutant sources, source distribution and prevailing wind characteristics. ② Site with greater regard for the minutiae in areas of dense population, when compared with other regions. ③ Site along border areas in consideration of the meteorological conditions and sources distribution of the neighboring local government. ④ Site in consideration of plans for future use of the land. ⑤ Do not site disproportionately in specific regions. <u>Positioning the specimen air sampling vent</u> ① Separate as far as possible from nearby buildings and obstacles so as to avoid being affected by turbulence, etc. ② Site so as to avoid being affected by specific sources such as chimneys, smokestacks, etc. ③ Set to an appropriate height so as to accurately comprehend the state of pollution due to the measurement target substances. "The ideal height is 1.5 to 10m above ground level." (Quoted from the notice.) <u>Points to consider when siting roadside air pollution monitoring stations (given in the latest revision.)</u> The state of roads, traffic volume, and so forth is considered so that the state of air pollution due to automobile exhaust gas can be effectively monitored. ① Considering the meteorological conditions of the region and the locality, the state of the topography, road construction, and the surrounding buildings, and the volume of traffic and the state of traffic jams, decide on sites that enable effective comprehension of the state of air pollution. ② Select sites and locations in the vicinity of traffic-laden roads or intersections, and where people normally live and work, or else a site close to such a location. Further, considering the fact that automobile vehicle exhaust gas wane with distance, the ideal site is as close to a road as possible.

### 9.3.3 Measurement sites and area of measuring station

Notice and notification from the Environment Agency concerning the height of the specimen sampling vent, the site of the ambient air measuring station, and so forth, when setting the environmental standards is shown for each air pollutant in Table. 9.3.3.

Table 9.3.3 Height of the "sampling vent" and "measuring site" of the air pollution continuous measuring station.

Measurement	Site	Collection vent height	Exceptions
CO	Site in the vicinity of a traffic-laden road or crossroads, or else close to such a site.	Approximately 1.5m	
SO <sub>2</sub>	Set the measuring points at one point per 25 km <sup>2</sup> as standard, and increase as necessary in regions of severe air pollution.	As a rule 1.5m or greater, 10m or below	For heights of 10m or more such as high-rise housing or other sites where many people are living, set appropriately given the conditions.
SPM	Measuring points are combined for sulfur oxides and carbon monoxides.	As a rule 3m to 10m above ground level	In order to alleviate the effects of sand and dirt being blown up from the ground, set the height as a rule to 3m to 10m above ground level.
NO <sub>2</sub>	Measuring points are combined for sulfur oxides and carbon monoxides.	As a rule 1.5m to 10m above ground level	Same as SO <sub>2</sub>
Ox	Site measuring points in consideration of the fact that the effects of pollution due to sulfur oxides, etc., have reached the suburbs, where they were not severe before.	As a rule 1.5m to 10m above ground level	Same as SO <sub>2</sub>

Further, in addition to being able to accommodate basic meteorological (wind direction, wind speed, temperature, humidity, sunlight, etc.) measuring equipment and measuring equipment for each air pollutant, the area of the measuring station also needs the space in which to install office desks for data processing, sluices for cleaning, lockers for storing parts, and so forth. The floor space for a measuring station requires 20 to 30m<sup>2</sup> as a rule.

### 9.3.4 Installation conditions of measuring equipment

The installation site for measuring equipment comes equipped ideally with the following conditions:

- (1) To prevent vibration: Countermeasures against vibration so as not to transmit to other equipment tremors from the suction pump, etc.
- (2) Avoid or prevent corrosion from corrosive gasses and dust.
- (3) Avoid high humidity or, when the room temperature is 40°C or below, ideally control temperature fluctuations to within  $\pm 5^\circ\text{C}$  in a room temperature range of 10 to 30°C, and avoid contact with direct sunlight as far as possible.
- (4) Ensure that there are few fluctuations in the electrical voltage and frequency. Also, discuss anti-lightening measures.
- (5) Set the equipment level so as to prevent tilting.

- (6) Ensure that maintenance work can be carried out easily and safely.
- (7) Manage with short air sample tubes. Further, ensure that rainwater, exhaust gasses, and so forth, are not absorbed directly.
- (8) Part of the waste solution from the wet method analyzer must be managed by qualified personnel with the “special industrial waste manager” because it is specific industrial waste matter.

### 9.3.5 Maintenance and management of measuring equipment

When supporting telemeter systems that accurately obtain measurement data, which fluctuates from moment to moment, accumulates, processes, and passes on that data, it is essential to religiously conduct spot checks and maintenance on the measuring equipment, and to pay close attention so that no fault arises in the data. Here, the types and content of the various measuring equipment support and management is shown in Table 9.3.4.

Table 9.3.4 Types and content of maintenance and management

Management section	Objective	Frequency of implementation	Content
Daily checks (Normal checks) (Rounds checks)	Continuous normal operation of automatic measuring equipment. (Data gathering)	Min. once/week	1. Check measuring equipment operations status. 2. Replace and resupply expendable parts. 3. Easy calibration and checks. 4. Easy cleaning and replacement of parts.
Periodic checks (Close checks) (Check transmission accuracy)	Support equipment functions and guard against breakdowns. (Keep within accuracy standards)	Min. once/year	1. Inspect flow paths 2. Inspect detector 3. Inspect control and transmission systems 4. Inspect amplifiers and recorders
Emergency checks	Rapid and prompt checks when a malfunction or breakdown occurs (restore normal status.)	During a malfunction	1. Identify the breakdown and execute minor repairs. 2. Identify and repair cause (depending on the manufacturer.)
Function tests	Prevent trouble and safeguard sustainability regarding maintenance or data evaluation (thoroughly comprehend the equipment characteristics.)	When purchasing equipment (including remodeling, replacement, and renewal) One month	1. Equipment function test (standard gas meter, etc.) 2. Equipment stability test (flow fluctuations, zero, span drift, etc.) 3. Measurement data evaluation (compatibility with obsolete equipment, etc.)
Dynamic calibration	Determine the contents of the accuracy range.	When necessary	1. Check by standard gasses 2. Compile calibration curve

## 9.4 Dust fall measurement methods

### 9.4.1 Introduction

Of all the air pollutants, dust fall is the name given to smoke, soot, or other impure particles that settle along with the rain, due to their own weight. Of the dust fall measurement methods<sup>1-7)</sup>, this explanation deals with the deposit gauge method, which is both the most widely used and the most general.

### 9.4.2 Sampling method

The dust fall totals are compiled as shown in Fig.9.4.1. The wire mesh A protects the trap funnel B somewhat, preventing birds from cutting off the connection. If a glass collecting bottle is being used during the winter, because there is a risk of it being damaged by frost, the bottle is best wrapped in a suitable insulating material, or else in place of glass, a polyethylene bottle should be used.

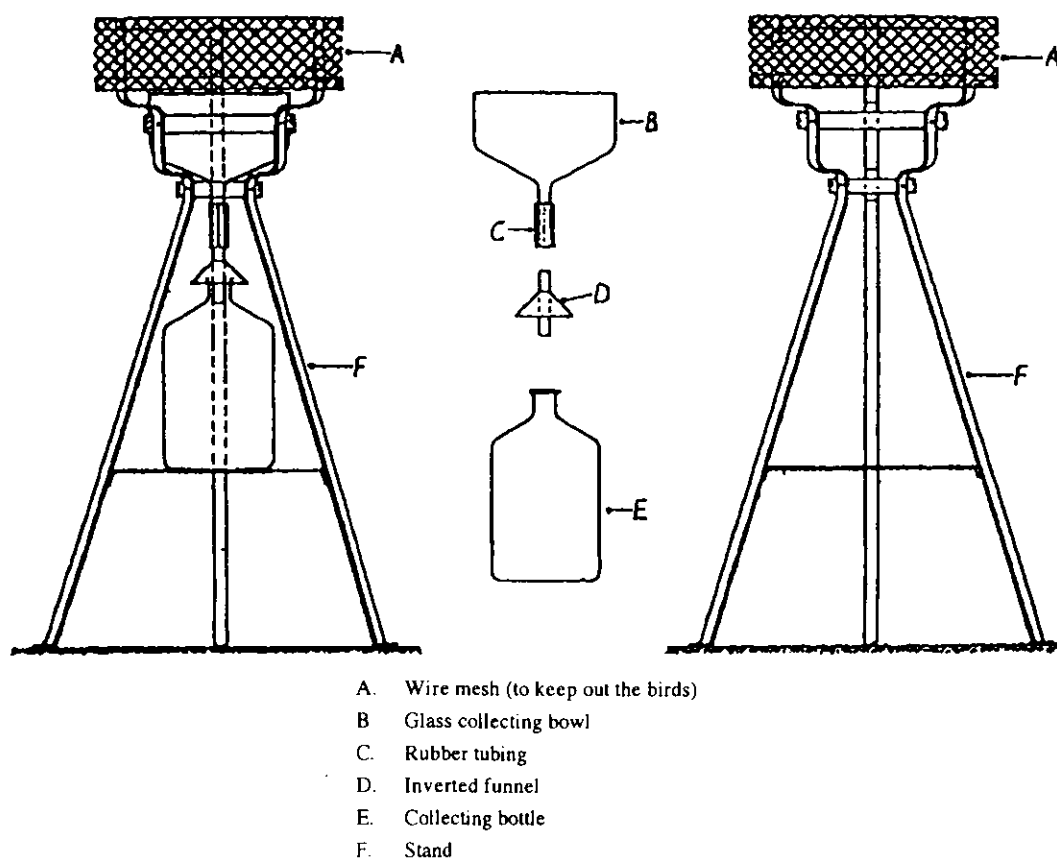


Fig.9.4.1 Deposit gauge assembly

The deposit gauge should be erected on the surface of a wide vacant space, and the distance from all surrounding buildings also needs to be a minimum distance of double the height from the collection funnel to the height of the

building.

In Japan, in order to prevent a sandstorm from blowing up from the ground surface, in many cases the gauge is installed on the roof of a building five to 15m in height above the ground, in which case, it is essential to pay attention to the positioning of funnels, and so forth.

The deposit gauge must be fixed firmly in place with either nails or bolts. Because the period of collection is one month, if lichens begin to form, there is a danger of the collected substances undergoing chemical changes. In order to prevent this, 0.02N copper sulfate solution 10 ml is added to the collecting bottle. One month later, the collecting funnel is examined, and any clearly foreign bodies such as leaves, insects, and so forth, are removed. Next, a burette brush is passed through the spigot of the glass funnel and moved up and down until it reaches inside the connecting tube. Approximately 250 ml of water, which has been collected in the bottle, is poured into the funnel (if the water in the bottle is insufficient, a fixed quantity of distilled water is used instead.) The sediment that has adhered to the inside of the collection funnel is passed through a policeman, and the water, which has been clouded by the solids is allowed to flow into the bottle, during which time the inside of the connecting tube is cleaned with a burette brush.

The bottle is then replaced with a pre-prepared clean one, and the dust meter is ready for another month of measurements. A cork stopper is placed firmly in the collecting bottle containing the water and the solids, and the following information is recorded on a label which is then affixed to the bottle.

Details of the site, the collecting period, an accurate measurement of the diameter of the collecting funnel and, if distilled water was used to clean the container, the quantity, and the effects, if any, of foreign bodies on the sediment, are all entered.

#### 9.4.3 Analysis

Generally, for analysis, the following items are needed for the report:

- ① Quantity of solution collected: The quantity of solution minus any distilled water that was added and the copper sulfate solution.
- ② Solution pH value: Unnecessary if water was added.
- ③ Quantity of undissolved substances.
- ④ Quantity of tar: Portion of the ③ that is soluble is  $CS_2$ . In Japan, acetone is used in place of  $CS_2$ .
- ⑤ Quantity of ash: Obtained by extracting the  $CS_2$ , and burning off the residue.
- ⑥ Flammable substances other than tar: Namely, (6)-(3)-(4)-(5).
- ⑦ Total of soluble matter.
- ⑧ Quantity of  $Ca^{++}$ ,  $Cl^-$ ,  $SO_4^{--}$ , etc.
- ⑨ Quantity of solids: This is the sum total of both the soluble and insoluble substances. An outline of the analytical operation is shown systematically in Fig.9.4.2.

When the solution quantity is 300 ml or less, distilled water is added to make it up to 300 ml. Filter paper such as Toyo filter paper No. 5C, or another ashless filter paper, is used. For extracting the tar, after drying and

weighing the water-insoluble matter on the filter paper, Soxhlet extraction is carried out, but normally several hours are required. The extracted insoluble matter is then heated forcefully in an electric oven at 800°C to determine the ash content. A fixed quantity of the filtered solution (1 to 2 0) is placed on either a platinum or porcelain dish for which the weight is known, dissolved in water, and evaporated to dryness. Next, this is dried for two to three hours in a dryer at 105°C, after which it is left to cool in a desiccator and weighed, the difference in weight both before and after the evaporation tray is determined, and is converted to the solution weight and the total weight of the soluble matter is thereby determined. At present, the ion components of multiple substances, such as SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and Na<sup>+</sup> can be analyzed using ion chromatography.

The quantity of dust fall (or its components) is expressed as t/km<sup>2</sup>/month (= g/m<sup>2</sup>/month), and expressed by Eq. (1).

$$\text{Quantity of dust fall} = 1.273 \frac{w}{D^2} \times 10^4 \dots\dots\dots (1)$$

Here,  $w$  = Total quantity (or components) of analysis values (g)

$D$  = Diameter of collecting bowl (cm)

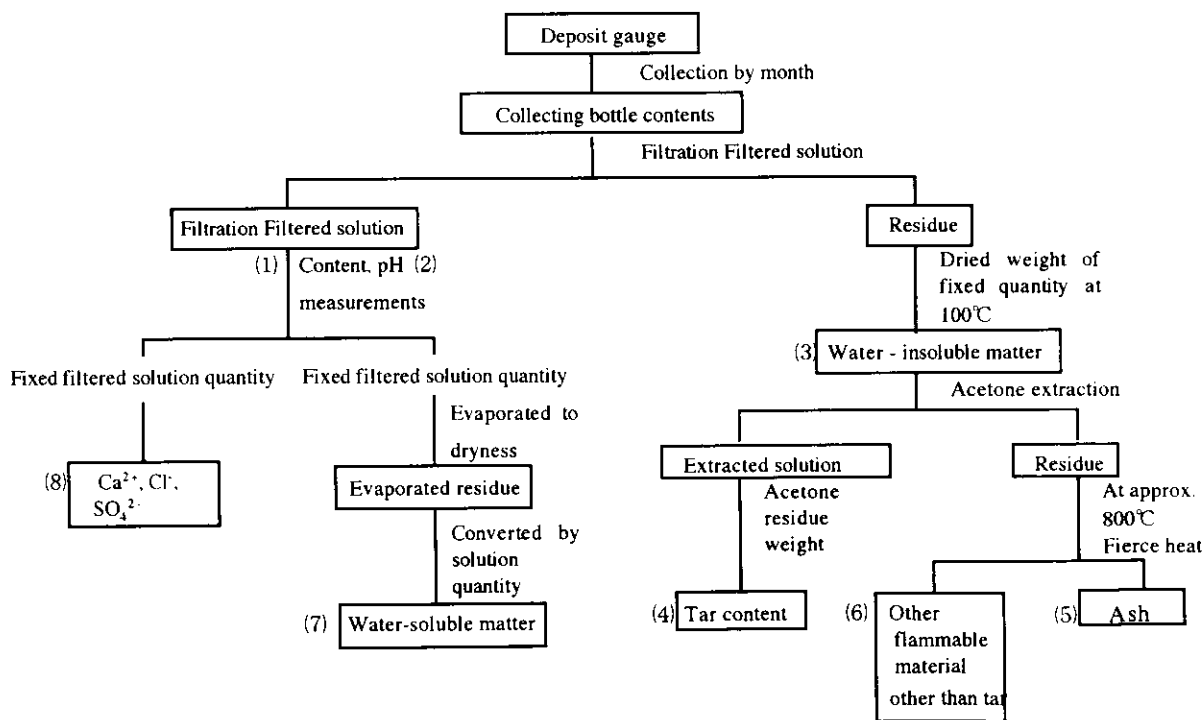


Fig.9.4.2 Deposited matter general analysis system

## 9.5 Measurement Techniques of Acid Deposition

### 9.5.1 Introduction

One of the well known global environmental issues is acid deposition. Acid deposition, sometimes socially referred to as “acid rain”, is air pollution on a continental scale. Combustion of fossil fuel injects sulfur and nitrogen oxides into the atmosphere. These oxides are subsequently converted to sulfuric and nitric acids by atmospheric photochemical processes, which result in the acidification of the atmosphere <sup>1)</sup>. Sulfuric and nitric acids thus formed are deposited onto the earth’s surface by wet and dry deposition processes. The deposited acids then acidify terrestrial and aquatic environments, which will cause adverse effects on various environmental elements including structural and ornamental materials and human health.

An important thing to note here is this environmental issue should not be recognized to be “acidification of rainwater” or “acidic fallout”. The deposition of chemical species from the atmosphere to the earth’s surface is the end result of a complex chain of physical and chemical processes in the atmosphere and is an input to another complex chain of processes that take place in terrestrial and aquatic ecosystems.

In other words, atmospheric deposition is also known to provide a primary pathway for the entry of nutrients and toxic chemicals into terrestrial ecosystems and oceans. The scientific description of this issue is acidification of the atmosphere as well as the environment. We should term this environmental issue “global acidification” because fossil fuel combustion also cause as global warming by emitting carbon dioxide.

Wet deposition is the deposition of chemical species associated with precipitation processes: in-cloud and below-cloud scavenging. Dry deposition is the deposition of the species in the gaseous and particulate forms by the atmospheric turbulent processes in the boundary layer and physico-chemical interactions with the surface of various substances in the receptor. Although numerical and experimental evaluations so far have reported that dry deposition will be as important as wet deposition, it is difficult to evaluate dry deposition on large spacial and temporal scales with sufficient certainty.

In this chapter, the basic concept of measurement techniques of wet deposition will be discussed below. Details of measurement techniques have already been addressed elsewhere <sup>3)-7)</sup>.

### 9.5.2 Points for Wet Deposition Measurement Techniques

Measurement techniques used in wet deposition programs depend heavily on the program objectives. Definition of program objectives requires consideration of all factors influencing the nature and outcome of the program. In practice, measurement program objectives are always constrained by monetary and logistical factors. It is extremely important that all such constraints be identified at the beginning of the program so that they can be accounted for in the design of the measurement system. Typical constraints include budget limitations, manpower availability, access to instrumentation, access to electrical power, proven chemical analysis capabilities, and availability of computers for data handling.

The most appropriate data is obtainable only when necessary and sufficient methods are utilized. Not only

will simple compromise or applications of unnecessarily sophisticated techniques produce data sets with inconsistent quality, they would also waste human energy, time, and funds. Proper measurement program design ensures that the program objectives will be satisfied, that the program constraints will be met, and that the program will be implemented in a timely and efficient manner. Adhering to this process reduces the amount of system alternation during the execution of the program.

Measurement techniques will be grouped into the four categories: 1) siting, 2) sample collection and handling, 3) chemical analysis, and 4) quality control and quality assurance. Different techniques will produce different descriptions of the same phenomenon. Before a comparison of different sets of data, one should clearly ensure the difference in the utilized techniques. In order to derive the proper conclusions from a data set, the impacts of the measurement quality on the data quality should be evaluated. The uncertainty of the conclusions derived from the data set should be consistent with the data quality. Each of the four categories made above will be individually discussed.

#### (1) Siting

Once a site has been selected, it is very difficult to relocate it with ease. The siting is the most decisive issue of all. What is important in site selection is whether the site is representative of the region of interest. Potential sources of pollutants to the samples are evaluated on three scales: on-site (ca. 1-100 m from collector), local (ca. 100 m-10 km from collector), and regional (ca. 10-50 km). Site representativeness is defined as the degree to which data accurately and precisely represents a characteristic of a population or parametric variation at a sampling point, a process condition, or an environmental condition. This element of data quality is generally evaluated in terms of semi-quantitative ratings. It is essential to make clear the nature of the measurement at the site which will provide different siting standards.

#### (2) Sample collection and sample handling

The wet deposition of a chemical species is calculated as the product of precipitation amount and species concentration. Precipitation amount is measured using standard precipitation gauges. The concentration is measured using precipitation chemistry collectors. The collector is usually a wet-only collector which is composed of a funnel-and-bottle with a movable lid on the funnel, that remains open only during precipitation. Another type of collector, bulk collector which remains open during both wet and dry periods, might be utilized on some occasions although the bulk sample will be contaminated by dry deposition during the dry period.

The sampling period is the length of time that a sample collection bottle stays in a wet deposition collector before being removed for shipping to a chemical analysis laboratory. There are a variety of sampling periods commonly used in precipitation sampling programs from daily to monthly. In major wet-deposition measurements networks of the world, samples are collected on a daily or event-basis.

#### (3) Chemical analysis

The dissolved inorganic ions of major concern to acid deposition measurement programs are the nine types of ions in Fig.9.5.1<sup>8)</sup>. The chemical analysis methods for the ions and conductivity are shown in Table 9.5.1. pH



measurement is still high problematic, and more research is needed to solve measurement problems. However, seen from the viewpoint of the entire measurement systems, the problems of chemical analysis are not very great. Rather, it is important to be aware of essential problems other than those of chemical analysis, such as siting, sampling, and sample handling.

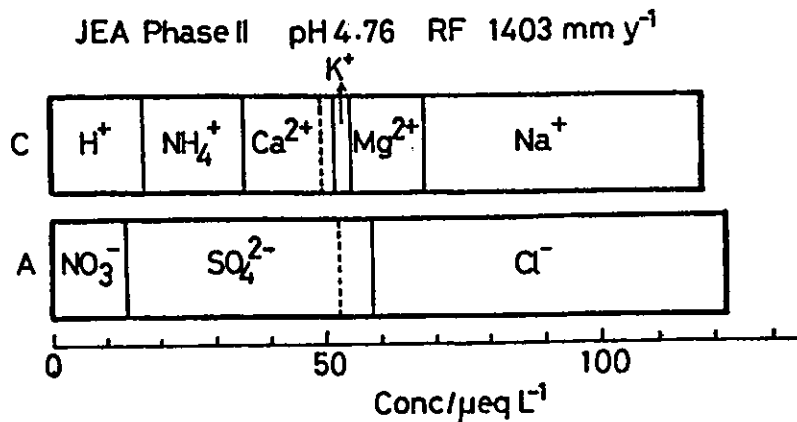


Fig.9.5.1 Nine types of ions<sup>8)</sup> usually measured by the chemical analysis of precipitation

Table 9.5.1 Methods recommended for the analysis of the main components of rainwater

Measurement item	Equipment analysis methods
pH	Glass pole (poles with non-leaking internal cells are ideal)
Electrical conductivity rate	Electrical conductivity meter
Ion chloride, ion nitrate, ion sulfuric acid, ion nitrite, ion fluorides	Ion chromatography (ideally with a sub-laser attachment)
Ammonium ion	Ion chromatography Spectrometry (indophenol blue)
Sodium, potassium, calcium, magnesium ions	Ion chromatography Atomic absorption spectrometry / emission analysis
Phosphoric acid ion	Spectrometry
Heavy metals, aluminum, mercury	Atomic absorption spectrometry using a graphite furnace, ICP emission spectrometry, ICP/MS, metal trough mercury analyzer
Organic acids	Ion chromatography

#### (4) Quality Control/quality assurance (QA/QC)

Quality control and quality assurance to date have only emphasized the chemical analysis of standard reference materials. However, quality control should apply to all the items ( (1) to (3) above) so that the overall data quality will meet the measurement objectives. Quality assurance is the quantitative evaluation of the data quality in terms of the above items. The purpose of the evaluation is to indicate to the data user the level of confidence he can place in the data for the analysis of interest so that one can select the pertinent data to their objectives. Even with data of low but clear quality, a proper interpretation is definitely needed to draw conclusions with such qualities are as consistent when the data quality itself<sup>9)</sup>. However, if the quality of the data is unclear, the interpretations and

associated conclusions will eventually be unclear as well.

The site is rated with respect to the representativeness in the three scales mentioned in the siting section. For the sample sampling, the sample history, such as the management of the sample collectors and storage bottles, shipping, storage until the chemistry is analyzed, should be recorded and handled as an important part of the data <sup>6)</sup>.

Validity of the chemical analysis is evaluated with respect to the ion balance and electric conductivity checks, in addition to analyzing standard reference materials. In the case of Fig.9.5.1, the balance of the full concentration of positive ions and negative ions is shown as being a very good one.

Data completeness is another element of quality assurance. This is defined as a measure of the amount of valid data obtained from a measurement system compared to the amount that was expected to be obtained under correct normal conditions. From a data set with poor data completeness, the missing values must not be estimated, nor the annual mean calculated. Wet deposition estimates should not be used if certain levels of completeness are not met.

The quality of a data summary is a function of the site representativeness and of the data completeness on which the summary is based. A qualitative assessment of the overall quality of a data summary can be obtained by combining the information in the categorization of the quality of a data summary. This overall data quality is an important attribute of the data one that plays a valuable role in assessing the usefulness of wet deposition data.

### 9.5.3 Summary

In order to make high quality wet-deposition measurements, the measurement system must be carefully designed, implemented, and operated. Particular care must be given to the selection of sites, sampling periods, instrumentation, sample collection and handling methods, analytical methods, data management, and data reporting. Ensuring that the measurements techniques produce data of suitable quality requires the operation of a quality assurance program. Considerable energy, money and attention should be dedicated to quality assurance programs <sup>6)</sup>.

## 9.6 Methods of analyzing airborne particulate matter

### 9.6.1 Introduction

The measuring of airborne particulate matter determines the concentration of the particles by collecting them on a filter using either a high volume sampler (hereinafter called an HV) or a low volume sampler (hereinafter called an LV), to provide an analysis of the metal content of the sample, and so forth <sup>1) 2) 3)</sup>. In addition, an Anderson Sampler <sup>4)</sup> or similar may be used as a means of collecting particles with different diameters.

If the metals are analyzed using either atomic absorption photometry or ICP (Inductivity Coupled Prazuma) luminescent analysis, it is first necessary to acid decompose the sample beforehand and liquefy the target substance, and then non-destructive analysis is possible through both radio activation analysis and X-ray fluorescence analysis.

Here, this article is concerned with atomic absorption photometry, which is the most widely used means of analyzing the metal content of particles, and explains in outline radio activation analysis, with which non-destructive analysis is possible.

### 9.6.2 Atomic absorption photometry

#### (1) Sample collection and measuring mass concentration

A high volume (HV) sampler is shown in Fig.9.6.1, and a low volume (LV) sampler is shown in Fig.9.6.2. The filter requires a collection rate of at least 99% for particulate matter with a diameter of  $0.3 \mu\text{m}$ , low pressure loss, low reactivity to adhesion of gas particles and humidification, and must not include particles that may interfere with analysis. Normally, in the HV, a quartz fiber filter, fluorine resin filter, and a nitrocellulose filter are used, and in the LV, in addition to the above filters, a membrane filter is also used. The collection period for the HV is normally 24 hours, and for the LV is between four days to one week.

The filter is weighed following measurement of the stable temperature (normally  $20^{\circ}\text{C}$ ) for a minimum period of 24 hours both before and after the sample collection, and after the constant room humidity (50%) has been measured, and the particle density ( $\mu\text{g}/\text{m}^3$ ) is determined.

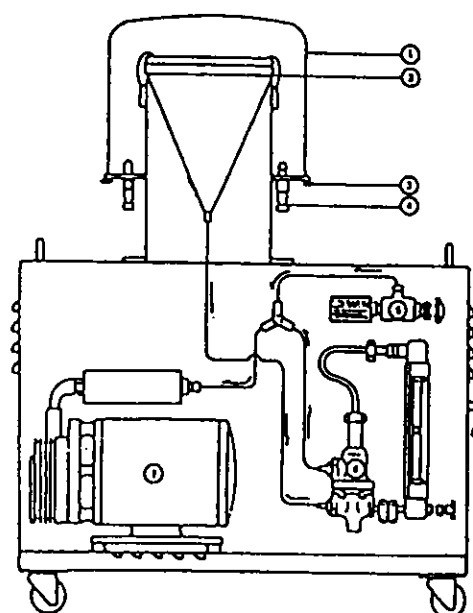
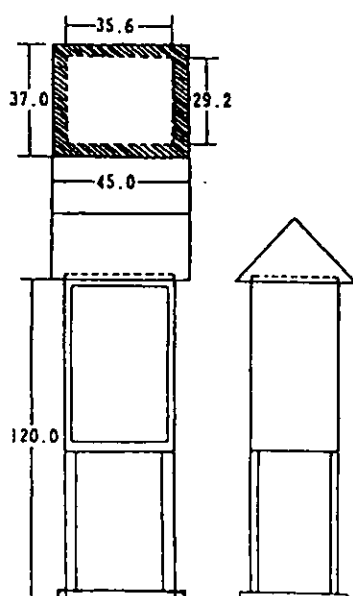
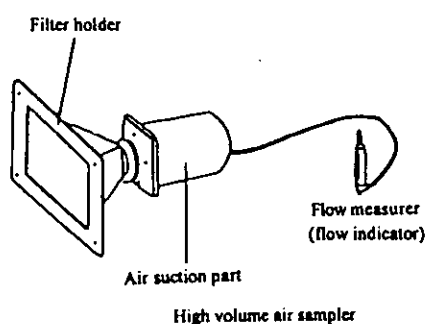
#### (2) Adjusting the sample solution (fluoric acid, nitric acid, perchloric acid method)

- a) The sample collection filter has an appropriate volume removed, is placed in a 100 ml tetrafluoride ethylene beaker, 20 ml nitric acid and 5 ml chloric acid are added, and the beaker covered by a tetrachloride ethylene clock dish, and heated gently on a hotplate at  $130^{\circ}\text{C}$  for approximately one hour. The clock dish is removed, and if the solution volume is about 5 ml, 5 ml of nitric acid, and the whole is reheated.
- b) After cooling, 10 ml nitric acid, 3 ml perchloric acid, and 3 ml hydrofluoric acid are all added, the clock dish is removed, the solution covered, and heated gently on a hotplate at approximately  $200^{\circ}\text{C}$ .
- c) When the perchloric acid begins to generate white smoke again, the solution is cooled, 5 ml nitric acid is added, and the whole heated. When the perchloric acid begins to generate white smoke again, the beaker is completely covered with the clock dish and continually heated, and the clock dish is removed once the contents

are no longer white, or pale yellow in color, whereupon they are dried and solidified, and once a thin white smoke begins to appear, the heating is stopped and the product cooled.

- d) 50 ml hot water and 10 ml nitric acid (1⇒10) are added, and heated for approximately a further 10 minutes either over boiling water or on a hotplate, and the solid matter is dissolved.
- e) After leaving to cool, the contents are filtered through a No.5C filter paper. The beaker and the filter paper are both washed in warm nitric acid (1⇒10), and filtered in the same way. All of the filtered solution is transferred to a 100 ml tetrafluoride ethylene beaker, gently heated either over boiling water or on a hotplate, and evaporated to a solid.
- f) After cooling, 10 ml hydrochloric acid (1⇒10) is added to the first beaker, which is then heated and dissolved over water. After cooling, the full volume is transferred to a flask (25 ml), water is added to a fixed volume, and the sample solution is thereby obtained.
- g) A blank specimen solution is prepared separately using a blank specimen filter by following the same steps a) to f) above.

In addition, the filter on which the specimen was collected is subjected to heat analysis in both a pressure analyzer and by aqua regia as a way of adjusting the sample solution, of which there are several <sup>3)</sup>.



#### Low volume air sampler

- ① Top cover (stainless steel)
- ② Filter holder, filter clamp (stainless steel)
- ③ Top cover tightening screw (5× plastic screws)
- ④ Cyclone (×3)
- ⑤ Regulation valve and filter (stainless steel)
- ⑥ Differential pressure valve (stainless steel, cover is acrylic)
- ⑦ Rotary pump (200 W)

Fig.9.6.1 HV (top) and its shelter (bottom)

Fig.9.6.2 LV construction

(3) Flame atomic absorption spectrometer conditions setting and equipment adjustment

The sample solution adjusted in 9.6.2 (2) is atomized by an oxy-acetylene flame, the absorption wavelengths of each target measurement substance is measured, and their volume determined from the calibration curve.

The flame atomic absorption analysis conditions should be set appropriately with reference to the following:

Analysis wavelength: Wavelength of each target substance (e.g., for nickel, 232.0 nm)

Lamp current: Depends on each target substance (e.g., for nickel, 10 mA)

Gas flow: Acetylene (1.7 l/min), air (15 l/min)

(4) Compiling the calibration curve

0.3 to 6.0 of the control solution for each target substance (100 μ g/ml) is transferred in steps into a total volume flask (100 ml), and after heating the acid under the same conditions as for the sample solution, water is added to make up the appropriate volume. A control concentration series with at least five steps, including zero, is then created.

The conditions in (3) are operated for each control concentration series created, and the calibration curve thereby compiled.

(5) The blank sample solution and the sample solution are both subjected to the operations in question, and both the blank value ( $M_b$ : μ g/ml) and the volume for each target substance ( $M_s$ : μ g/ml) are determined.

(6) Calculating the concentration

The airborne concentration of each target measurement substance is calculated using Eq. (1).

$$C = \frac{(M_s - M_b) \times E \times S}{s \times V_{20}} \dots\dots\dots (1)$$

Here,

$C$  = Airborne concentration of each target measurement substance (μ g/m<sup>3</sup>)

$M_s$  = Concentration of each target substance in the sample solution (μ g/ml)

$M_b$  = Concentration of each target substance in the blank sample solution (μ g/ml)

$E$  = Volume of sample solution obtained in (2) (ml)

$S$  = Surface area of filter that collected the sample (cm<sup>2</sup>)

$s$  = Surface area of filter used in the analysis (cm<sup>2</sup>)

$V_{20}$  = Air collection volume at 20°C (m<sup>3</sup>)

9.6.3 Activation analysis

(Principles and features)

Activation analysis is the general name given to analytical methods based on nuclear reaction. Of these,

thermal neutron radio activation analysis, which uses a semiconductor detector to measure gamma rays from radio active nuclides created by irradiating the sample with high density thermal neutrons in an atomic reactor can analyze trace amount of multiple chemical elements. As with airborne particulate matter, it is a method highly suited to the analysis of samples containing many trace elements, or where the amount of sample obtained is small, and it possesses the following features:

<Advantages>

- A highly sensitive method for analyzing multiple elements, and can analyze approximately 30 elements in airborne particulate matter.
- Because it can analyze non-destructively without chemical processing, there is little contamination (pollution) of the analysis.

<Disadvantages>

- Needs a semiconductor Ei-ray detector and atomic generator facilities for the experiment.
- Takes approximately one month for all analysis results to be obtained, due to the half-life of radioactive elements.
- Poor sensitivity for elements such as lead and cadmium.

(Analysis example)

Using a low volume air sampler at the Environment Agency's national air surveillance network, airborne particulate matter collected by a nitrocellulose membrane filter was irradiating and measured under the following conditions, and approximately 30 elements were analyzed <sup>5)</sup>.

- Atomic reactor and thermal neutron density: Rikkyo University Atomic Research Institute, thermal neutron density  $0.5$  to  $1.5 \times 10^{12}$  n/cm<sup>2</sup>/sec.
- Thermal neutron irradiation: Irradiation for three minutes (short-lived nuclide) and six hours (long-lived nuclide).
- Gamma ray measurement: For five minutes after several minutes (short-lived nuclide), for 20 minutes after several days (medium-lived nuclide), and for one hour after several weeks (long-lived nuclide).
- Elements measured: Ag, Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Fe, Hf, K, La, Lu, Mn, Na, Sb, Sc, Se, Sm, Th, Ti, V, W and Zn
- Other elements: Cd, Ni, and Pb were analyzed using X-ray fluorescence analysis.

Using radio activation analysis, and by measuring the various constituent elements in the atmosphere, the future of airborne particles could be characterized. Further, using the CEM (Chemical Element Balance) method, the rate of contribution from typical sources such as the soil (Al, Sc, etc.), sea salt (Na, Cl), steelworks (Mn, Fe, etc.), waste substance incinerators (K, Sb, etc.), and petroleum combustion (V, Ni, etc.), could be estimated from the measurement values of indicator elements. In addition, it is also used in earth sciences research to explain the yellow sands phenomenon.



③ Sample solution measurements

Approximately 20 μℓ of the sample solution, which has been adjusted in ①, is extracted using a microsyringe, and infused into the HPLC, the chromatograph is recorded, and about of the peak for the benzo (a) pyrene (hereinafter called BaP), either the peak surface area or the peak height is determined. The BaP concentration in the sample solution is determined from the calibration curve drawn up beforehand.

④ Drawing up the calibration curve

- a) The BaP first standard solution (10 μ g/ml) is diluted with acetonitril so that its concentration is 1 to 10 ng/ml, and a control series is compiled for the calibration curve. The control series has five or more steps, including zero.
- b) Using the ③ operation, either the peak area or peak height is determined equivalent to each BaP concentration.
- c) The calibration curve is drawn up from the relation between the peak surface area or height, and the BaP concentration.

⑤ Blank test

For the sample and the filter from the same lot, the sample solution is adjusted by means of the ①, the ③ operation is carried out, and the blank value (A<sub>b</sub>; ng) is determined.

⑥ Sensitivity test

The ③ operation is carried out with regard to the standard solution around the middle of the calibration curve from the control concentration series, and fluctuations in the sensitivity are checked. This operation is carried out at least once for every 10 sample measurements.

(3) Calculating the concentration

From the results of ③ and ⑤ in (2), the airborne BaP concentration is calculated using next Eq. (1).

$$C = \frac{(A_s - A_b) \times v_e \times E \times S \times 1,000}{v \times \left(\frac{4}{3}\right) \times v_c \times V \times s \times \frac{293}{273 + t} \times \frac{P}{101.3}} \dots\dots\dots (1)$$

Here,

C = Airborne BaP concentration at 0°C (μ g/m<sup>3</sup>)

A<sub>s</sub> = BaP in the sample solution injected into HPLC

A<sub>b</sub> = BaP blank (ng)

S = Filter area that collected the specimen (cm<sup>2</sup>)

s = Filter area that was used for the measurement (cm<sup>2</sup>)

E = Extracted solution volume (mℓ, normally 4 mℓ)

v = Volume of solution injected into HPLC (μ ℓ)

v<sub>e</sub> = Final sample solution volume (mℓ, normally 1 mℓ)

v<sub>c</sub> = Solution volume extracted after alkaline clean up (mℓ, normally 1 mℓ)

V = Air suction volume (ℓ)

t = Mean air temperature during sampling (°C)

P = Mean atmospheric pressure during sampling (kPa)



## 9.8 Methods of Analyzing Toxic Airborne Compounds (Volatile Organic Compounds)

### 9.8.1 Introduction

Volatile organic compounds are separated into aldehydes and benzene and chlorine-based hydrocarbons (hereinafter called VOCs), depending on their collection method. Moreover, I would like to refer to particulate matter concerning metallic toxic substances.

### 9.8.2 Methods of analyzing benzene and other volatile organic compounds (VOCs) <sup>1)-3)</sup>

#### (1) Sample collection

Generally, following electrolytic polishing of the inside, the specimen is collected (see Fig.9.8.1) using a system container (canister) that employs a special inert processing to create an even purer thin chrome/nickel oxidant membrane. In addition, there is also the solid absorption and solvent extraction method <sup>4)</sup>.

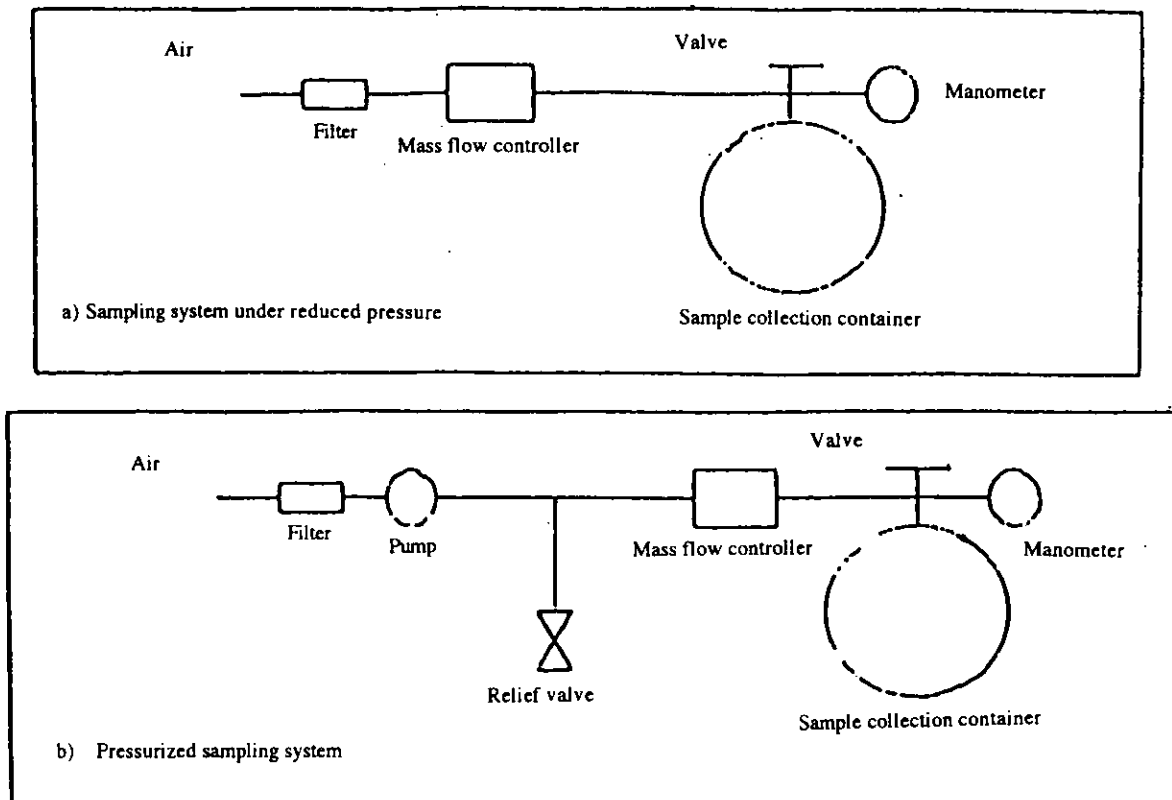


Fig.9.8.1 Overview of the sample collection container <sup>3)</sup>

#### ① Subatmospheric pressure sampling method (Standard atmospheric pressure collection method)

The tip of the sample collection container is connected to the sample collection equipment. The sample

collection container valve is opened, and collection begins to a previously determined flow volume, then after 24 hours the valve is closed and the tip of the sample collection container is stoppered. The sample collection start time, finishing time, and the pressure within the sample collection container are all recorded.

② Pressurized sampling system

The tip of the sample collection container is connected to the sample collection equipment. The valve is opened while the sample collection equipment pump is running, and collection begins to a previously determined flow volume, then after 24 hours the valve is closed and the tip of the sample collection container is stoppered. The sample collection start time, finishing time, and the pressure within the sample collection container are all recorded.

(2) Analysis operation

① The GC/MS analysis conditions are set and the equipment is adjusted.

The GC/MS analysis conditions are set as appropriate with reference to the following items:

- Column used : Methyl silicon uncovered capillary column  
Internal diameter 0.25 mm, length 60 m, membrane thickness 0.25  $\mu$  m
- Column temperature : 40°C (kept for 5 mins.)  $\rightarrow$  4°C/min  $\rightarrow$  140°C
- Interface temperature : 220°C
- Carrier gas : Helium 1 to 3 ml/min
- Ion source temperature : 200°C
- Ionization voltage : 70 eV
- Detection method : SIM or scan

Either PFTBA (Perfluorotri-n-butyl amine) or PFK (Perfluoro kerosine) is introduced to MS as a control substance for calibrating the volume, and using the subatmospheric, specific values are calibrated such as mass balance and analysis function (one unit of volume or more is taken to be in the range of volume (m/z) = 18 to 300), depending on the measurement objectives. The measurement results following calibration are also stored.

② Test concentration

If collecting is done using the sampling system under reduced sub atmospheric pressure, pressure is added to the sample collection container until it reaches approximately 200 kPa ( $1.5 \times 10^3$  mmHg), and the pressure  $P$  (kPa) within the sample collection container is read accurately. The dilution rate ( $n$ ) caused by the additional pressure is given by Eq. (1).

$$(n) = \frac{P}{p} \dots\dots\dots (1)$$

Here,  $p$  is the pressure (kPa) in the container following collection.

Specimens collected using either added pressure with zero gas following sampling system under reduced pressure or specimens collected using the pressurized sampling system are concentrated by a fixed flow of sample during dehumidification, with the sample collection container collected to the sample introduction equipment. Control of the flow volume is done using a mass flow controller, and concentration is completed in a fixed amount

of time. At this juncture, a fixed volume of control gas, the same as for when the calibration curve was compiled, is also concentrated within the concentrator.

The concentrator is heated (the adsorption concentration tubes are heated for one minute to 250°C, and the low temperature concentration tubes are heated to 90°C), and the target measurement substance is removed, and reconcentrated in the cooled cryofocusing trap using liquid nitrogen.

### ③ Sample introduction

In equipment which uses an air tube as a cryofocuser, the air tube is heated for a fixed period and the VOCs removed, an analysis column is introduced, and the GC temperature program is started.

The column tank is cooled, and analysis is started using equipment to cool the tip of the analysis column, by starting the GC column tank temperature increase program.

### ④ SIM method

- The measured flow volume (see Table 9.8.1 for reference examples) is set for each target measurement substance.
- The chromatogram is recorded to both confirm quantitative mass fragment and fix characteristic mass fragment for each of the target measurement substances that were set in a), and a comparison between the two is sought.
- The peak surface area or peak height of the target measurement items detected, and either the peak surface area or peak height of the internal standard are measured, a comparison between the two is sought and, using the previously compiled ⑥, the volume of the target measurement substances in the specimen (As: ng) is determined.

Table 9.8.1 Volume for measuring GC/MS in VOCs

Compound	Primary key Ion	Secondary Key Ion
Acrylnitril	52	53
Vinyl chloride monomer	62	64
Dichloromethane	84	86, 49
1, 2-Dichloromethane	62	64
Tetrachloroethylene	166	164, 129
Trichloroethylene	130	132, 95
Benzene	78	77
1, 3-Butadiene	54	53, 51
internal standard	98	
Toluene d8	96, 70	
Fluorobenzene	117	
Chlorobenzene d5		

### ⑤ Scan method

- Set the measurement parameters.
- Record a total ion chromatogram (TIC) for the conditions set in a).
- Either the peak surface area or the height corresponding to both the internal standard and each target measurement substance on the TIC are determined. Either the peak surface area or the height is compared for both the internal standard and each target measurement substance and, using the calibration curve compiled in advance by the ⑥, the volume of each target measurement substance in the sample (As: ng) is determined.

⑥ Compiling the calibration curve

- a) After directly introducing a fixed volume of both the mixed control gas and the control gas to the condenser on the sample introducer, an operation is conducted from ② to either ④ or ⑤, and the chromatograph for each target measurement substance is recorded. The volume of the mixed control gas introduced is made to correspond with the volume range, and changed to five or more stages (including zero), after which the above operation is repeated.
- b) The volume infused into the GC/MS for each target measurement substance from the mixed control gas that was used for the calibrations in a) select the midrange of the calibration curve, and both target ion and the secondary ion chromatograms are recorded for each target measurement substance. Using either the peak surface area or the peak height for each, a comparison of the strength is determined between the target ion and the secondary ion for each substance.
- c) The relative strength of either the peak surface area or the peak height for both the target ion and the secondary ion are determined for each target measurement substance at each of the concentrations, and the fact that they agree with the relative strengths of each target measurement substance as sought in b) is checked. The difference between the peak surface area or the peak height for both each target measurement substance and the internal standard is determined, and the calibration curve is compiled by comparing the volume of each target measurement substance and either the peak surface area or the peak height of each target measurement substance.

⑦ For the blank test, in accordance with ②, following the introduction of a humidified zero gas to the container, both the sample and the same quantity of humidified zero gas are concentrated by the concentrator, operations are performed from ② to either ④ or ⑤, and the blank value ( $A_b$ : ng) is measured.

(3) Calculating the concentration

From the results obtained in (2) in the preceding paragraph of ④ and either ⑤ or ⑦, the concentration of each target measurement substance within the sample is calculated using Eq. (2).

$$C = \frac{n(A_s - A_b)}{V \times \frac{293}{273 + t} \times \frac{P}{101.3}} \dots\dots\dots (2)$$

Here,

$C$  = Concentration of each airborne target measurement substance at 20°C ( $\mu$  g/m<sup>3</sup>)

$n$  = Dilution rate (in the case of reduced pressure collection)

$A_s$  = Volume of each target measurement substance in the sample gas determined by the calibration curve (ng)

$A_b$  = Volume of each target measurement substance in the blank gas determined by the calibration curve (ng)

$V$  = Sample gas volume supplied for analysis (ℓ)

$t$  = Temperature during sample analysis (°C)

$P$  = Air pressure during sample analysis (kPa)

### 9.8.3 Formaldehyde and acetaldehyde analysis methods <sup>1)-3)</sup>

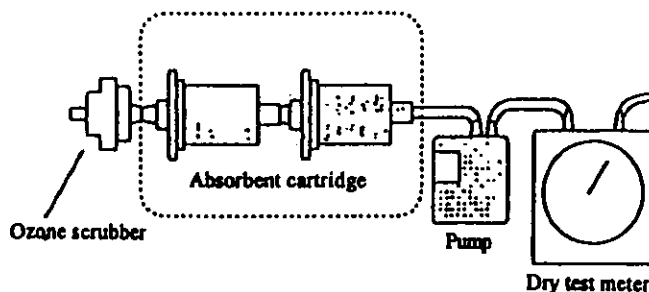
#### (1) Sample collection and sample solution adjustment

A fixed volume of about 1 mg of 2, 4-DNPH (2, 4-Dinitrophenyl hydrazine) is placed in a resin tube (internal diameter 10 mm, length 20 mm) and covered with 350 mg silica gel with particles of 50 to 250  $\mu$  m in diameter. This is then designated the specimen collection tube. In addition, in the case of a comparatively high concentration, the specimen is trapped in a 2, 4-DNPH solution and, following extraction with carbon tetrachloride, is subjected to either gas chromatography or the fixed volume method <sup>1) 2)</sup>.

##### ① Collection method

Fig.9.8.2 shows a typical collector. The absorbent cartridge tube and the ozone scrubber are both opened, and the ozone scrubber, absorbent cartridge, pump, and gas meter are connected, and collection is continued at 0.1  $\ell$ /min flow for 24 hours. After the sample has been collected, the absorbent cartridge is stoppered tightly.

The contents of the absorbent cartridge should be sent for extraction as fast as possible. The ozone scrubber should not be reused.



Source: Shimadzu catalogue No. G179

Fig.9.8.2 Sampling of aldehydes

##### ② Test solution adjustment

The absorbent cartridge is removed from the storage container and, following removal of the stoppers at both ends, the top end is connected to a solution syringe (10 ml) holding 5 ml of acetonitril, which is then passed gently into the absorbent cartridge at a flow rate of 1 ml/min, and subsequently allowed to flow into a (graded) full volume flask containing 5 ml of the aldehyde hydrozone conductor  $\ell$ .

The acetonitril is added to the end solution, brought into line with the standard curve for the full flask, stoppered tightly, and shaken well to mix it. This solution is then separated into two vials, and one vial is used as the HPLC (High Performance Liquid Chromatograph) test solution. The remaining vial is stored in a refrigerator until its analysis values can be checked.

③ For the blank test, the ② operation is carried out on a absorbent cartridge from the same lot, but in which no specimens have been collected, and the blank test solution is thereby adjusted.

#### (2) Test operation

① Set the HPLC analysis conditions and adjust the equipment.

The HPLC analysis conditions should be set appropriately with reference to the following:

Separation column : ODS, 5  $\mu$  m

4.6 mm  $\times$  50 mm + 4.6 mm  $\times$  250 mm

Mobile phase: Acetonitril: water/ 60 : 40

Flow volume: 1.0 ml/min.

Sample injection volume: 20  $\mu$  l

Column temperature: 40°C

Detector: UV (360 nm)

② Sample solution measurements

(1) ② Approximately 20  $\mu$  l of the sample solution, which was adjusted by ② in (1), is extracted using a microsyringe, infused into the HPLC, and its chromatogram recorded, and either the peak surface area or the peak height is determined for the FA-2, 4-DNPHz, and AA-2, 4-DNPHz sustainable peak period.

Using either the peak surface area or the peak height for the FA-2, 4-DNPHz, and AA-2, 4-DNPHz, the substance volume ( $A_s$ ; ng), which was converted as formaldehyde and acetaldehyde from the calibration curve that was compiled earlier, is determined.

③ Compiling the calibration curve

- a) 0 to 2 ml of the prototype control solution mix (each 10  $\mu$  g/ml) of formaldehyde and acetaldehyde are removed in steps to a total container flask (10 ml), adjusted to a fixed volume using acetonitril, and the control series for compiling the calibration curve is compiled. The control series has five stages, including zero.
- b) The ② operation is carried out, and either the peak surface area or peak height is determined equivalent for both the formaldehyde and the acetaldehyde.
- c) The calibration curve is then compiled from the peak surface area or peak height and the flow volume (ng) for both the formaldehyde and the acetaldehyde.

④ The blank test solution is infused into the HPLC, the ② operation is carried out, and the blank value ( $A_b$ ; ng) is determined.

⑤ As for the fluctuation sensitivity test, a control solution close to the middle of the calibration curve is infused into the HPLC, and the ② operation is carried out. This operation is carried out at least once for every 10 sample measurements.

(3) Calculating the concentration

The concentration of each airborne target measurement substance is calculated using Eq. (3) from the ② and

④ Obtained in (2).

$$C = \frac{(A_s - A_b) \times E \times 1,000}{V \times V \times \frac{293}{273 + t} \times \frac{P}{101.3}} \dots \dots \dots (3)$$

Here,

$C$  = Concentration of for maldehyde and acetaldehyde the atmosphere at 20°C ( $\mu \text{ g/m}^3$ )

$A_s$  = Volume of each target measurement substance in the sample solution determined by the calibration curve (ng)

$A_b$  = Volume of each target measurement substance in the blank solution determined by the calibration curve (ng)

$E$  = Test solution volume (mℓ)

$V$  = Solution volume infused into HPLC ( $\mu \text{ ℓ}$ )

$V$  = Absorbed air volume measured by the gas meter (ℓ)

$t$  = Mean air temperature during sample collection (°C)

$P$  = Mean atmospheric pressure during sample collection (kPa)

## 9.9 Dioxin analysis methods

### 9.9.1 Introduction

The substances called either dioxin or dioxins are the general name given to polychlorinated dibenzo-*p*-dioxins PCDDs and to polychlorinated dibenzofurans PCDFs, whereby PCDD has 75 isomers, and PCDF has 135 isomers. Because dioxins are severely toxic substances 5,000 times more poisonous than potassium cyanide, the analyst must undergo special training in its handling. Sample should be processed in a well-managed laboratory facilities, written in 9.1.7 GC/MS with a high resolution function (¥60 million to ¥100 million) and standard substances (¥10 million to ¥80 million), and waste substance processing and storage facilities are essential.

Moreover, because analysis handles values at an extremely low level of concentration, it is important to establish accurate data processing and quality control. Laboratories with just such a system are limited to special facilities in developed countries.

### 9.9.2 Sampling

#### ① Ambient air sampling <sup>1)</sup>

A collection tube (inner diameter 84 mm × length 200 mm) with two sheets of polyurethane foam fitted is attached to the latter stage of a high volume sampler filter, as described in 9.6, and suction is continued for approximately 24 hours at a rate of 700 mℓ/min.

Both the filter and polyurethane foam shall be treated by heating and washed for a minimum of 16 hours in Soxhlet to remove all of the interfering substances prior to the sampling. To assure the data accuracy, both filter and polyurethane foam from the same lot must be prepared as a travel blank.

#### ② Exhaust gas sample collection <sup>1) 2)</sup>

The particulate components are isokinetically drawn from an emission source by collection apparants of Type 2 under regulation JIS Z8808 as stated in 8.1, and the filter holder is set to a maximum of 120°C.

The gaseous components are collected in an sorbent column (XAD-2 resin) and impingers. By both methods of the Ministry of Health and Welfare and of the Environment Agency, three impingers are connected in front of the sorbent column and two are connected behind, in series, as shown in Fig. 9.9.1, and two of the ones in front are filled with water (hexane sterile water), and at the fourth impinger is filled with diethylene glycol. Never use any grease on the connecting parts. Moreover, the US EPA's method 23 A <sup>4)</sup>, uses a condenser coil in place of an impinger, which is filled with water in front of the sorbent column, thereby changing from the collection methods of semi-volatile organic substances that have been used heretofore <sup>3)</sup>.

Specimen collection sets its standard at 3 m<sup>3</sup> or more (four hours average) as a rule, following the passage of a minimum of one hour after the burning has stabilized.

The filter and absorbent should be removed all interfering components washing by Soxhlet. Further, the rolles are regulated for both the sample storage and the blank sample in detail.



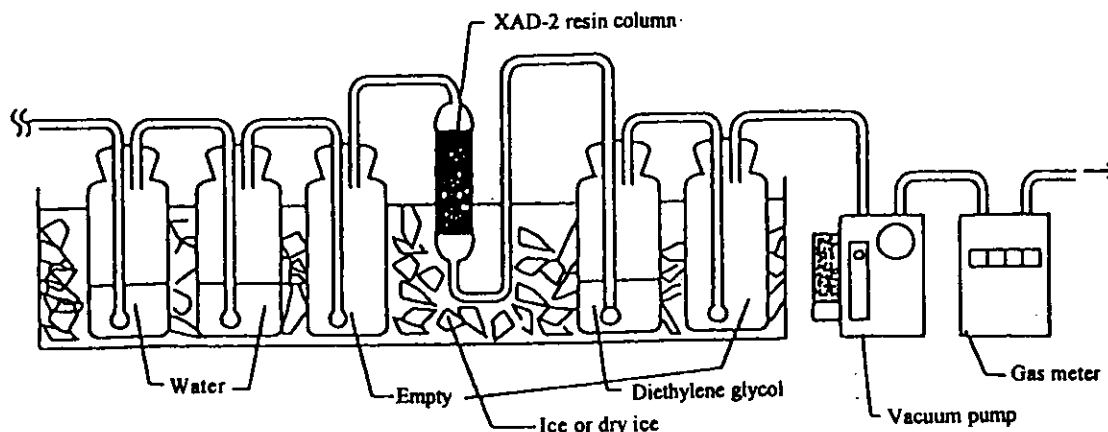


Fig.9.9.1 Configuration of exhaust gas sampling unit below the filter holder

### 9.9.3 Analytical operations

The dioxins in the collected sample are extracted individually, removed water, and concentrated, and cleaned up by means of different types of column chromatography, until it becomes colorless, the final solution is injected into high resolution GC/MS (HRGC/HRMS), and analyzed. Fig.9.9.2 gives an example of the analytical flow chart for emission samples described in the Ministry of Health and Welfare Manual <sup>2)</sup>.

The analysis is performed for all isomers of dioxins including the chlorine isotope <sup>37</sup>Cl, from tetrachlorides (T<sub>4</sub>CDD, T<sub>4</sub>CDF) to octochlorides (O<sub>8</sub>CDD, O<sub>8</sub>CDF), by means of the SIM (Selected Ion Monitor) method.

In order to identify and determine the dioxins by internal standard method, 17 types of C<sub>12</sub>-PCDD and PCDF are prepared as standard materials. As internal standard substances, 16 to 19 types of <sup>13</sup>C<sub>12</sub>-PCDD and PCDF, and <sup>37</sup>Cl-PCDD are used. Different isomers are used to add the internal standard for each sampling spike, cleanup spike, and syringe spike.

The cleanup spike uses a total of 10 types of PCDD and PCDF as an internal standard made by <sup>13</sup>C<sub>12</sub> for each chlorinate, which are added into the sample before the cleanup (however, in the US EPA method, as OCDFs are not added to avoid the interference, a total of nine spikes are used). These purpose are to make possible the ultra trace analysis by an isotope dilution method, and to check if the cleanup has carried out definitely through the recovery rate. Moreover, the US EPA method, prescribes that the internal standard is added immediately before the extraction operation and check the total recovery rate through the extraction and the cleanup operation.

The syringe spike is added immediately before the injection of sample into the GC/MS, in order to avoid the effect of the sensitivity change of the analyzer. The total concentration is formed from the relative concentration and the syringe spike responses.

In order to guarantee that the sample contamination is small, a sampling spike is added into the absorbent used, and its recovery rate is obtained. Further, all experimental procedures in the laboratory are conducted as same as actual samples. The total operation blank is analyzed, and the blank values are confirmed below the detection limit.

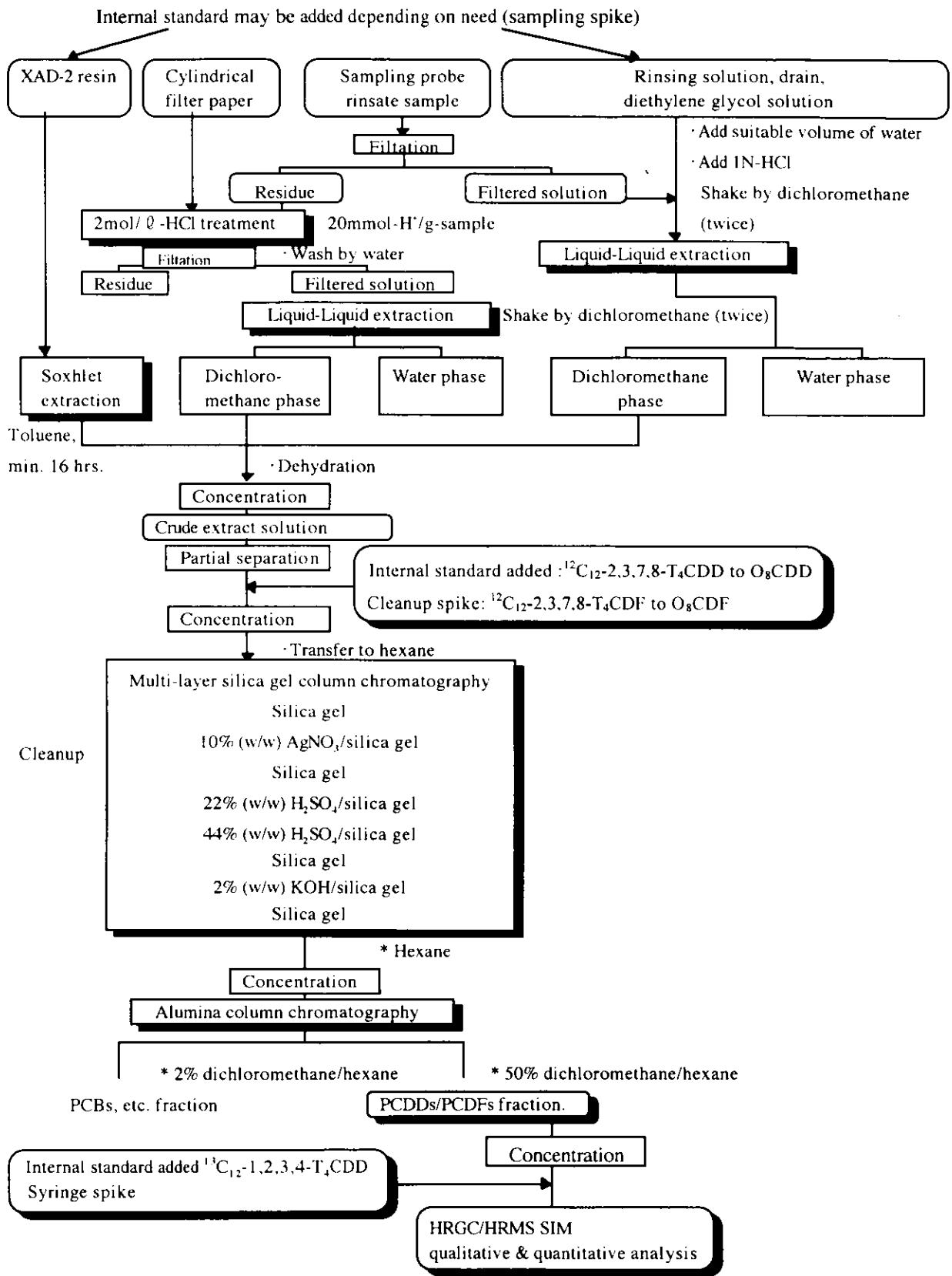


Fig.9.9.2 Analytical Scheme of Exhaust Gas Sample

#### 9.9.4 Indication of concentration

The measuring concentration is indicated for all homologues of the tetrachlorides and octochlorides as shown in Table 9.9.1. The concentrations of 2,3,7,8 position substitution isomers and the total of other isomers are indicated, together with the grand total concentrations of all other isomers.

Further, with regard to 17 isomers of extremely potent toxicity, the TEQ (Toxic Equivalents) are determined through multiplying the concentration by the TEF (Toxicity Equivalency Factor), and then totalized them. These results are expressed as pg-TEQ/m<sup>3</sup> in the ambient air, and ng-TEQ/m<sup>3</sup><sub>N</sub> (dry gas volume) for exhaust gasses.

Table 9.9.1 Indication of Dioxins

	PCDDs		PCDFs	
	homologue	Isomer	homologue	Isomer
4-chlorinated substituents	T <sub>4</sub> CDDs	2, 3, 7, 8 - Other	T <sub>4</sub> CDFs	2, 3, 7, 8 - Other
5-chlorinated substituents	P <sub>5</sub> CDDs	1, 2, 3, 7, 8 - Other	P <sub>5</sub> CDFs	1, 2, 3, 7, 8 - 2, 3, 4, 7, 8 - Other
6-chlorinated substituents	H <sub>6</sub> CDDs	1, 2, 3, 4, 7, 8 - 1, 2, 3, 6, 7, 8 - 1, 2, 3, 7, 8, 9 - Other	H <sub>6</sub> CDFs	1, 2, 3, 4, 7, 8 - 1, 2, 3, 6, 7, 8 - 1, 2, 3, 7, 8, 9 - 2, 3, 4, 6, 7, 8 - Other
7-chlorinated substituents	H <sub>7</sub> CDDs	1, 2, 3, 4, 6, 7, 8 - Other	H <sub>7</sub> CDFs	1, 2, 3, 4, 6, 7, 8 - 1, 2, 3, 4, 7, 8, 9 -Other
8-chlorinated substituents	O <sub>8</sub> CDD	1, 2, 3, 4, 6, 7, 8, 9 -	O <sub>8</sub> CDF	1, 2, 3, 4, 6, 7, 8, 9 -
Σ (4-chlorinated~8-chlorinated)	PCDDs	—	PCDFs	—

## 9.10 Simple NO, NO<sub>2</sub> measuring methods

### 9.10.1 Introduction

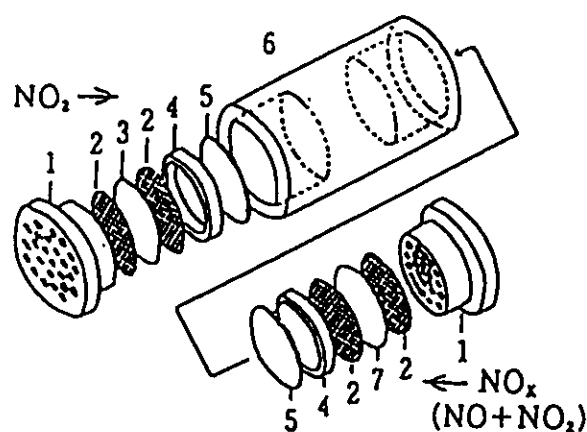
A simple measuring method centered around NO<sub>2</sub><sup>1)-5)</sup> is one of the methods concentrated upon in the field of epidemiological surveying that was looked at in 9.12. When a compound of TEA (triethanolamine), which is a collection reagent, is used with PTIO (2-Pheny 1-4.4.5.5-tetramethylimidazoline-3-oxide-1-oxyl), NO and NO<sub>2</sub> can be measured simultaneously.

This section focuses on simple methods of measuring airborne NO and NO<sub>2</sub> using a small sampler through the principle of molecular diffusion, using the above compound<sup>5)</sup>. Both the sampler and the peripherals for this method are available commercially, in addition to which, a batch personal sampler<sup>4)</sup> that is used for NO<sub>2</sub> measuring is also available.

### 9.10.2 Sampler construction and collection method

The sampler collector is divided into NO<sub>2</sub> and NO<sub>x</sub>, and the airborne NO and NO<sub>2</sub> both collected separately in the collector. The sampler construction is shown in Fig.9.10.1.

The NO<sub>2</sub> is collected in part 2 of Fig.9.10.1, while on the other hand, the NO<sub>x</sub> (NO + NO<sub>2</sub>) is collected in part 7 of the same figure, by which means, the collected NO volume can be determined simply by  $\text{Nox} - \text{NO}_2$ .



- 1 Polyethylene multiporous seal (cylindrical, with 25 absorption vents)
- 2 Stainless steel mesh (80 mesh)
- 3 NO<sub>2</sub> collection element
- 4 Teflon ring (2 mm thick)
- 5 Teflon panel (1.5 mm thick, 14 mm diameter)
- 6 Acrylic cylinder (15 mm internal diameter, 19 mm external diameter, 26 mm length)
- 7 NO<sub>x</sub> collection element

Fig.9.10.1 PTIO method NO<sub>x</sub> sampler construction<sup>5)</sup>

### 9.10.3 Sampler preparations

#### (1) reagent adjustment

##### ① NO<sub>2</sub> absorption solution (10% V/V TEA/acetone solution)

Triethanol amine (high quality) 20 ml dissolved in acetone (high quality), making 200 ml.

##### ② NO<sub>x</sub> absorption fluid (PTIO and TEA solution)\*

PTIO 0.3 g dissolved in NO<sub>2</sub> absorption fluid, making 10 ml.

\* Because PTIO separates even at room temperature, should ideally be prepared when necessary, and stored chilled.

#### (2) NO<sub>2</sub> collection element preparation

Place a cellulose fiber filter paper (Toyo No.50) with 14.5 mm diameter perforations on top of a Teflon mesh and, using a micro syringe, drip 50  $\mu$  l of NO<sub>2</sub> absorption solution onto the filter paper to make the NO<sub>2</sub> collection element.

#### (3) NO<sub>x</sub> collection element preparation

Place a cellulose fiber filter paper (Toyo No.50) with 14.5 mm diameter perforations on top of a Teflon mesh and, using a micro syringe, drip 50  $\mu$  l of NO<sub>x</sub> absorption solution onto the filter paper to make the NO<sub>x</sub> collection element.

### 9.10.4 Sampler assembly

Insert a Teflon board and a Teflon ring as deep as possible on both the left and right sides of the sampler. Next, squeeze both the NO<sub>2</sub> and NO<sub>x</sub> collection elements between two sheets of stainless steel mesh (80 mesh), insert one each into each collector, and seal tight and fix the polyethylene multiporous seal (see Fig.9.10.1 construction diagram). Attach this sampler unit to a metal safety pin clip and attach the fixing batch, then immediately insert the zippered polythene bag, and store and seal tightly in the Styrofoam bottle (a 70 ml wide mouth agent preserving anti-humidity container is suitable).

### 9.10.5 Collection of nitrogen oxides

- (1) Expose the sampler in a pre-examined site. An exposure time of 24 hours is standard. When the sampler has been exposed for 24 hours, it is possible to measure the concentration of NO<sub>x</sub> within the concentration range to several ppm from the ppb number.
- (2) When using measurements from the outdoor air, fix the sampler in the accompanying shelter, and in addition to avoiding direct sunlight, ensure that the sampler does not come directly into contact with any rain.
- (3) When using individual exposure volume measurements, fit the sampler with a hood before exposure.

### 9.10.6 Analysis methods

#### (1) Preparing the reagent

##### ① Sulfanil amide solution

Dissolve a mixture of 80g sulfanilamide (high quality), 200 ml phosphoric acid (high quality) and approximately 700 ml of water together in solution, and add more water to make 1 ℓ. Store in a cool, dark place.

##### ② NEDA solution

Dissolve 0.56g N-(1-naphtyl) ethylene diamine dichloric acid in 100 ml of water. Store in a cool, dark place.

##### ③ Colorproducing reagent

Mix 10 containers of sulfanil solution and 1 container of NEDA solution. Prepare when needed for use.

##### ④ NO<sub>2</sub> standard original solution

Heat sodium nitrite at 105 to 110°C for at least four hours, adjust accurately to a weight of 1.50g, and dissolve in water to make 1 ℓ. Include NO<sub>2</sub> 100 μg in the 1 ml solution.

##### ⑤ NO<sub>2</sub> standard solution

Dilute NO<sub>2</sub> standard original solution 100 times in water, take a further 0, 2, 4, 6, 8, 10 ml, dilute each of those with water, make 100 ml, and prepare the standard solution (0 to 1.0 μg NO<sub>2</sub>/ml) when needed for use.

#### (2) Analysis operation

- ① Remove with pincers the collection element together with the NO<sub>x</sub> and NO<sub>2</sub> collector wire meshes from the exposed sampler, and insert each of them into a 25 ml combined stopper test tube.
- ② Add 8 ml of water, and following extraction for 30 minutes, shake lightly to mix.
- ③ After cooling this at 2 to 6°C, add 2 ml colorproducing reagent while simultaneously agitating rapidly, and leave to cool for 30 minutes.
- ④ Return to room temperature, and measure the spectrometry at the maximum absorption wavelength lose to 545 nm.
- ⑤ Conduct an identical operation for the unexposed collection element, and measure the air test values.
- ⑥ Accurately take a fixed volume (8 ml) of NO<sub>2</sub> control solution (0 to 1.0 μg NO<sub>2</sub>/ml) that has been prepared in stages, add 2 ml of color reagent, conduct the same coloring operation and compile a calibration curve.

#### (3) Calculating concentration

NO and NO<sub>2</sub> concentrations are calculated using Eq. (1) and (2).

$$\text{NO concentration (ppb)} = \alpha_{\text{NO}} \times \frac{W_{\text{NO}_x} - W_{\text{NO}_2}}{t} \dots\dots\dots (1)$$

$$\text{NO}_2 \text{ concentration (ppb)} = \alpha_{\text{NO}_2} \times \frac{W_{\text{NO}_2}}{t} \dots\dots\dots (2)$$

Here,

$W_{NOx}, W_{NO_2}$  :  $NO_2$  volume (ng) collected by the  $NOx$  and  $NO_2$  collection elements determined with reference to the calibration curve

$\alpha_{NO}, \alpha_{NO_2}$  : ppb concentration conversion factor (ppb/min/ng)

$t$  : Exposure time (mins.)

Normally, conducted with air temperature  $20^\circ C$ , relative humidity 70%, and calculated as

$$\alpha_{NO} = 60$$

$$\alpha_{NO_2} = 56$$

## 9.11 Offensive odor measurement methods

### 9.11.1 Overview of offensive odor measuring methods

Offensive odor measurement methods can be classified into two, instrumental analysis methods, which use analytical equipment to measure offensive odor substances, and namely sensory tests that determine the strength of an odor from the human olfaction. Offensive odor can be seen from the point of view as sensory pollution, and sensory tests can be considered as a means of easily obtaining the sense of damage inflicted by the odor in question directly. But there are problems when it comes to the issue of results objectivity. On the other hand, instrumental analysis methods evaluate as their results the relationship between the concentration of each substance and the strength of odor (the Weber-Fechner's rule)<sup>1)</sup>, and are deemed a method of measuring offensive odors, but it is not easy to accurately determine the odor components from each source and evaluate the condition of that source. In this way, both instrumental analysis methods and sensory tests have their pros and cons, so by employing them both, a more accurate evaluation of the offensive odor can be made.

The Offensive Odor Control Law initially designated 22 offensive substances to be regulated, determining their concentration by instrumental analysis method, and basing its regulations on the concentration in question, but due to a revision of the law in 1995, it was decided to use sensory tests (triangle odor bag method) in conjunction with the equipment. Moreover, as an additional way of measuring offensive odors, odor sensors have come onto the market, but an odor sensor that can select the target substance and one that can be used by the general public has not yet to be developed.

### 9.11.2 Instrumental analysis methods

#### (1) Overview of instrumental analysis methods<sup>2) 3)</sup>

In many cases, offensive odor substances can be sensed as a "smell" even with extremely low concentrations. For this reason, the offensive odor substance often goes undetected if it is introduced to an analyzer as it is. Consequently, in measuring offensive odor substances, it is necessary to introduce an odor to the detector in concentrated form. Further, these are many substances mixing with the odor, so it is necessary to take the offensive odor substance alone by pretreatment, collect a sample selectively at the sampling stage, and to use a detector that has ability to select specific substance.

The measurement of offensive odors is comprised of the following processes: sampling, concentration, preprocessing, and analysis. In some cases, collection, concentration, and preprocessing are often carried out simultaneously, or some of the operations are abridged. The measuring methods can be classified by sampling method.

- ① The odor is collected in a container such as a bag or bottle (glass, stainless steel, etc.) (Vessel sampling method.)
- ② Collection of the odor in a solution that reacts with the odorous substance (Solution absorption method.)
- ③ Collection of the odor in a tube filled with an adsorbent coated with a reaction agent (Reactor tube method.)
- ④ Collection of the odor on filter paper coated with a reaction agent (Filter paper sampling method.)



⑤ Collection of the odor in a collection tube packed with an adsorbent (Adsorption method.)

Of these sampling methods, ② to ⑤ are carried out the concentration process at the same time. A separate concentration operation is only needed with the container collection method ①. ① targets the volatile components, and for the concentration method, basically a U-shaped glass tube packed with an adsorbent is cooled in a cooling medium such as liquid oxygen, and the odor substances, which have been collected in a container, are concentrated by passing it through the U-shaped glass tube (see Fig.9.11.1) <sup>4)</sup>. This glass tube is then heated, and the odorous components are introduced to the equipment (cold-trap method). In addition, measurement is possible by combining sampling methods ② to ⑤ in the container. Methods ② to ④ all target reactive compounds gas. In the case of an acid gas, a basic reagent is added to each medium, the sample gas is passed through the medium and collected through the acid/base reaction. The acid gas thus collected is added a strong acid, separated, and supplied for analysis. In the case of a basic gas, the opposite operation is performed. The ordinary temperature adsorption method ⑤ is suited to substances with a higher boiling point than the cold-trap method, and after passing the sample gas through a sampling tube at ordinary temperature, the substance is extracted by either heating the tube or using the solvent, and then introduced to the equipment.

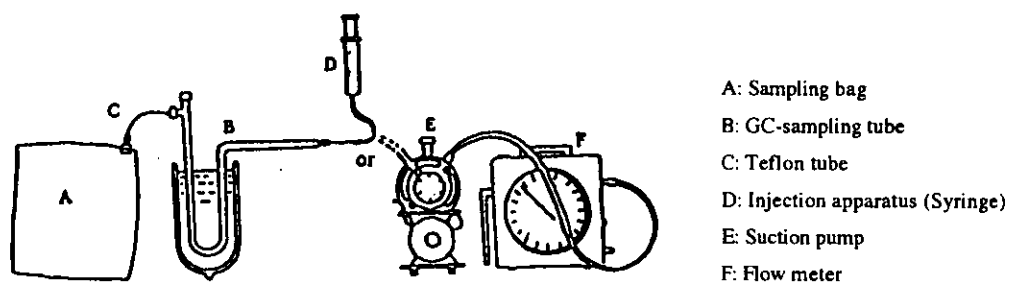


Fig.9.11.1 Overview of the cold-trap method <sup>4)</sup>

(2) Analytical instruments of offensive odor substances

The components of the odor are in a gaseous state, many components are mixed together, and detection sensitivity is high, for which reasons, either a gas chromatograph or a gas chromatograph-mass spectrometer is used. Such equipments are basically composed of an injection port, columns, and detector, and the sample gas that has been injected is separated into each of its component parts by the columns, and introduced to the detector for analysis. Both detectors which demonstrate ordinary responses, and detectors possessing selectivity to functional group are commercially available. Table 9.11.1 shows them in overview.

Table 9.11.1 Overview of gas chromatograph detectors

Detector	Target analysis substance	Detection limit (pg)	Linear range
Flame ionization detector (FID)	General organic compounds	50~	10 <sup>6</sup>
Flame photometric detector (FPD)	Sulfur and phosphorous compounds	10~	10 <sup>2</sup>
Flame thermionic detector (FTD, NPD)	Nitrogen and phosphorous compounds	10~	10 <sup>4</sup>
Electron capture detector (ECD)	Organohalogen, organic metals	0.1~	10 <sup>3</sup>
Photoionization detector (PID)	Unsaturated compounds	50~	10 <sup>6</sup>
Electron conductivity detector (ELCD)	Halogen, sulfur, and nitrogen compounds	5~	10 <sup>6</sup>
Mass spectrometer (MS) scanning method	General organic compounds	10~	10 <sup>6</sup>
Mass spectrometer (MS) SIM method	General organic compounds	0.01~	10 <sup>6</sup>

**(3) Analysis Methods of specified odorous substances**

The Offensive Odor Control Law determines measurement methods for 22 substances starting with hydrogen sulfide as substances targeted for regulation (specified odor substances). An outline of the measurement methods for each substance is shown in Table 9.11.2.

Table 9.1.1.2 Outline of the measurement methods for specified odor substances

Specified odor substance	Attached tables	Classification	Collecting method (sampling method)	Concentration method	Sampling volume (minimum determination limit)	Measurement method	Apparatus used
Ammonia	1	Site boundaries Exhaust outlet	Absorption method (Boric acid solution)	Solution collection (sample solution adjustment)	50 0 (0.05 ppm)	Absorption spectrophotometry	Absorption spectrophotometer
Hydrogen sulfide Methyl mercaptan Methyl sulfide Methyl disulfide	2	Site boundaries Exhaust outlet (excluding hydrogen sulfide)	Sampling bag (bag collection)	Cold-trap/heated introduction method	1 0 (0.0002 ppm) 1 0 (0.0002 ppm) 1 0 (0.0005 ppm) 1 0 (0.0005 ppm)	Gas chromatography	Gas chromatograph analyzer with flame photometric detector (FPD)
Trimethylamine	3	Site boundaries Exhaust outlet	Absorption method	Solution collection Alkali decomposition Cold-trap/heated introduction method	50 0 (0.0005 ppm)	Gas chromatography	Gas chromatograph analyzer with flame ionization detector (FID)
Acetaldehyde Propion aldehyde n-Butyl aldehyde Isobutyl aldehyde n-Valer aldehyde Isovaler aldehyde	4	Site boundaries Exhaust outlet (excluding acetaldehyde)	Sampling bag Cartridge method	2, 4-DNPH collection and sample adjustment	50 0 (note)	Gas chromatography	Gas chromatograph analyzer with flame thermionic detector (FTD)
Isobutanol	5	Site boundaries Exhaust outlet	Sampling bag (bag collection)	Adsorption/heated introduction method	2 0 (0.0005 ppm)	Gas chromatograph-mass spectrometer	Gas chromatograph-mass spectrometer which has electron shock ionization (EI method) (SIM method)
Ethyl acetate Methyl isobutyl ketone	6	Site boundaries Exhaust outlet	Sampling bag (bag collection)	Cold-trap/heated introduction method, Adsorption/heated introduction method	1 0 (0.01 ppm)	Gas chromatography	Gas chromatograph analyzer with flame ionization detector (FID)
Toluene Styrene Xylene	7	Site boundaries Exhaust outlet (excluding styrene)	Sampling bag (bag collection)	Cold-trap, Adsorption (heated introduction method)	1 0 (0.01 ppm)	Gas chromatography	Gas chromatograph analyzer with flame ionization detector (FID)
Propionic acid n-Butyric acid n-Valeric acid Isovaleric acid	8	Site boundaries	Adsorption method	Cold-trap/Adsorption (heated introduction method) Alkali beads collection, Formic acid injection/heated introduction method	25 0 (0.005 ppm) 25 0 (0.005 ppm) 25 0 (0.005 ppm) 25 0 (0.005 ppm)	Gas chromatography	Gas chromatograph analyzer with flame ionization detector (FID)

(Note): Acetaldehyde (0.005 ppm), n-Butyl aldehyde (0.005 ppm), Isobutyl aldehyde (0.005 ppm), n-Valer aldehyde (0.002 ppm), Isovaler aldehyde (0.002 ppm)  
 \* "Attached tables" indicate separate tables for measurement methods of specified offensive odor substances (Environment Agency report), and 1, 2, ... signifies table 1, table 2, ... etc.

#### (4) Other instrumental analysis methods

The measuring methods shown in (3) require equipment, preparation, and an experienced surveyor, and establishing of the sufficient measuring system is difficult. For this reason, the use of detection tubes that can be employed in a wide variety of fields such as work environment measurements and so forth in offensive odor analysis as a simple measuring method is currently under discussion. Of the specified odorous substances, detection tubes for use in the work environment have been improved for five substances: ammonia, methylmercaptane, hydrogen sulfide, acetaldehyde, and styrene, making them available for actual use. The specifications of the detection tubes for use of odorous substances are shown in Table 9.11.3<sup>5)</sup>. Moreover, sampling by using these detection tubes use large quantities of sample gas, and the tubes are connected to both a suction pump and an integrated flowmeter (a dry test meter).

Table 9.11.3 Specifications of the detection tubes for offensive odor use<sup>5)</sup>

Target substance	Manufacturer	Detection range (ppm)	Sampling gas volume	Reaction principle	Interfering gas	Discoloration state
Ammonia	G. Co.	0.5~5.0	750 ml	$2\text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4$	Discoloration of amines and diamines at 2 ppm or more. High error margin coexisting with 1/15 or more of $\text{NH}_3$	Pink→Yellow
	K. Co.	0.3~5.0	500 ml	$2\text{NH}_3 + \text{H}_3\text{PO}_4 \rightarrow (\text{NH}_4)_2\text{HPO}_4$	Amines	Pink→Yellow
Hydrogen sulfide	G. Co.	0.01~0.2	750 ml	$\text{H}_2\text{S} + \text{HgCl}_2 \rightarrow \text{HSHgCl} + \text{HCl}$	No effect on $\text{CH}_3\text{SH}$ , $\text{SO}_2$ , $\text{NO}_2$ , $\text{NH}_3$	Yellow→Purple
	K. Co.	0.02~0.2	1500 ml	$\text{H}_2\text{S} + \text{HgCl}_2 \rightarrow \text{HSHgCl} + \text{HCl}$	$\text{CH}_3\text{SH}$ has no effect at 0.02 ppm or below.	Yellow→Pink
Styrene	G. Co.	0.2~4.0	1500 ml	Concentration polymerization due to fuming sulfuric acid smoke emission	Patchy generation of a yellowish-brown color with butadiene 2 ppm, slight discoloration with tenfold or more of alcohol, ketone, ester, and aldehyde.	White→yellow
	K. Co.	0.4~4.0	2000 ml	Concentration polymerization due to fuming sulfuric acid	No effect at 10 ppm or less with each of acetone, benzene, toluene, xylene, or ethyl acetate.	White→yellow

#### 9.11.3 Sensory test methods<sup>6) 7)</sup>

Sensory test methods involving offensive odors are methods of measuring offensive odors using the human olfaction. The human olfaction has the advantage of catching all smells. Sensory test methods involving offensive odors are applied suitably as follows: methods that determine the dilution rate of odors (odor concentration, odor index) through methods of assigning a numerical value, and methods that determine the intensity of an odor. The triangle odor bag method is used as a way of determining the dilution rate, and methods using the six-state odor intensity scale are used as ways of determining the strength of an odor.

##### (1) Triangle odor bag method

As methods by which the odor is gradually diluted to determine the dilution rate at which the human olfaction can no longer detect the odor, the following have all been reported: the scentometer method, the olfactometer method, and the syringe injection method, but the triangle odor bag method, which solves the flaws in the syringe injection

method, is most widely used in Japan, and due to a revision in the Offensive Odor Control Law in 1995, has been introduced as a new method of measuring offensive odors.

The triangle odor bag method panels are passed 3  $\ell$  of odor bags filled with one odor and two odorless bags, and they select the odor-filled bag from among them by an operation shown in Fig.9.11.2. If the answer is correct, the same operation is repeated with an odor-filled bag that has been further diluted, and this experiment is repeated until an incorrect answer is given, by which specific method the dilution rate (odor concentration, odor index) is determined. The dilution method from a threefold series downwards. The methods for calculating the dilution index from the triangle odor bag method is shown in Fig.9.11.3.

Moreover, specimens are collected by either plastic bag (Mylar bag, Tedlar bag, etc.), or glass bottle.

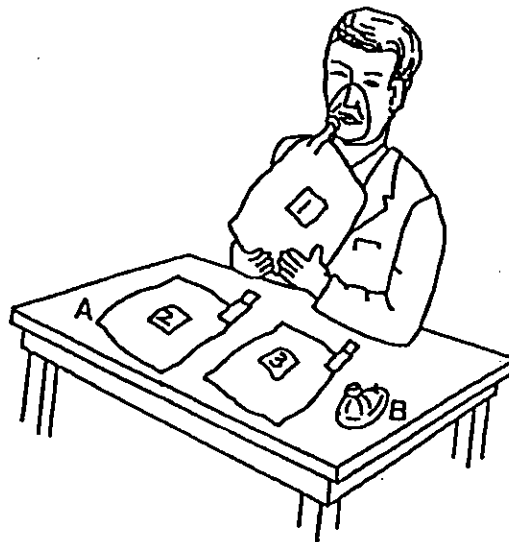


Fig.9.11.2 Triangle odor bag method operation <sup>7)</sup>

Dilution rate		30	100	300	1,000	3,000	10 million	Threshold value for each panel	Cutting of Max. and Min. values
log. values		1.48	2.00	2.48	3.00	3.48	4.00		
Panels	A	/	/	○	○	×		3.24	Cut Cut
	B	/	/	○	×				
	C	/	/	○	○	○	○		
	D	/	/	○	○	○	×	3.74	
	E	/	/	○	○	×	3.24		
	F	/	/	○	○	○	×	3.74	

Fig.9.11.3 Method of calculating the triangle odor bag method <sup>6)</sup>

(2) Six-stage odor intensity

The measurement of odor intensity is a method of assigning numerical values based on a measurement that shows in tabular form how the strength of an odor is sensed when the surveyor directly inhales that smell in the

environment. The measurement method is to plot on a graph as shown in Fig.9.11.4<sup>6)</sup> the strength of an odor directly smelled at ten-second intervals by three or more surveyors at each survey site. This operation is repeated continuously for five minutes at the same survey site. The mean value of the surveyors is calculated for each time period, and the strength of the odor at the measuring site is evaluated.

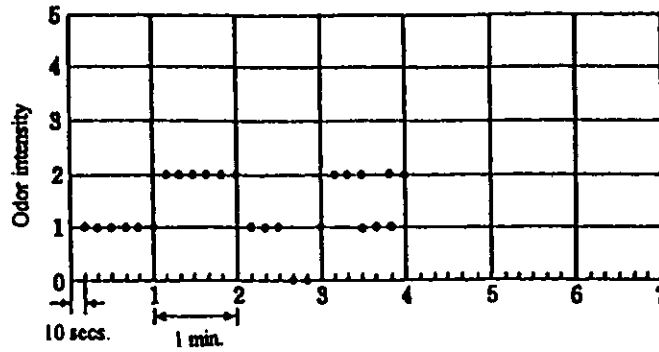


Fig.9.11.4 Six-stage odor intensity scale<sup>6)</sup>

### Calculating the odor index

#### (1) Environmental specimens

The odor index is calculated according to the following formula. However, if the mean correct rate concerning the initial dilution ratio is less than 0.58, the odor index values are displayed as less than  $10 \log M$ .

$$Y = 10 \log \left( M \times 10^{\frac{\gamma_1 - 0.58}{\gamma_1 - \gamma_0}} \right)$$

With this formula, the mean correct rate is expressed when  $Y$  is the odor index,  $M$  is the initial dilution ratio,  $\gamma_1$  is the mean correct rate relating to the initial dilution ratio,  $\gamma_0$  is the initial dilution ratio times 10.

#### (2) Outlet vent specimens

A. The threshold values for each panel relating to the dilution ratio for the specimen odor are calculated according to the following formula.

$$X_i = \frac{\log M_{1i} + \log M_{0i}}{2}$$

In this formula,  $X_i$  expresses the threshold value for a given panel relating to the dilution ratio of the sample odor,  $M_{1i}$  is the maximum value from among the dilution ratio relating to the relevant odor bags when the panel in question has been selected with an odor bag attached, and  $M_{0i}$  is the dilutant value relating to an attached odor bag when either selection is dysfunctional or when an odorless bag is selected from the panel in question.

B. Of the  $X_i$  calculated for each panel, one of the maximum and minimum values each are excluded, the values other than the two excluded ones are added together, and the values obtained are excluded from the values reduced by two from the number of people on the panel.

C. The calculation is made using the following formula.

$$Y = 10 X$$

With this formula,  $Y$  expresses the odor index, and  $X$  is the value calculated in B. above.

## 9.12 Surveys into the impact on health (epidemiological survey methods)

### 9.12.1 Introduction

Surveys in order to examine the impact of air pollutants on health, are broadly divided into epidemiological surveys on people and animal experiments. The results of animal experiments cannot be directly extrapolated to human, but animal experiment is highly suited to study on the damage mechanism, because it is possible to expose animals to various dose of a single pollutant and, further, is also useful especially into carcinogenic experiments. This chapter devotes itself to human epidemiological surveys, leaving methods of animal experiment for another occasion, and is centered on especially attention which should be paid when conducting those surveys into the impact of air pollution on human health.

### 9.12.2 Steps of epidemiologically evaluating air pollution

Pulmonary diseases induced by air pollutants are non specific diseases, and in many cases difficult to elucidate the causal relationship between air pollution and these diseases. For this reason, it is important to pay sufficient attention to the epidemiological design. Generally, a hypothesis is established as the epidemiological approach, and then examined for proof. Steps to this end can be cited as follows:

- ① Adequate understanding of the characteristics of the subjects.
- ② Understanding of the air pollutants exposure conditions cited hypothetically as being related to cause and effect.
- ③ Understanding of the distribution and frequency of the disease and the damage to health thought to be related to the cause and effect.
- ④ Understanding of the dose-response relationship, and estimating and determining the causal relationship.
- ⑤ Discussion of suitable countermeasures and preventive measures.

### 9.12.3 Actual process of epidemiological survey

#### (1) Preparing a protocol of survey

In order to examine the validity of the hypothesis, both a design and a protocol (written survey plan, written action plan) are necessary. Because the survey often involves many people, ideally everything should be put in writing, and all of the people can share and mutually understand the contents of the survey.

#### (2) Selecting subjective group

If examining the hypothesis that air pollution is bad effect on the health, the subjects should be selected from a region of high concentrations of air pollution, and the compared group should be from the countryside, or other areas where there is little air pollution. In this case, it is essential to understand the characteristics of the population. For example, it is important if the region does or does not have many work sites where people are exposed to the various causes of pulmonary disease. Further, there is the problem of disturbance factors such as smoking, age, sex,



and so forth. To process these out, the disturbance factors are accepted as they are, and the same methods and stratified sampling method as intended for the control group such as social class and age are stratified and equalized, thereby eliminating the disturbance factors.

### (3) Selecting the subjects of the survey

When the survey subjective group has been decided upon, a survey of the people belonging to that group is conducted. If possible, all subjects in the group should ideally be surveyed, but in many cases, a sample survey is conducted, due to the pressures of budget and staff numbers. Sampling methods are as follows: Case control study, and random sampling. Regardless of which, it is important that the subjects represent the population as far as possible. The proportion of numbers selected to the population is also important. During the analysis, it is important whether the examination is statistically correct or not. For this reason, because the necessary number of subjects can be determined by calculating backwards, if possible, it should be determined on that basis.

### (4) General items of a survey

- \* Items relating to the person: Sex, age, human race, physical condition, past history, social and economic status, residential history, occupation, religion, education, personal habits (smoking, dietary habits, etc.)
- \* Items relating to the person's residential environment: Residential site (beside a road, near a factory, etc.), detached house, multiple dwellings, airtight, age of building, types of coolers and heaters, etc.
- \* Items relating to the environment: Temperature, humidity, wind direction, topography, etc.

### (5) Survey into the respiratory symptoms

#### \* BMRC (British Medical Research Council) questionnaire

Following the problem of excessive deaths due to smog in London, the BMRC developed its standard questionnaire as a method of epidemiologically understanding chronic bronchitis, and published it in 1960. It was subsequently revised and the version actually in use dates from 1976. Because the BMRC questionnaire is for an interview, in order to obtain uniform results, the interviewers are required for training. To date, it has been translated into approximately 12 languages, but it is important to use appropriate terms suited to the customs and habits of the people in question, and if one part is changed, its suitability also needs to be discussed.

#### \* ATS-DLD (American Thoracic Society-Division of Lung Diseases) questionnaire

This questionnaire was compiled by experts in the Division of Lung Diseases at the American Thoracic Society, and made public in 1978. Because the American people felt somewhat alienated by the terms in BMRC questionnaire, an adults' version and children's version were both compiled with reference to the original. It is a self-completed questionnaire concerning both asthma-type symptoms and chronic bronchitis. In Japan, a version that has been partially amended from the original is used.

#### \* Test of pulmonary function

Both of the above are surveys based on subjective symptoms, but a test of pulmonary function is conducted as an objective index. Although it takes both time and effort, both objectivity and changes across the years can both be pursued. The indices are vital capacity, forced vital capacity, the forced expiratory volume % in one second, the

maximum peak flow, and so forth. Further, airway hyperreactivity in asthma can also be examined.

\* Immunological test

Blood is collected, and both IgE (Immunoglobulin E) and its complement, which form the allergy reaction indices, are measured.

(6) Exposure assessment

In the case of air pollution, because it is difficult to understand the individual exposure dose within a certain period, in many cases the local measurement point values are replaced with exposure levels for the local population. Recently, in some surveys, a personal sampler is used, and it is attempted to understand their exposure dose, but such factors as the limits to the sample numbers and that measurements can only be taken over one or two days at best are proving to be problematic.

(7) Analysis and evaluation

When investigating a hypothesis, a null hypothesis is established from the survey results, and as an opposing hypothesis to counter this, the alternative hypothesis is selected when the null hypothesis is rejected, and if it cannot be rejected, then the null hypothesis can be accepted. If a statistical correlation is identified, the quality of that data is sufficiently good, and if the disturbance factors are excluded as far as possible, it is possible to presume causal relationship between air pollutants and diseases.

## 9.13 Field survey of effects on vegetation

As a method of evaluating the effects on plants, evaluation of the visible damage to the leaf surfaces, phytometer, the open top chamber method, and survey method for evaluating state of plants grown in the field are used.

### 9.13.1 Methods of indexing the appearance of visible damage (plant indicator)

Higher plants can not generally change their domain like animals do, so their growth is induced, due to environmental impact upon their habitat, or they change their lifestyle in response to the environment. Further, compared with animals, plant sensitivity to air pollutants is extremely high.

Plants that are highly sensitive to air pollutants, even a comparatively low concentration of air pollution results in the manifestation of visible damage to the leaf surface. For example, with the morning glory (strain "Scarlett O'Hara") visible damage is manifested with an ozone concentration of 4 ppb or above, and when the concentration increases above that level, the damage becomes proportionally great. When visible damage manifests on the leaf surfaces, plant growth thereafter is affected. In alfalfa, when 5% or more of the leaf surfaces are visibly damaged, not only does the rate of damage accelerate, but the growth in plant dry weight is linearly reduced <sup>1)</sup>.

### 9.13.2 Open top chamber method

With the Open Top Chamber (OTC) method, two transparent chambers with open ceilings are installed in the field. On one chamber, the surrounding air is introduced directly into the chamber, which is designated as the unfiltered plot. On the other one, clean air from which all of the airborne pollutants have been removed by means of an activated charcoal filter is introduced into the filtered plot. The growth of indicator plants grown within both chambers is compared, and the effect on plants of air pollution at a given site is evaluated. When the air at a site in which the OTC has been installed is polluted, visible damage occurs to the leaf surfaces of plants raised in the unfiltered plot, and dry weight growth of the plant and its total leaf area is retarded when compared with plants raised in the filtered plot. By comparing the growth reactions of plants in both plots, the effects of air pollution on plants in the field can be evaluated.

Fig.9.13.1 shows an outline of OTC <sup>2)</sup>. The chamber (30 cm long × 30 cm wide × 70 cm high) is made of transparent acrylic sheet. A hole is made in the bottom of the wooden pot to allow water to drain from the base (90 cm long × 45 cm wide × 30 cm high) for growing plants, and a suitable culture medium is used for growing plants in the pot.

As a plant indicator used in the OTC, when using plants that are highly sensitive to air pollution, even the extremely low concentrations of air pollution on-site can be detected from the plant reaction, and it is thus also possible to evaluate the air quality. For example, the radish plant, *Raphanus sativus*, highly sensitive to photochemical oxidants like O<sub>3</sub>, grows rapidly, and is comparatively easy to cultivate <sup>3)</sup>.

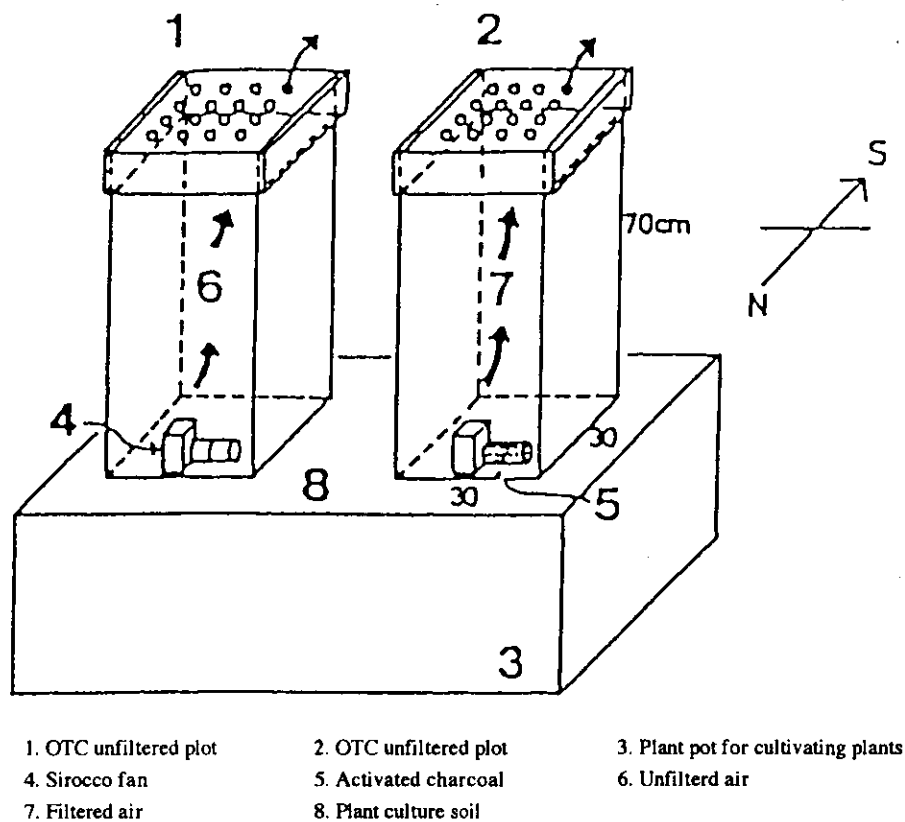


Fig.9.13.1 Outline of open top chamber (OTC) (Aihara et. al., 1988)

### 9.13.3 Phytometer

Among the survey methods of the air pollution effect on vegetation, phytometer can compare the growth rates of plants in the vegetative growth period in different regions. This requires potted young plants to be placed in each region under a fixed condition regarding inorganic nutrient status and water content of the soil. The dry weight of each plant is measured at regular intervals. From the obtained data the extent of the impact of air pollution on plants at the relevant site can be evaluated. As a plant for this purpose, the following characteristics may be cited:

- 1) Easy cultivation and synchronized growth in an initial growing stage;
- 2) A high sensitivity to air pollution, and resistant to disease and insect damage;
- 3) Possibility of evaluation in a relatively short period of time, such as from one week to one month, regardless of the season.

The plants hitherto used often are as follows: buckwheat (*Fagopyrum esculentum*), sunflower (*Helianthus annuus*), oats (*Avena sativa*), perennial rye grass (*Lolium perenne* L. cv. S23), and radish (*Raphanus sativus* L. cv. comet). In addition, trees such as Zelkova and Japanese cedar are used for long-term evaluation.

At the sites in which the potted plants are installed, the state of air pollution and meteorological conditions are also ideally measured, using equipment for simple measurements of air pollution, an accumulated solarimeter, maximum and minimum thermometers, and so forth.

Fig.9.13.2 shows an example of phytometer for field survey <sup>4)</sup>. Ten potted plants showing homogeneous growth are installed in a given site and then collected back in one to two weeks later. After measuring the leaf area, the plant is divided into foliage, stem, and roots, and the dry weight of each organ is measured. The initial sampling of plants is also necessary. As an indicator for evaluating the impact of air pollution on plants, the Relative Growth Rate (RGR) is determined.

RGR (g/g/day) can be determined by Eq. (1).

$$RGR = \frac{\ln W_2 - \ln W_1}{t_2 - t_1} \times 100 \quad \dots \dots \dots (1)$$

Here,  $W_1$  and  $W_2$  denote plant dry weight at initial sampling  $t_1$  and that at final sampling  $t_2$ , respectively.  $(t_2-t_1)$  is the number of days between two sampling dates, which is normally one or two weeks.

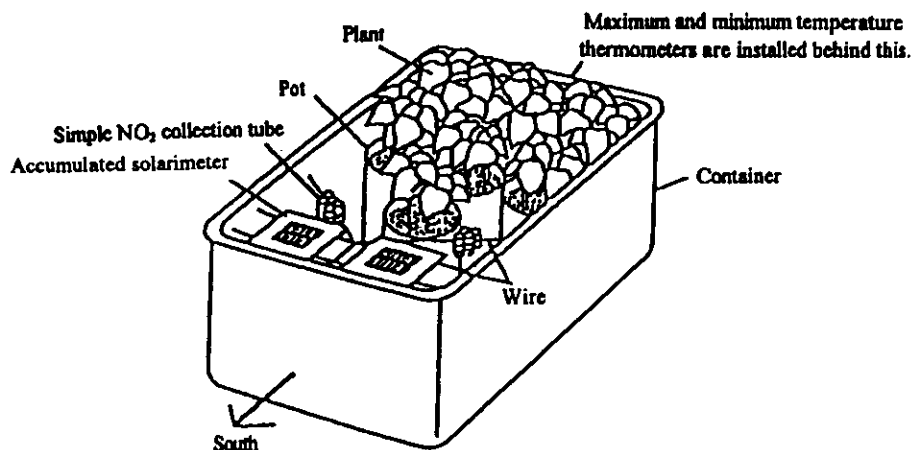


Fig.9.13.2 Example of a phytometer (Ushijima, 1981)

#### 9.13.4 Survey method for plants growing in the field

In order to evaluate the impact of air pollution on plants, as a method of monitoring plants growing under natural conditions there is a method which evaluates changes in the form of trees, and in the width of their annual rings evaluated.

Fig.9.13.3 shows an example of changes in form of tree by air pollution impact, where evaluation in fifth stages is shown in Japanese cedar and Zelkova <sup>5)</sup>. Further, Table 9.13.1 indicates an example of evaluation items for a tree vitality index <sup>5)</sup>.

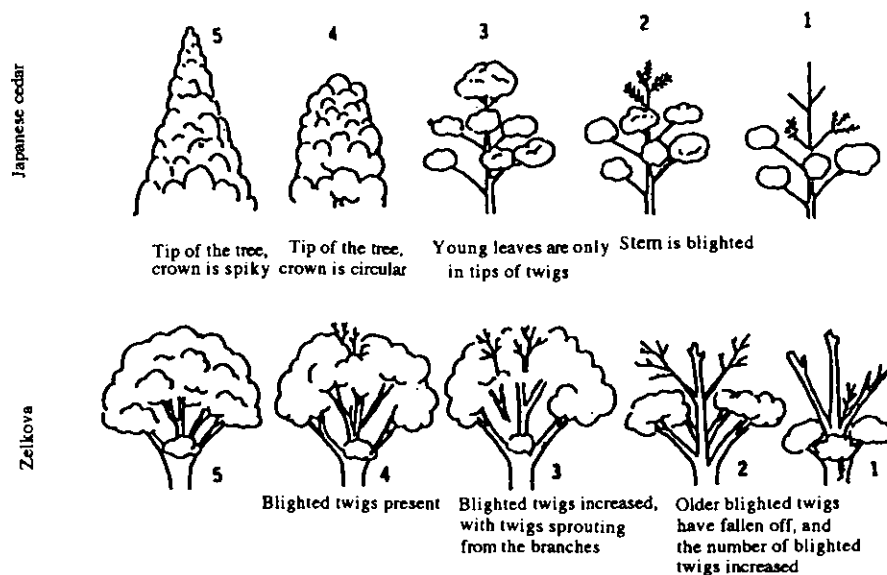


Fig.9.13.3 Decline scale of trees in Japanese cedar and Zelkova (Matsunaka, 1975)

Table 9.13.1 Evaluation standards for tree vitality index (Matsunaka, 1975)

Measurement items	Evaluation standards			
	1. Good, normal	2. Affected, but nearly normal	3. Deterioration is considerably advanced	4. Striking deterioration
Tree vitality	No damage	Slightly damaged	Evidently damaged	Severely damaged
Form of tree	Normal	Some branches lacking	Evidently wanting	Severely wanting
Branch growth	Normal	Somewhat reduced	Short and slender	Extremely short and slender
Dieback of stem	None	Slightly diebacked	Evidently diebacked	Completely dead
Density of foliage	Healthy foliage	Unbalanced foliage	Sparse foliage	Very sparse with dead leaves
Leaf shape	Normal	Slightly deformed	Evidently deformed	Completely deformed
Leaf size	Normal	Somewhat small	Evidently small	Strikingly small
Leaf color	Normal	Somewhat abnormal	Abnormal	Strikingly abnormal
Injury of leaves	None	Slight	Considerable	Striking
Red (yellow) leaves	Leaves shed normally in spring or autumn Annually	Slightly early compared with normal Annually	Leaves shed at odd times Biannually	Leaves shed at odd times Three or more times a year
Leaf shedding	Normal	Color slightly off	Leaves partially red and yellow, but color is bad	No red or yellow colors; leaves shed in dirty state

## 9.14 Survey on the Impact on Materials and Cultural Assets

### 9.14.1 Visible observation

The equipment used for on-site surveys are limited in scope to what is portable for a single individual. As a rule, survey paper and recording paper is prepared in advance, and the format is determined experimentally. The monument survey forms introduced by Tokyo are shown in Table 9.14.1.

Items to be carried

Camera (if possible, macro, with telescopic lens), compass, small polyethylene bag, plastic bottle, desalinated gauze, bamboo spatula, ion exchanged water 100 to 200 ml, a small syringe, a loupe, recording paper.

Table 9.14.1 Effect on Metal Statues, etc. Installed Outdoors: Survey Form

Installation site	( ) Contact address
Installed object's name	( ) Data
Installation date	Date: Survey date:
Material data	Metal (Material: Processing method: )
	Other (Material: Processing method: )
	Compound
Surface processing	No Yes (Type: )
Environmental conditions	Sunlight (good / bad) Trees: Obstacle: Illumination: Air pollution source: Ventilation: Meteorological conditions: Main wind direction, location of rivers and seas, etc.
Visible observation	Extent of damage: Severe medium minor
Photographic record	Vicinity, whole, partial, partial magnified
Preservation state	Whole: Partial:
	Surface: Preservation management: Cleaning methods, surface management, management body
Sampling	Precipitation flow, dent part residue, etc.

Note: Visible observation: Bronze statue (Damage severe: patina corrosion 80%<, damage medium: Verdigris corrosion 30 to 80%, damage minor: 30%>) Stone, concrete statue (Damage severe: Profile damage 80%<, damage medium: Profile damage 30 to 80%, damage minor: Profile damage 30%>)

### 9.14.2 Methods of measuring and analyzing deterioration <sup>1)</sup>

#### (1) Observation of outside

When collection of samples for analysis cannot be done and direct observation of the target alone is possible, it is necessary to divide them into ranks, as the first stage of outdoor monitoring, by standardizing the extent of deterioration of the sample by each material. Reference is also made to the method shown by the percentage area, as when evaluating photochemical plant damage, and methods whereby soot is measured according to the Ringelman's smoke chart.

#### (2) Measurement and analysis using equipment

X-ray diffraction, X-ray fluorescent analysis, color differentiation, and microscopic observation can all be applied with regard to samples obtained from the survey targets. If appropriate, chemical processing can also expand the methods of analysis. Atomic absorption analysis, emission spectroscopic analysis, radiation analysis, and ion chromatography can also all be applied.

#### (3) Measuring air pollution and meteorological factors

As much data as possible is collected on typical air pollutant concentrations and meteorological items that are considered to be responsible for the deterioration of the target materials. When it is impossible to use data collected by other organizations, the surveyor must take measurements directly themselves. Measurements of NO<sub>2</sub>, atmospheric temperature, and so forth, through the simple measurement method can be taken easily.

#### (4) Field exposure tests using standard samples

When it is possible to conduct comparative long term surveys, standard samples are prepared for each substance, and exposure tests are conducted on-site to examine the relationship between environmental factors and the extent of damage. From these results, a damage function can be compiled, which can also be used for assessments.

#### (5) Artificial exposure tests using standard samples

Controlled concentration exposure tests in an artificial meteorological chamber are used as experiments with the aim of quantitatively elucidating the effects of environmental factors on materials individually. Through this method, for example, the correlation between the extent of the damage, and sulfur dioxide concentrations can be quantitatively evaluated when artificially exposing just the sulfur dioxide in a given substance, for a certain time period, by concentration. By comparing these results with the field data, the accuracy of the mutual function can be improved.

#### (6) Reports

The format of the report is also extremely important to secure the standards of the survey, and it is necessary to prepare this in advance depending on the survey objectives.



## 9.15 Meteorological observation measurements

### 9.15.1 Introduction

Air pollution is closely related to the meteorology of lower atmosphere. Here the meteorological measuring methods which are necessary to understand the movements of air pollution and for which no particular special knowledge or technology is needed have been chosen for explanation. Further, an overview of surface and high altitude meteorological observation that is currently taking place is also shown.

### 9.15.2 Surface meteorological observation

(1) Meteorological observation using buildings: Many meteorological observations are made using the rooftops of buildings. When installing a meteorological tower, it is important to select a site that avoids wind currents created by the building itself, and where there are no building obstacles in the vicinity. Further, topographically also, a site must be selected that preserves the regional character. As measurement items, wind direction and wind velocity, temperature, humidity, solar intensity, intensity of the ultraviolet radiation, and air pressure must all be measured automatically.

(2) Meteorological measurements using a tower: Meteorological observation using a tower or chimney are often used as a method of learning the meteorological characteristics of that particular region. Depending on the wind direction, the meteorological tower can be affected, so that ideally, two or three identical measuring systems should be installed at the same height, and depending on the main wind direction, measurement values should be used from the itself can affect the result equipment that remains unaffected. Data on the vertical distribution of the ambient temperature is extremely valuable because it elucidates the frequency of manifestation of the ground surface inversion layer. Further, by installing constant air pollution monitoring equipments meteorological observation using a tower will make it possible to comprehend in detail the region's composition of air pollution.

### 9.15.3 Upper air meteorological observation

(1) "Pilot balloon" observation: "Pilot balloon" observation is a simple and easy method for learning the wind direction and wind velocity in the upper atmosphere. Because their practicability is also extremely high, they are introduced here in detail. The method is as follows. First a balloon, called a pilot balloon, generally weighing 20 g and filled with helium, is allowed to rise after its buoyancy has been adjusted. Next a balloon theodolite follows the moments of the balloon and measures the azimuth and the angle of elevation at fixed time intervals. The wind direction and wind velocity in the upper atmosphere are determined from changes in the numerical values.

The buoyancy is adjusted in a site on ground surface with no wind, using a plumb bob. When the speed of elevation is set to 150 m/min., a 57.6 g bob is used. At night, a water battery (13.3 g in weight in its water-laden condition) is used which emits light when being soaked in water and current flows through it. At this time, a 74.9g

bob is used. When the speed of elevation is set to 200 m/min., bobs of 136.4 g and 154.8 g are used respectively. Since the speed of elevation is given, when readings are taken 20 seconds apart, if the speed of elevation is 150 m/min., the mean wind speed and wind direction are determined every 50 m in height.

(2) Non-lift balloon observation: This is used to determine the wind currents and the extent of their vertical turbulence at a fixed height. The movements of a non-lift balloon that has been balanced, between its buoyancy and its weight are used. Its movements can be followed by the signals sent from a transmitter fitted to the balloon to the earth, or helicopter can be used for following.

(3) Radio sonde observation: With pilot balloon observation, only the wind velocity and wind direction can be determined. However by using low-level radiosonde, vertical distributions of temperature, humidity, can be determined for up to two to three kilometers. The temperature distribution is indispensable in order to learn the height of the temperature inversion layer and the stability of the air. Further, by learning the vertical distribution of the humidity, it is possible to accurately determine the ceiling height and the history of the air layer above.

(4) Rawinsonde observation: This system can simultaneously measure the wind direction and wind velocity by catching the movements of the radiosonde by an antenna which follows them automatically. In the case of pilot balloon observation, measurements from the upper air above the clouds can obtain data even when visibility is poor or there is cloud cover, by implementing rawinsonde observation for data that otherwise could not be obtained.

(5) Ozonesonde observation: This system can measure vertical ozone distribution up to 30 km high by further attaching ozone measuring equipment to the rawinsonde measuring system. The ozone sensors use a potassium iodide solution. Terrestrial sensor adjustment and calibration is required.

(6) Kytoon observation: A kytoon is a large balloon shaped like an airship, and possessing buoyancy. By operating a winch on earth and raising and lowering the kytoon sensor, the kytoon can be raised to the target height, and measurements taken. Kytoons come in all different sizes, from small, lightweight models that measure air temperature, humidity, and ozone, to those that measure wind current composition, and wind speed and wind direction. Signal transmission can be either by wireless, or by fiberoptic cable. By using the large model kytoons, an air sampling pipe can be raised, and it is possible to take all types of air pollutant measurements using automatic air pollutant measuring equipment installed on earth, while sampling the air of the upper atmosphere. The measurement height is approximately 1 km, but at times of strong wind or rain, measurements are problematic.

(7) Aircraft observation: In order to understand the layered distribution of air pollution, and the meteorology thereof, the most effective measurements can be taken by using an aircraft. Aircraft used include aircrafts with fixedwings, helicopter with rotary wing, and airships. According to aviation law, generally measurements may not be taken below 300 m. Fixed-wing aircrafts are used for measurements over a wide area, helicopters for measurements centering on vertical distribution, and airships prove to be most effective for measurements at low speed. In any

case, the greatest problem is a stable power source.

#### 9.15.4 Continuous remote sensing of upper atmosphere meteorology

(1) Laser radar: If changes over time to the mixed layers of the atmosphere can be understood, it would be useful in monitoring and forecasting air pollution. To this end, a Mie scattering laser radar can be used. Mie scattering is the name given to the diffusion of light due to airborne particulate matter, but this system determines the vertical distribution of particulate matter in the upper atmosphere by receiving and amplifying through a telescope on the ground the Mie scattering intensity following the firing of a laser into the upper atmosphere. The possibility of installing this system in a small van or other such vehicle has already been made practicable, and it is now possible to make continuous measurements to a height of 3 km.

(2) Doppler acoustic radar: Information pertaining to the wind velocity and direction in the upper atmosphere are most useful in elucidating the movements of air pollution. The movements of vertical wind direction are especially important. By using Doppler acoustic radar, it is possible to determine in real time the composition of wind in the upper atmosphere. Doppler acoustic radar emits a sound wave into the upper atmosphere, which is reflected back by airborne particulate matter, and from changes in the sound frequency it is possible to determine both the vertical wind velocity and the horizontal wind velocity. Specifically, with a 2,100 Hz, 30 W output, measurements can be made up to approximately 1,000 m aloft. Measurement accuracy is good, and it is possible to measure up to weak breeze with approximately 0.01 m/s velocity both in the horizontal and vertical directions. By increasing the output, an even higher altitude measurement can be obtained, but it is important to consider the sound disruption to the neighborhood. Further, measurements at a site where major noise is generated in the surroundings are interfered with by that noise.

#### 9.15.5 Meteorological network

The regional meteorological measuring system known colloquially as AMeDAS (Automated Meteorological Data Acquisition System) has been developed at approximately 1,300 sites throughout Japan, and the mean distance between stations in a square is 17 km. Of these, approximately 840 sites measure wind velocity and direction, air temperature, duration of sunshine, and amount of precipitation, and the data obtained is useful in analyzing the regional distribution of air pollution. However, because weak breeze cannot be measured, the system is only suitable for measuring the movement of air pollution over a wide area. The mean distance between the stations with meteorological measuring functions for these four main in a square is 21 km. On the other hand, meteorological elements such as wind speed and direction, air temperature, and humidity are also continuously measured at atmospheric pollution regular measuring stations. Nevertheless, these measuring sites are concentrated in urban areas, and are insufficient for analysis over a wide area, but are extremely useful in the analysis of plane distribution of high concentrations of air pollution in urban areas when the breeze is weak.

In Japan, radiosonde observation is conducted two to four times a day at upper air observations. The

measuring times are at 3:00, 9:00, 15:00, and 21:00. The time of the upper air observation is based on midnight and noon of Greenwich mean time, which are 9:00 and 21:00 respectively in Japan.

### (9.1) References

- 1) S. Butcher and R. Charlson (Takashi Araki et al, trans.); *The Chemistry of Air Pollution* (1975)
- 2) Japan Society of Air Pollution (now the Japan Society for Atmospheric Environment) ed.; *Revised Air Pollution Handbook, Volume 1, Measurement* (1975)
- 3) Hori, Masahiro: *Measurement method of harmful factors* Japan Society for Safety Engineering ed.; *Safety Engineering Lecture 4, Health Damage in Workshop* p.149 (1982)
- 4) Kawaguchi, Hiroshi; *Detection Limits and Sensitivity*, *Bunseki*, 1982, No.1, pp.36 to 42
- 5) Sugimae, Akiyoshi; *Bunseki Kagaku* 28, p.559 (1979)

### (9.3) Literature cited

Air Quality Bureau, Environment Agency; *Ambient Air Continuous Monitoring Manual* (1980)

### (9.4) Literature cited

- 1) British Standards Institution: *Deposit Gauges for Atmospheric Pollution, British Standard 1747* (1951)
- 2) Department of Scientific and Industrial Research: *Measurement of Air Pollution*, Her Majesty's Stationery Office (1957)
- 3) Terabu, Motoji; *The Chemistry of Air Pollution*, Gihakudo (1951)
- 4) Air Pollution Measurements Committees: *Recommended Standard Method for Continuing Dust Fall Survey (APM-I. Revision 1)*, *J. Air Poll. Control Assoc.*, 16 (7), (1966)
- 5) Japan Environmental Measurement and Analysis Association; *Commentary of Environmental Measurement and Analysis Methods, Volume 1*, Maruzen (1984)
- 6) National Air Pollution Research Council; *Revised Air Pollution Handbook (r), Measurements*, Corona Ltd. (1975)
- 7) Terabu, Motoji (trans.), *British Science and Engineering Association (ed.); Methods of Measuring Air Pollution*, Gihakudo (1958)

### (9.6) Literature cited

- 1) Japan Environmental Measurement and Analysis Association; *Notes on Environmental Measurement and Analysis, Volume 1 (Air Pollutant Sample Collection Methods), Volume 2 (Air Pollutant Measurement and Analysis Methods)*, Maruzen (1984)
- 2) Air Quality Bureau, Environment Agency; *Air Pollutants Measuring Methods Indication* (1987)
- 3) Air Quality Bureau, Environment Agency, Air Regulation Section; *Hazardous Air Pollutants Measuring Manual* (1997)
- 4) Japan Environmental Sanitation Center; *Analysis and Forecasts of Suspended Particulate Pollutants* (1987)
- 5) Environment Agency Commission Results Report; "National Air Surveillance Network, Analysis of Suspended Dust and Suspended Particulate Matter", 1975 to 1996, Japan Environmental Sanitation Center

#### (9.7) Literature cited

- 1) Gotoh, Sumio, Matsushita, Hidetsuru, Shiozaki, Takuya, Kato, Yukihiko; Aroutin analysis of benzo (a) pyrene in airborne particulates by high performance liquid chromatography, *Bunseki Kagaku*, vol.30, 362 (1981)
- 2) Air Pollution Control Division, Air Quality Bureau, Environment Agency; Hazardous Air Pollutants Measuring Manual (1997)

#### (9.8) Literature cited

- 1) Japan Environmental Analysis Measurement Committee; Interpreting Environmental Measurement and Analysis Methods, Volume 2 (Methods of Measuring and Analyzing Air Pollutants), Maruzen (1984)
- 2) Air Quality Bureau, Environment Agency; Indicators in Air Pollutant Measurement Methods (1987)
- 3) Air Pollution Control Division, Air Quality Bureau, Environment Agency; Toxic Air Pollutants Measurements Manual (1997)
- 4) Kanagawa Prefecture Coastal Areas Air Pollution Survey Association; Survey of Atmospheric Concentrations of Fluorine and Organic Chlorides, Kanagawa Prefecture Coastal Areas Air Pollution Survey Report, pp.4 to 58 (1990)

#### (9.9) Literature cited

- 1) Air Quality Bureau, Environment Agency, Air Pollution Control Division; Hazardous Air Pollutants Measuring Methods Manual (October 1997)
- 2) Ministry of Health and Welfare, Hygiene Bureau Aqueduct Environmental Regulation Section; Standard Dioxin Measuring Methods in Waste Substance Disposal, (February 1997)
- 3) U.S. EPA: Method 0010, Modified method 5 sampling train (1986)
- 4) U.S. EPA: Method 0023A, Sampling method for polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofuran emissions from stationary sources (1996)
- 5) Shiozaki; Dioxins Analysis, *Bunseki* 1997 (1), pp.32 to 38

#### (9.10) Literature cited

- 1) A. C. Barton and H. G. McAdie; A cumulative survey technique for atmospheric nitrogen dioxide. In : 67th annual Meeting of the Air Pollution Control Association. Denver, Colorado, June 9-13, pp.1-20 (1974)
- 2) Sato, Shizuo, et. al., Methods of Measuring Relative Airborne Concentrations of NO<sub>2</sub>, Pollution and its Countermeasures, 13 (3), 52-57 (1997)
- 3) Simple Nitrous Oxides Measuring Methods Research Society; Research Concerning Methods of Measuring Relative Concentrations of nitrogen dioxide, Environment Agency-sponsored research paper (1977)
- 4) Nishimura, Hajime, Yanagizawa, Yukio; NO<sub>2</sub> Personal Samplers for Use in Measuring Concentrations in the Living Environment, *Journal of the Japan Society of Air Pollution* 8, 316-323 (1980)
- 5) Hirano, Koichiro, et. al; Simple Methods of Measuring NO and NO<sub>2</sub>, *Environment and Measuring Technology* 12, 32-39 (1985)

#### (9.11) Literature cited

- 1) Nagata, Yoshio, Takeuchi, Norifumi; Threshold Measurement Values for Offensive Odor Substances using The Triangle Odor Bag Method, Lecture Essays from the Japan Society for Atmospheric Environment, p. 528 (1988)
- 2) Kato, Tatsuo; Air Pollution Gas Chromatograph Technology, Sanyo Publishing (1975)
- 3) Ishikuro, Tomohiko, Kato, Tatsuo, Shigeta, Yoshihiro; Offensive Odor Equipment Measuring, Kodansha (1984)
- 4) Furukawa, Osamu, Hasegawa, Takashi, Namiki, Sho; Revisions to Measuring Methods of Offensive Odor Substances, Offensive Odor Research 24, pp.351-364 (1993)
- 5) Air Quality Bureau, Environment Agency, Special Pollution Section; Easy Offensive Odor Substances Measuring Manual (1990)
- 6) Ishikuro, Tatsuyoshi; Basic Odor Countermeasures and their Application, Ohm Co. (1997)
- 7) New Environment Management Facilities Dictionary Editorial Board; Anti-Air Pollution Equipment, Industrial Survey Council (1996)
- 8) Otaki, Toshikazu, Yamagata, Noboru.; Environmental Pollution Analysis Methods 12, Dainihon Tosho (1973)

#### (9.13) Literature cited

- 1) Totsuka, T.; Effects of sulfur dioxide on the growth of plants. Res, Rep, Natl. Inst. Environ. Stud. No.10, pp. 317-332 (1979)
- 2) Aihara, K., Omichi, S., Sinozaki, M., Totsuka, T., Yajima, I.; Small Open Top Chambers (OTC) for Evaluating the Impact of Air Pollution on Plants, Bulletin of Kanagawa Prefectural Environ. Center 10, 12-21 (1988)
- 3) Horie, H., Izuta, T., M. Takigawa, K., Miyake, T., Totsuka, T.; Evaluation of the Atmospheric Environment Using a Small Open Trop Chamber (OTC) as an indicator of Radish Growth, *Japan Society for Air Quality* 23, pp.284-289 (1988)
- 4) Ushijima, T.; Phytometer, Rep.of Jap. Environment Agency 1980, Manual of Environmental Impact Assessment through the Impact of Air on Plants (proposed), Separate volume, pp.64-70, Japan Public Hygiene Association (1981)
- 5) Matsunaka, S.; Bioindicator, Kodansha Scientific, p.182 (1975)

#### (9.14) Literature cited

- 1) Air Quality Bureau, Environment Agency, Guideline on Measuring Corrosion of Metallic Substances Due to Air Pollution, 1988

#### (9.15) Reference

- 1) Meteorological Handbook Compilation Committee; Meteorological Handbook, Asakura Shoten (1979)

## Chapter 10 Air Pollution Estimating Techniques

### 10.1 The Theory of Atmospheric Diffusion

As for smoke which is discharged into the atmosphere, advection by the wind and diffusion by the wind turbulence work to dilute it. The dilution by the advection of the wind is proportional to the wind velocity. Fig. 10.1.1 illustrates the dilution by the wind. The  $\bigcirc$  in the figure represents smoke as separate masses (called puffs). When we assume that the puff is discharged 1-second intervals, the distance between contiguous puffs is proportional to wind velocity. In other words, when the wind velocity is 1 m/s, the distance becomes 1 m and at a wind velocity of 2 m/s, it becomes 2 m. The distance between contiguous puffs shows the dilution effect. In another words, the concentration is inversely proportional to the velocity.

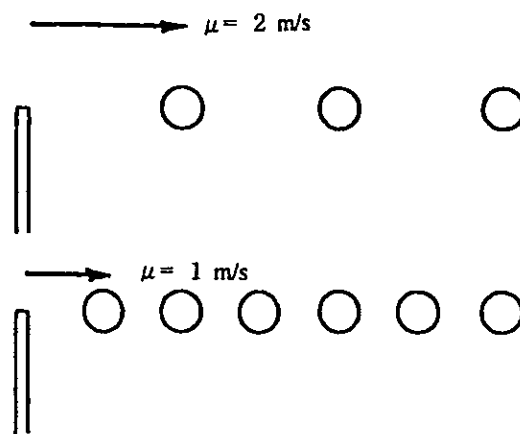


Fig. 10.1.1 The dilution effect of wind velocity

Therefore, although this overlaps with some of Chapter 4, we consider in this chapter the wind and the structure of the surface layer of the atmosphere. The wind blows around, over or through trees, forests, mountains and clusters of high-rise buildings in cities. The irregularity of these ground-surface is called roughness. The roughness disturbs air flow and produces eddies and turbulence (Fig. 10.1.2). In the turbulent air flow, wind velocity and direction changes time to time. The flow where the direction of the wind and its velocity change randomly is called the turbulent flow. The wind in the atmosphere is the most typical turbulent flow. The turbulence of the wind created by the roughness of the ground is the strongest near the ground surface and decreases as it rises in the sky. The atmospheric layer where the airflow is influenced by the ground surface is called the atmospheric boundary layer. The wind velocity and the turbulence changes with the altitude in the atmospheric boundary layer. In other words, the wind velocity is weak near the ground surface due to the drag force of the ground, but it increases at higher altitudes.



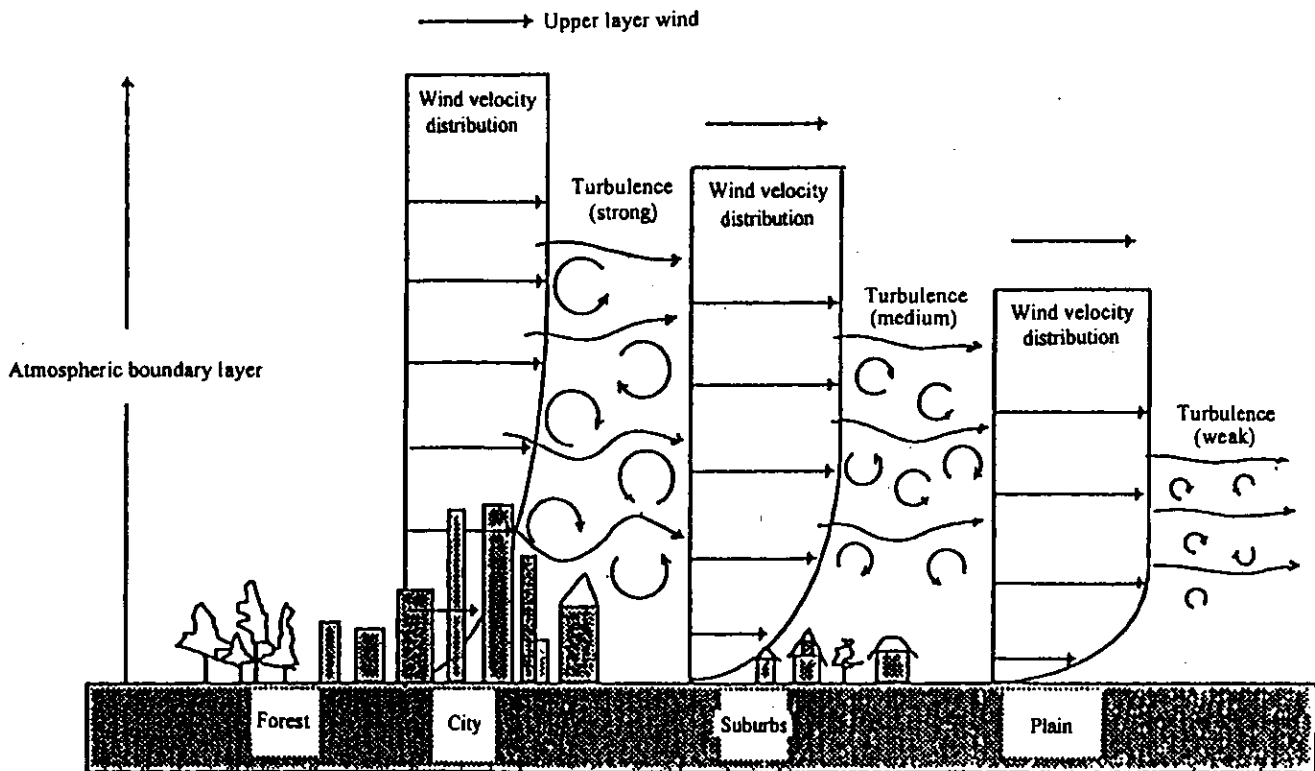


Fig. 10.1.2 Generation of turbulence in the atmosphere by the ground surface incongruity and the atmospheric boundary layer

Differences in temperature exist at ground level such as the temperature of the surface of the water and the ground in addition to the roughness. Also, convection occurs because solar radiation can warm the ground surface during the day in fine weather and the temperature of the air near the ground level rises. This convection motion is one of the biggest causes of turbulence in the wind. The atmospheric boundary layer where the convection is active in the daytime is called the mixing layer. On the other hand, convection is weak when it is cloudy, but the atmospheric boundary layer where the effect on wind velocity is dominant is called a neutral boundary layer.

On a night of fine weather, heat escapes from the ground surface into the sky through infrared radiation. Thus, the temperature of the ground surface falls and the air near the ground is also chilled. The vertical temperature of the atmosphere falls rapidly near the ground level. This air layer is called the ground inversion layer. The turbulence of the wind decreases in the inversion layer. The concept of the atmospheric boundary layer with the mixing layer, the neutral boundary layer and the ground inversion layer is shown in Fig. 10.1.3.

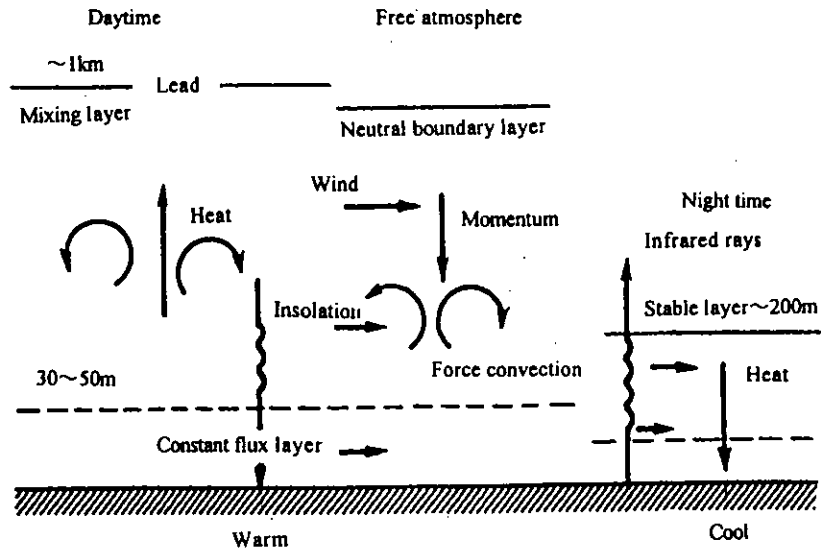


Fig.10.1.3 Origin and the classification of the atmospheric boundary layer

Generally, the temperature decreases up wards in the atmosphere. The temperature lapse rate is  $0.98^{\circ}\text{C}$  for every 100 m when the atmosphere is of dry air and the movement of the air mass is supposed to be adiabatic. This is called a dry adiabatic lapse rate and is shown by the symbol  $\gamma_d$ . When the rate of temperature decrease is larger than  $\gamma_d$ , then the vertical motion of the air mass accelerated by the buoyancy effect, it is said that the atmosphere is in an unstable condition (Fig. 10.1.4). On the other hand, when the lapse rate is smaller than  $\gamma_d$ , the vertical motion of the air mass is suppressed by the buoyancy force and turbulence become small. It is said that the atmosphere is in a stable condition. The layer of the atmosphere where the temperature increases up wards, is called temperature inversion layer. The inversion layer is a very strong stable condition and it emerges on a night of fine weather when the wind is weak. The relationship between the vertical temperature profile and the thermal stability of the atmosphere is shown in Fig.10.1.4.

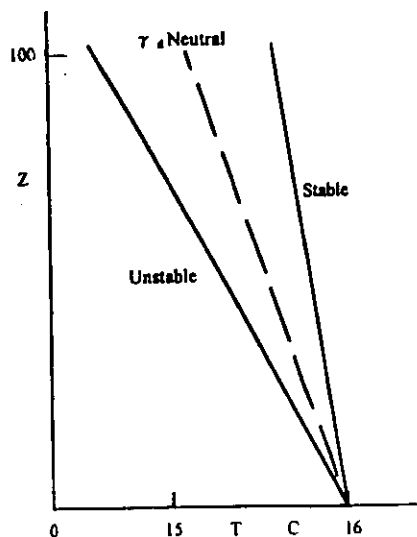


Fig.10.1.4 The atmosphere temperature inclination and atmospheric stability

Next, we will consider the relationship between turbulence and the diffusion by the wind. As for the wind, velocity and direction change every moment. The wind's vector deviates from the average to average. The deviation velocity from the average is called turbulent velocity. The turbulence component is deviled into the horizontal wind component and the vertical wind component. The root mean square of the turbulence component is called turbulence intensity. The relationship between the change of the wind and the diffusion is easily understand by considering movement of a particle in the turbulent flow (Fig.10.1.5). The particles in the smoke, drift up and down, left and right. If the turbulence of the wind is strong, the diffusion of the smoke is also big.

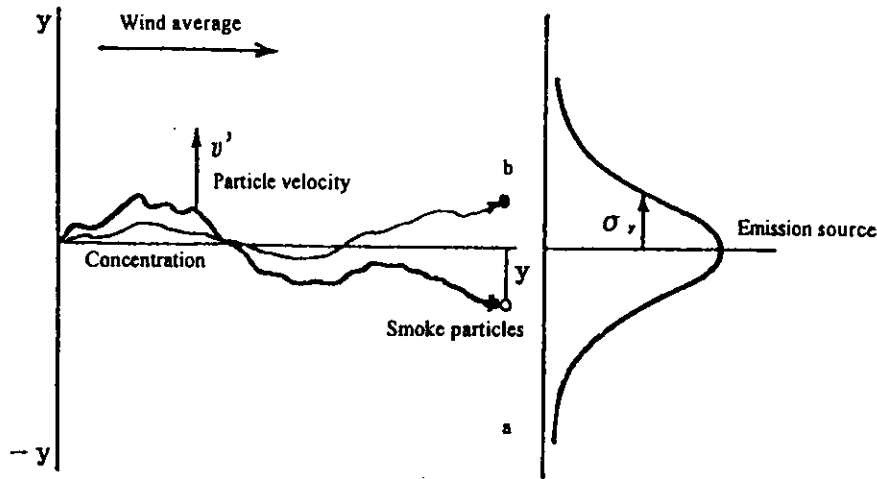


Fig.10.1.5 The diffusion of particles and the concentration profile

The idea of diffusion easily understand by releasing and tracking many particles as can be seen in Fig.10.1.5. The weight and size of the particulate in the smoke can be disregarded. When looking at the expanse of the particulate group on the downwind side, the number diminishes as most of the particles leave the flow as the average direction of the wind becomes prolonged. This shows the particulate concentration. The concentration profile of the smoke is the biggest in the center and this distribution profile is called a logarithmic profile or a Gaussian profile. The standard deviation of this distribution function is called the plume width of the smoke (Fig.10.1.5). This is illustrated in Eq. (1) when showing a concentration profile mathematically.

$$C(y) = C_0 \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \dots\dots\dots (1)$$

Here,  $\sigma_y$  is the direction of the plume width. Also,  $C_0$  shows the concentration at the plume axis. The turbulent flow in which the turbulence characteristic is independent to direction and place is called an isotropic turbulence. In the isotropic turbulence, plume width  $\sigma_y$  is given by Eq. (2) with  $\sigma_v$  the size of the turbulent flow.

$$\sigma_y = \left\{ 2\sigma_v^2 \int_0^T (T - \xi) R(\xi) d\xi \right\}^{\frac{1}{2}} \dots\dots\dots (2)$$

Here,  $T$  is the floating time of the smoke in the airflow and  $R(\xi)$ , the statistical characteristics of the turbulent flow, called auto-correlation function.  $R(\xi)$  is 1 when  $\xi$  is 0, and, as  $\xi$  increases  $R(\xi)$  approaches 0. The plume width is changeable by stability of the atmosphere, but this is described in the following paragraph.

The diffusion coefficient is another way of describing diffusion. For example, it is possible to show the distribution of the concentration as in Fig.10.1.6 with  $z$  at some point, flux  $F(z)$  by the diffusion of the smoke in the direction as shown in Eq. (3).

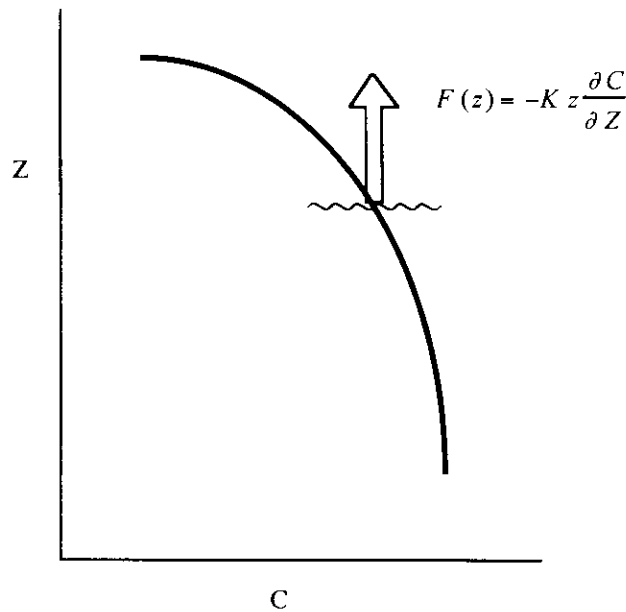


Fig.10.1.6 The relationship between the flux material  $Fz$  and the concentration gradient

$$F(z) = -Kz \frac{\partial C}{\partial z} \dots\dots\dots (3)$$

The diffusion coefficient  $Kz$  changes by the stability of the atmosphere.

The above is an analytical solution for diffusion and there is a theoretical method to treat diffusion by using the diffusion coefficient.

## 10.2 Outline of the Model to Estimate Atmospheric Diffusion

Up to the present, numerical solution solving the differential equation of diffusion and the plume and the puff model, both offering an analytical solution are used to estimate the atmosphere diffusion. Furthermore, the particle method in which many particles released and traced in the compute to estimate diffusion is recently developed the computer calculates the value.

### 10.2.1 Plume and Puff Models

The plume model supposes that the concentration of the smoke diffusion takes a logarithmic profile during a windy condition, and the concentration is calculated by giving plume width. In the atmosphere, the smoke diffuses up and down, left and right floated by wind. Assuming that the distribution of the concentration both in the vertical and horizontal directions takes a logarithmic profile, then the concentration of the smoke can be shown by the Eq. (4).

$$C(x, y, z) = \frac{Q}{2\pi\sigma_y\sigma_zU} \exp\left\{-\frac{y^2}{2\sigma_y^2}\right\} \left[ \exp\left\{-\frac{(z+He)^2}{2\sigma_z^2}\right\} + \exp\left\{-\frac{(z-He)^2}{2\sigma_z^2}\right\} \right] \dots\dots (4)$$

The symbols for this formula are;

$C(x, y, z)$  : The concentration at the point of downwind of emission source.  $x, y, z$

$\sigma_y, \sigma_z$  : Plume width in  $y$  and  $z$  direction respectively

$Q$  : The emission rate of pollutants per unit of time ( $m^3/s$ )

$U$  : Wind velocity ( $m/s$ )

$He$  : The height of the plume axis above ground and is called the effective source height ( $m$ ).

Because  $z$  is 0 when considering the concentration at the ground surfaces, the plume formula is simplified as shown in Eq. (5).

$$C(x, y, z) = \frac{Q}{\pi\sigma_y\sigma_zU} \exp\left\{-\frac{y^2}{2\sigma_y^2}\right\} \exp\left\{-\frac{He^2}{2\sigma_z^2}\right\} \dots\dots\dots (5)$$

Eq. (6) gives the ground surface concentration by the puff model which assumes a regular concentration profiles describing diffusion in a calm condition. The puff model is temporarily discontinuous model, assuming smoke is released serially for same time duration.

$$C(x, y, z) = \frac{q}{\pi^{\frac{3}{2}}\sigma_y\sigma_z} \exp\left\{-\frac{(x+y)^2}{2\sigma_y^2}\right\} \exp\left\{-\frac{He^2}{2\sigma_z^2}\right\} \dots\dots\dots (6)$$

By giving the plume widths  $\sigma_y$  and  $\sigma_z$  in the above plume model, the concentration profile in the horizontal

direction can be computed. However, in estimating the pollution of an industrial area with many the emission sources, a simplified diffusion model is used because the long term average for an annual average is the object of the estimation. For an estimate of a long-term average concentration, the wind is divided into 16 directions, and it is assumed that the concentration perpendicular to the wind is uniform in each wind sector. This equal to replace  $\sigma_y$  to  $\pi x/8$  and is equal to integrate the concentration profile with  $y$ .

The diffusion formula for this can be written as in Eq. (7).

$$C(x,0) = \left(\frac{1}{\pi}\right)^{\frac{1}{2}} \frac{Q}{u \pi x \sigma_z} \exp\left(-\frac{H e^2}{2 \sigma_z^2}\right) \dots\dots\dots (7)$$

For the puff model, it gives the concentration by a single puff, therefore, it is necessary to use the puff formula to sum up all of the puffs which are floating in space from the continuing source at one time. The plume widths  $\sigma_y$  and  $\sigma_z$  are given in floating time  $t$  and represented by Eq.(8), and integrating all of the puffs which were discharged by that time.

$$\begin{aligned} \sigma_y &= \alpha t \dots\dots\dots (8) \\ \sigma_z &= \beta t \end{aligned}$$

When integrating with the time, the ground surface concentration is given by Eq. (9).

$$C(R,0) = \frac{2Qp}{(2\pi)^{\frac{3}{2}}} \left\{ \frac{1}{R^2 + \alpha^2 H e^2 / \gamma^2} \right\} \dots\dots\dots (9)$$

The symbols in Eq. (9) are as follows:

- $R$  : The distance from the point source to the computation point  $(x^2 + y^2)^{\frac{1}{2}}$
- $Qp$  : Amount of pollutant in a puff for every time unit
- $\alpha$  : The rate of increase of the time of the horizontal plume width
- $\gamma$  : The rate of increase of the time of the vertical plume width

### 10.2.2 The Numerical Simulation Model

The numerical simulation model for diffusion is used to the diffusion over complex terrain and the diffusion of the temporarily changing location of air currents. It is inadequate to use the usual plume model for logarithmic profile for diffusion over complex terrain and diffusion of unsteady state condition.

The numerical simulation model calculates the airflow first, using the Navier Stokes equation for the turbulent flow. The next step is to calculate the diffusion in the flow field.

The basic form of the Navier Stokes equation is shown in Eq. (10).

$$\rho \left\{ \frac{\partial u}{\partial t} + u * \nabla u \right\} = - \nabla p + \rho g + \mu \nabla^2 u \quad \dots\dots\dots (10)$$

The formula of mass conservation is given by Eq. (11).

$$\nabla u = 0 \quad \dots\dots\dots (11)$$

Here, symbols are as follows:

$$\nabla : \frac{\partial}{\partial x} + \frac{\partial}{\partial y} + \frac{\partial}{\partial z}$$

$u$  : Vector flow velocity

$\rho$  : Fluid density

$g$  : Gravitational force

$\mu$  : Molecule viscosity coefficient

The air flow is normally a turbulent flow. Thus, the actual place of the flow is obtained by an equation rewritten from the Navier Stokes equation to meet the place of the turbulent flow.

Numerical simulation models of the turbulent flow are the model in which the turbulence structure is processed statistically. They are the k-ε model, the closure model which considers a higher turbulent structure, the Large-eddy simulation model which reproduces turbulence directly.

These models are used for the computation of the air flow over complex terrain, the simulation the of land and sea breezes, etc. and give the special structure of the air flow.

There are several numerical model for the simulation of diffusion such as solving the differential equation of diffusion by finite difference method, or the particle method in which particles are released and the transport and diffusion of them are traced.

The differential equation of discussion (the Fick equation) of diffusion in the turbulent boundary layer is given by Eq. (12)

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y} + w \frac{\partial C}{\partial z} = kx \frac{\partial^2 C}{\partial x^2} + ky \frac{\partial^2 C}{\partial y^2} + \frac{\partial}{\partial z} \left( kz \frac{\partial C}{\partial z} \right) \quad \dots\dots\dots (12)$$

Here, the symbols are as follows:

$C$  : Concentration

$t$  : Time

$x, y, z$  : Coordinate axis of the perpendicular coordinate

$u, v, w$  : The components average flow in the directions of  $x, y, z$

$kx, ky, kz$ : The turbulent flow diffusion coefficient for the directions of  $x, y, z$

In the differential equation of discussion model of diffusion, the field of diffusion is divided into the meshes,

and the concentration, at the points of intersection or at the mid-point of the mesh points are calculated. An example of the mesh division is shown in Fig. 10.2.1.

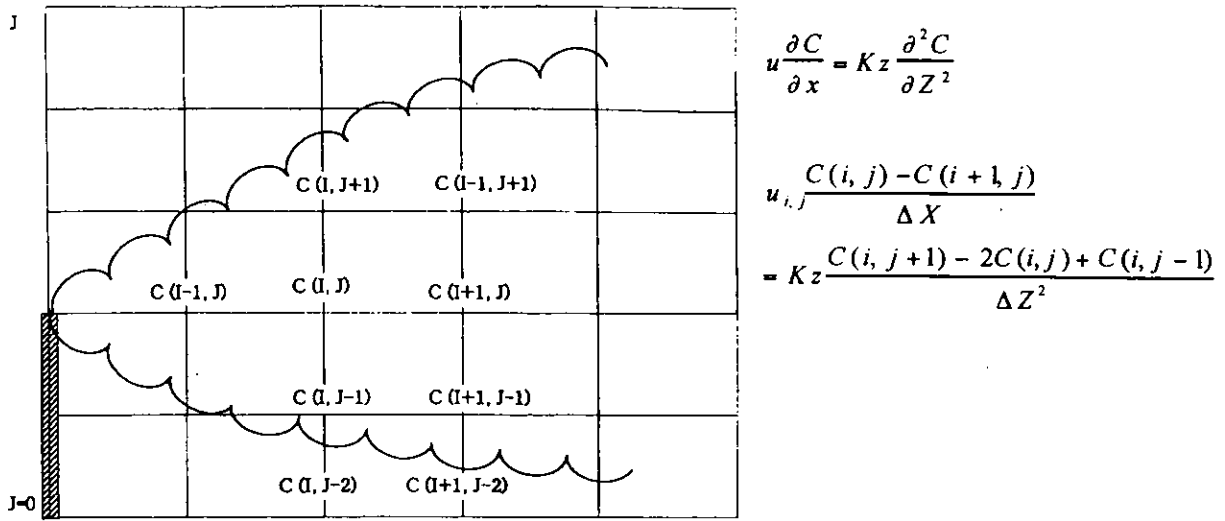


Fig. 10.2.1 An example of the Mesh division for the finite difference method

### 10.2.3 The particle method model of diffusion

Next, diffusion simulation by the particle method is introduced. The conditions can be set to meet the statistical nature of the turbulent flow in the particle method. In other words, in a differential equation of discussion, the condition of the diffusion is dependent on the diffusion coefficient. Generally, in atmospheric diffusion, near the emission source, the plume width is proportional to the distance from the emission source and, on the downwind side, it is known to be proportional to the square root of the distance. This shows that the diffusion coefficients are dependent on the distance from the source. However, it is difficult to change the diffusion coefficients by distance in the Fickian equation. For the particle method, on the other hand, it is possible to simulate actual turbulent diffusion by reproducing turbulent structure and releasing particles in the flow field. To get a smooth concentration profile when using the particle method, however, several hundreds of thousands of particles must be discharged, therefore it requires long computer time. Therefore, a simpler particle method is also proposed.

The velocity  $v(t)$  of a particle in the particle method is give in Eq. (13).

$$v(t) = v + v'(t)$$

$$v'(t) = R(\Delta t) v'(t - \Delta t) + n(1 - R(\Delta t)) \sigma v \dots\dots\dots (13)$$

Here,  $v$  is the average velocity,  $v'(t)$  is the turbulent component and  $n$  is a random number with an average value of 0 and a standard deviation of 1. In the particle method, the diffusion condition can easily be grasped visually by expressing the distribution of particles like a cloud.



#### 10.2.4 The model of the plume rise

Smoke which is discharged into the air, rises up the atmosphere because of its own inertial force and buoyancy. In the diffusion calculation of the actual smoke, the distance of the plume rise is calculated and it is assumed that the smoke is horizontally discharged from the maximum height that smoke reaches (effective source height  $H_e$ ). Many models are proposed as a method to estimate the plume rise. Introduced here are the Briggs and CONCAWE (Conservation of Clean Air and Water, Western Europe) models which have been adopted by Japan in the manuals for the total emission control method.

The CONCAWE model gives the distance  $\Delta H$  that the plume rises through Eq. (14)

$$\Delta H = 0.157 \frac{Q_H^{1/2}}{u^{3/4}} \dots\dots\dots (14)$$

Here, each symbol is represented as follows:

- $\Delta H$  : Distance of the plume rise (m)
- $Q_H$  : Thermal effluent (cal/s)
- $u$  : Wind velocity (m/s)

where,  $Q_H$  is shown in Eq. (15).

$$Q_H = \rho C_p Q \Delta t \dots\dots\dots (15)$$

- $\rho$  : Exhaust gas density at 0°C ( $1.298 \times 10^3$  g/m<sup>3</sup>)
- $C_p$  : The specific heat at constant pressure (0.24 cal/kg/K)
- $Q$  : Exhaust gas quantity for unit time (m<sup>3</sup>N/S)
- $\Delta t$  : The difference between the exhaust gas temperature (T<sub>g</sub>) and the atmospheric temperature (T<sub>g</sub>-15°C)

The Briggs model gives the plume rise under a calm condition with no wind in Eq. (16).

$$\Delta H = 1.4 Q_H^{1/4} \left( \frac{d\theta}{dz} \right)^{-3/8} \dots\dots\dots (16)$$

Here,  $\frac{d\theta}{dz}$  is the heat gradient (C/m).

Also, the effective source height is a summation between the stack height and the plume rise.

## 10.3 Investigation of atmospheric diffusion

New factories are being built the environmental impact on the atmosphere of each factory needs to be evaluated accurately. Or, the atmospheric environment needs to be improved in industrial areas where many factories are located. To satisfy these requirements field research must be done on the environment and diffusion in these areas. A field study is necessary, especially, when the object area lies over complex terrains, or weather characteristics which are disadvantageous to preserving the atmospheric environment.

Research is divided into a meteorological condition research to grasp the situation of the field of diffusion and an environmental investigation which examines the environmental concentration and condition of the diffusion.

### 10.3.1 Meteorological conditions investigation

Investigation of the meteorological condition of an object area acquires not only the data used to diffusion simulation, but also the general characteristics of the atmospheric on the diffusion that in the area. The items for field for diffusion measurement are as follows:

- ① The hourly wind direction and velocity at ground level
- ② The hourly solar radiation and net radiation
- ③ The temperature and the vertical distribution of the temperature
- ④ The atmospheric turbulence
- ⑤ The wind's direction and velocity vertical distribution and velocity

The wind direction and velocity at ground level are measured by either a windmill type anemometer or a ultra-sonic anemometer. It is desirable to have a density of one observation site with a 5-kilometer radius, or at least several sites in the whole area. The height of the measurement equipment should be more than 10 meters above the ground and in a location where there is no influence from buildings and trees, etc. An ultra-sonic anemometer set on a pole is shown in photo.10.3.1.

The solar radiation and the net radiation are necessary to estimate the degree of atmospheric stability and the development of the mixing layer or the ground inversion layer. The equipment is installed in an open place with no shade and, if possible, on grassland. A net radiometer is shown in Photo.10.3.2.

Temperature and vertical temperature distribution near the surface of the ground are necessary, to make clear the conditions of the mixing layer and the inversion layer as well as the meteorological conditions and to use diffusion model. The ground level temperature is measured with a thermometer housed in an instrument shelter installed on the grassland. To measure a continuous vertical temperature distribution, a thermometer is installed in a building such as a tower or a chimney. The thermometer used is usually a ventilated platinum wire thermometer. In areas where towers or chimney cannot be used to measure the vertical temperature distribution continuously, a remote measurement method such as a radio sonde can be used (Photo.10.3.3).

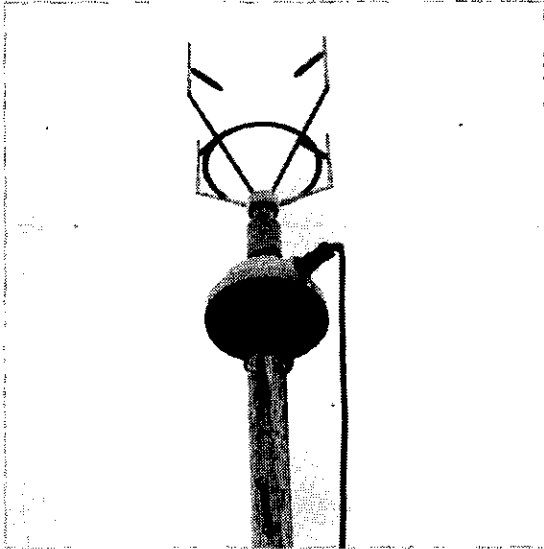


Photo.10.3.1 An ultra-sonic anemometer

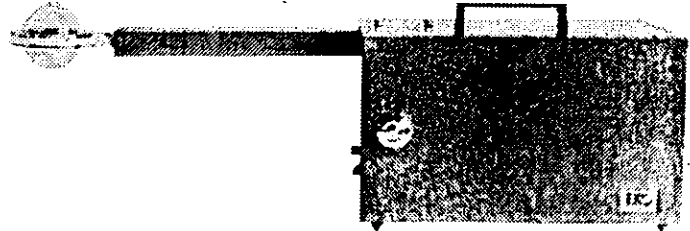


Photo.10.3.2 A net radiometer

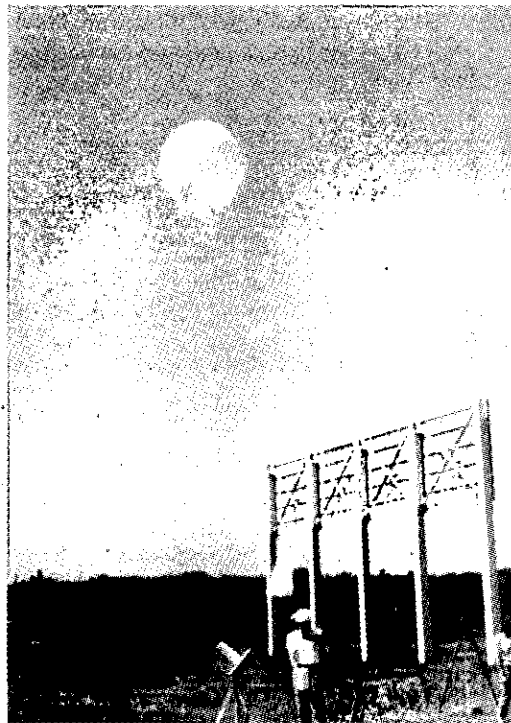


Photo.10.3.3 A radio sonde

Turbulence in the atmosphere is necessary to introduce the influences on diffusion by the terrain or structures (This word covers buildings, towers and other man-made artifacts. The word "constructions" doesn't usually pertain to buildings). An ultra-sonic anemometer is installed on a tower 20 meters or higher off the ground and it measures the two components of turbulence of horizontal direction and vertical direction.

The vertical distribution of the wind direction and velocity is necessary information to deal with diffusion from

high sources such as tall stacks. Wind direction/velocity meters and an ultra-sonic anemometers are mounted on high towers and chimney to continuously measure the vertical distribution of the wind direction and velocity. Where towers and chimney can not be used, remote methods can be utilized such as the pilot balloon method which tracks balloons released by theodolites and, also, the Rawin sonde can be used.

### 10.3.2 Investigation of the diffusion condition

Research into the diffusion condition of pollutants discharged from chimney among others is expensive and needs much manpower, but they are very important. The first is to measure the diffusion distribution of pollutants from existing industrial areas and the areas where the emission sources already exist. A tracer gas diffusion experiment is conducted to make clear the influence from specific sources in an area where the emission source is not operating yet and an area where many emission sources exist. In the tracer gas experiment, a material which has a low background concentration and highly sensitive to analysis is released instead of pollutants and is collected on the downwind side for analysis in a sampling network which has been arranged in an arc. As tracer material, sulfur hexa-fluoride ( $\text{SF}_6$ ) and fluorocarbon 13B1 ( $\text{CBrF}_3$ ), and so on, were used from the past but these chemicals are the greenhouse gases and their use is becoming impossible.

Fig.10.3.1 is an example of a diffusion concentration profile on the ground which was acquired by a tracer gas experiment using sulfur hexa-fluoride. The release height was 147 meters, and the emission duration and sampling time were both about one hour. The tracer material was collected by the sampler which was arranged in a half arc and analyzed, and an contour distribution of concentration was made. By these data the maximum concentration, the distance from the source point, the plume width and so on are acquired, and they are related to the meteorological condition and analyzed.

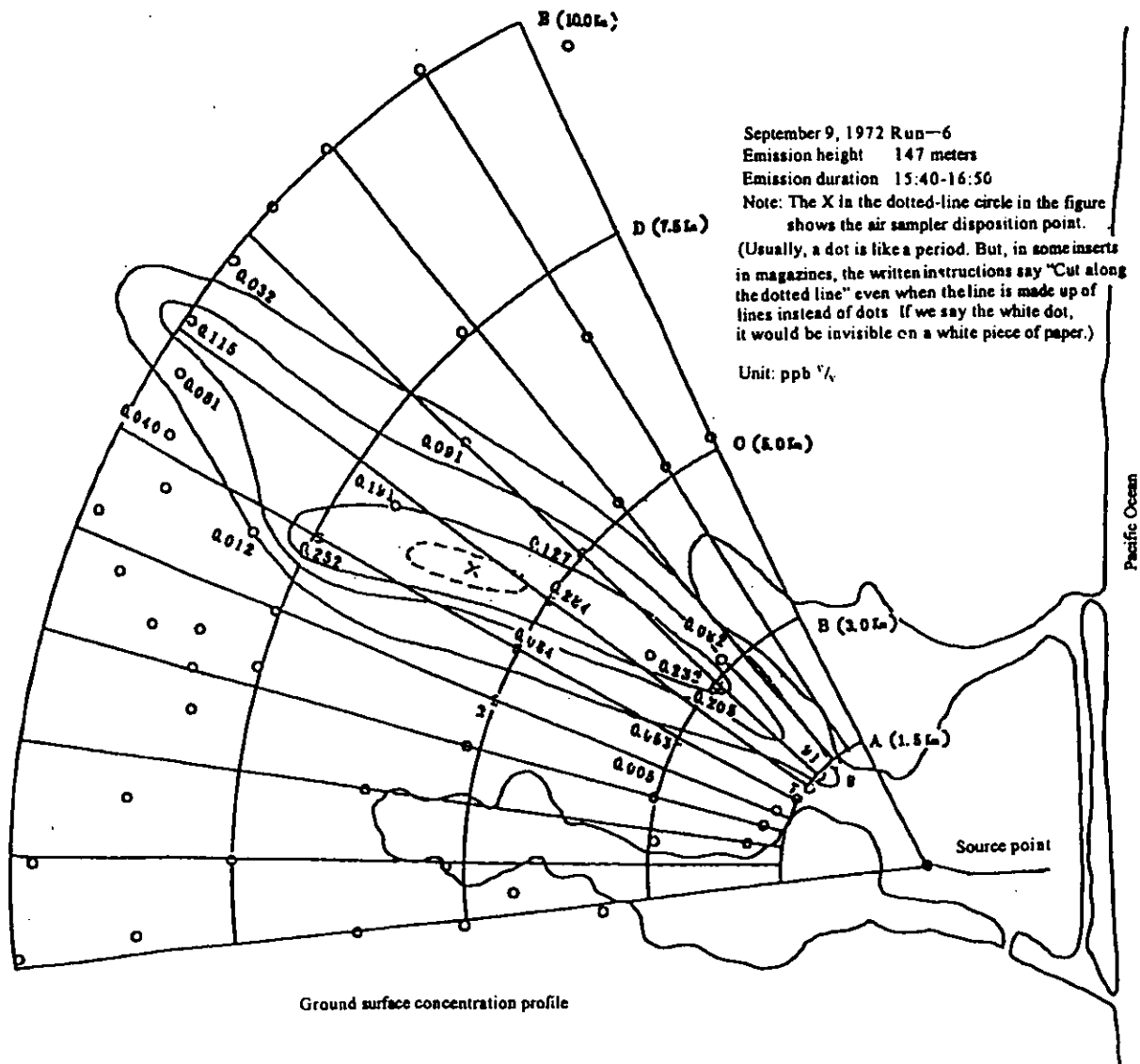


Fig.10.3.1 Ground surface concentration profile acquired by a tracer diffusion experiment

## 10.4 The Practical Method of Estimating Air Pollution

How is the diffusion simulation model which was described above used in the estimation of actual atmospheric environment? Also, what kind of data are gathered, analyzed and used? Here, an example is given of the forecasting model with an actual atmospheric environment concentration.

### 10.4.1 Estimating the long-term concentration

The type of concentration that can be most accurately diffusion by the present air pollution forecasting model is the long-term average such as the annual average or the seasonal average concentration. The primary reason for this is, the fact that the diffusion phenomenon is basically a statistical phenomenon and the ability to estimate the concentration at a point and time is only statistical. However, the long-range averages like the annual average or the seasonal average are made up of the average of the collections of many one-hour averages. Thus, the statistical uncertainty decreases. Other error factors are the errors in the field of diffusion and estimation errors at the emission source.

At present, the long-term average density estimate which uses analytical solution models such as the plume and puff models is regularly used for the above reason and is used to estimate the concentration of sulfur oxides and nitrogen oxides. The emission source, the weather and the environment concentration data are all necessary to estimate the long-range average concentration. The flow chart of the simulation of long-term average for the total emission control method of sulfur oxides and nitrogen oxides in Japan is conducted with the procedures in Fig. 10.4.1.

For example, from the data on the solar radiation and the wind direction and velocity for every hour of 365 days or one year, the occurrence frequency of the meteorological condition is calculated for the stability of the atmosphere and for the class of wind velocity of each of the sixteen directions.

The sorting of the wind velocity class and the stability of the atmosphere is due to the Japan standard stability classification as shown in Table 10.4.1. This classification list is an improved version of the one which was proposed by Pasquill of the UK. Pasquill's method displayed the strength of the solar radiation with the cloud volume, but, in the Japan standard stability classification, Pasquill's method has been changed into solar radiation which is a quantitative indicator. This method evaluates the convection and the strength of the inversion layer by the surface wind velocity and the solar radiation or the net radiation, and classifies stability. As for stability, A-C are unstable, D is neutral and E and F are stable. Pasquill and Gifford gave plume width to each stability as seen in the graphs in Fig. 10.4.2.

The annual average concentration uses the data of pollutant emission which was estimated according to the time zone and season, and is computed using the wind direction, the wind velocity and the occurrence frequency of thermal stability of the atmosphere which were ordered according to the time zone and season. To estimate the pollutant emission in this case, the emission coefficient which was introduced in Chapter 6 section 8 is utilized.

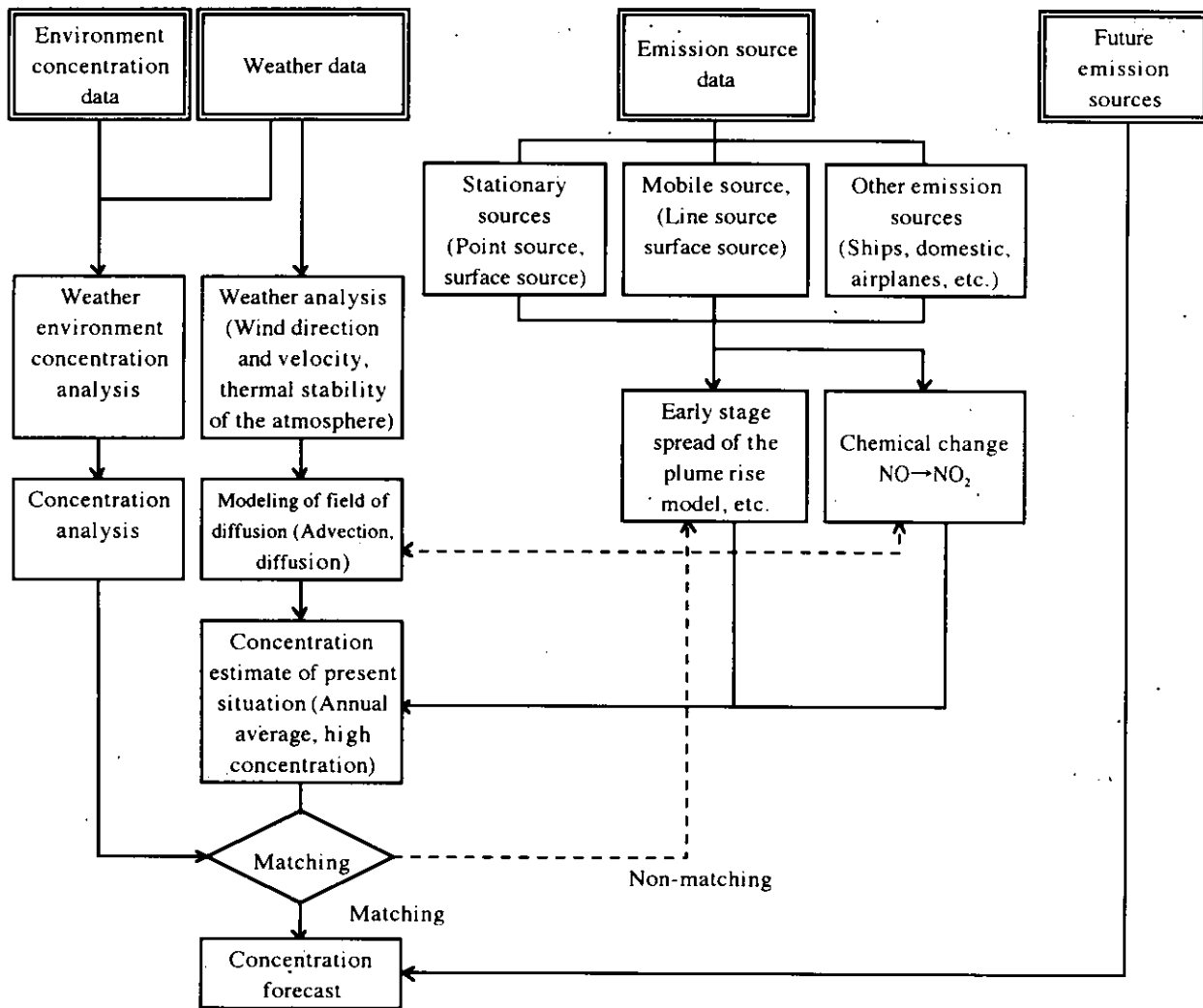


Fig. 10.4.1 The estimated flow of the long-term average concentration

Table 10.4.1 The Japan standard stability classification

Ground level Wind velocity (m/sec)	Daytime Solar radiation amount cal/cm <sup>2</sup> /hr			Thick Cloud (8-10) Thick night cloud	Nighttime	
	>50	49~25	<24		Upper layer cloud (5-10) Middle, low layer cloud (5-7)	Cloud volume (0-4)
<2	A	A~B	B	D	(F)	(G)
2~3	A~B	B	C	D	E	F
3~4	B	B~C	C	D	D	E
4~6	C	C~D	D	D	D	D
>6	C	D	D	D	D	D

Note: The numerical value considered Japan's climate for the solar radiation ranking of Pasquill's stability classification table.

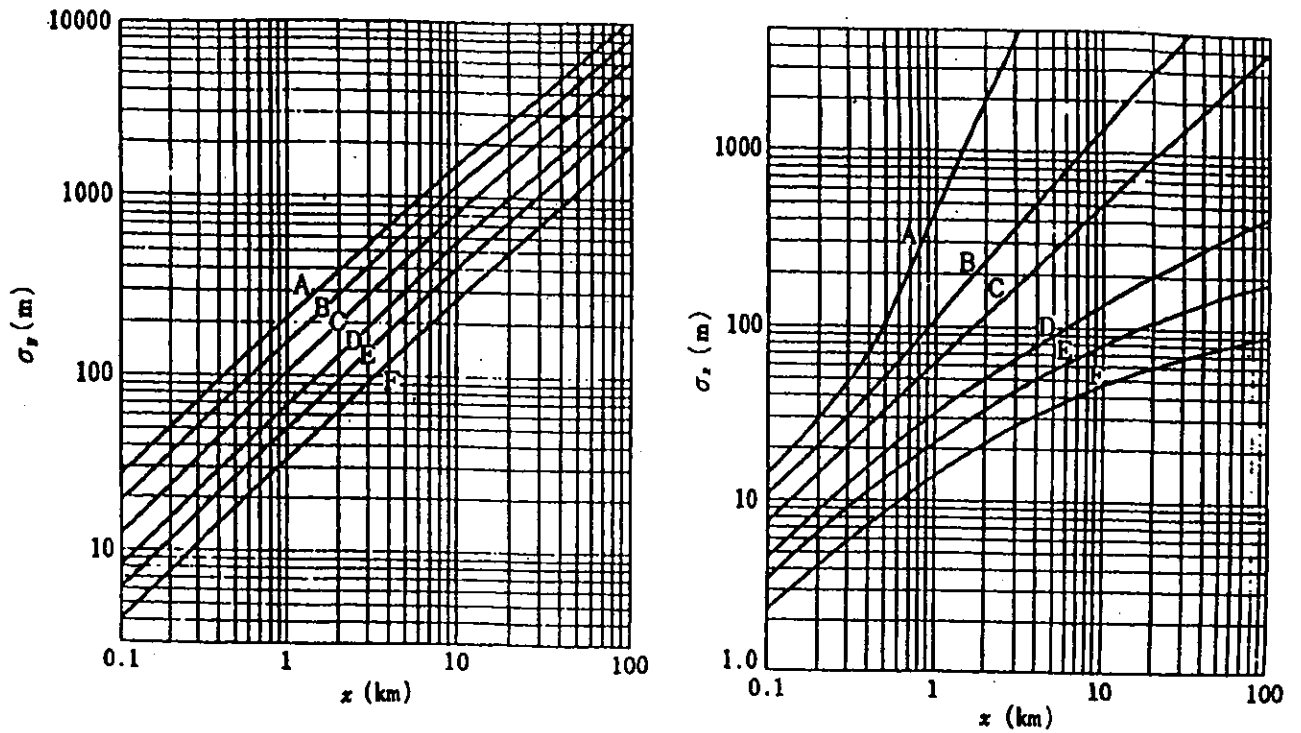


Fig. 10.4.2 Plume width for the atmospheric stability by Pasquill's method

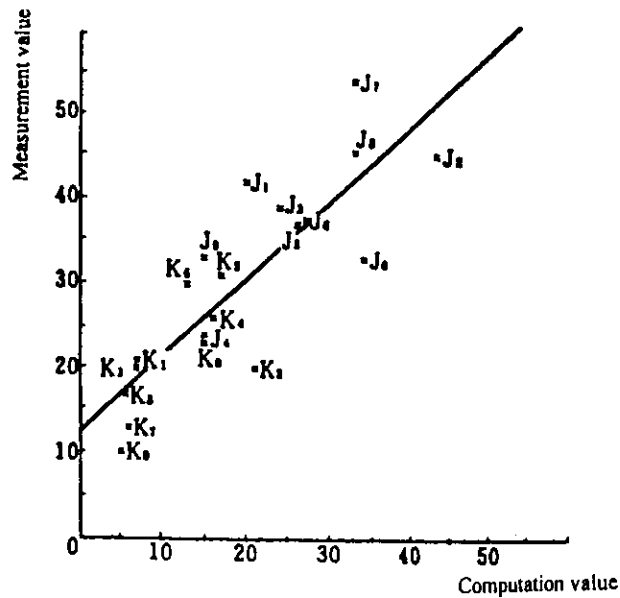


Fig. 10.4.3 Scatter diagram showing the computation value and the measurement value of  $\text{SO}_2$  annual average from the puff and plume models

Fig. 10.4.3 scatter diagram comparing the measurement value (horizontal) and the calculated value (vertical) computed by using the above method of simulation. If the point is located on the 45 degree line, the calculation value and the measurement value agree totally. However, the computational value becomes lower than the measurement value because there are unknown sources which is not able to cover in the simulation, and, also, because



the pollutant is transported from an outside area. This difference is called background concentration. If the computational value is  $x$ , the measurement value is  $y$ , and the background is  $b$  then the relationship between the measured and the computational value is approximated using Eq. (17).

$$y = ax + b \quad \dots\dots\dots (17)$$

Here,  $a$  is a proportional coefficient with the measurement value and the computation value.

The agreement of the simulation is evaluated by the correlation coefficient with the measurement value and the computation value along with coefficient  $a$ . The simulation becomes highly precise when the correlation coefficient is near 1 and  $a$  is near 1 in the calculation. A case of a non-heating period is shown in Fig. 10.4.4 as an example of the diffusion calculation results with an seasonal average concentration in the area.

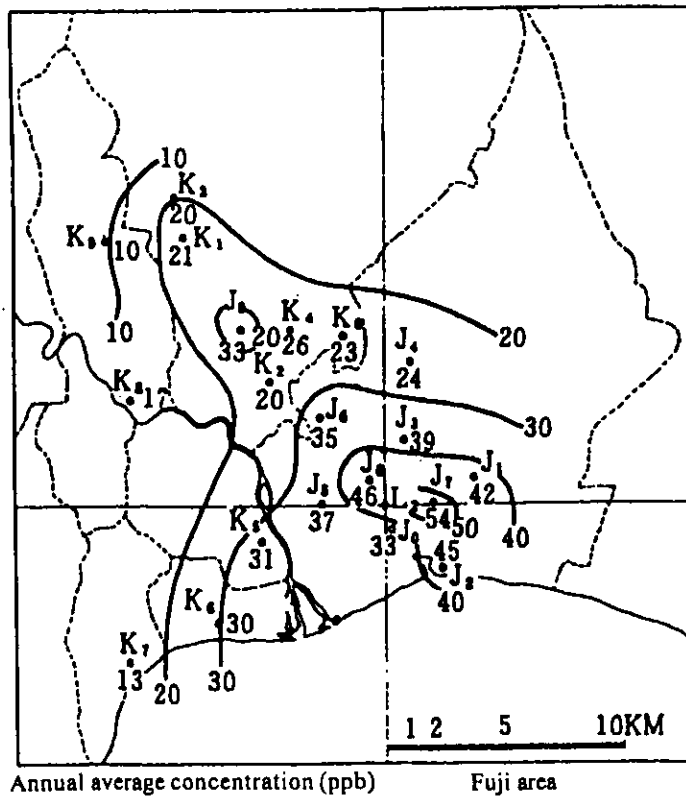


Fig.10.4.4 A long-term average concentration profile from the puff and plume models

A personal computer program, which estimates the long-term average concentration of the plume and puff models is sold. A program which statistically analyzes the meteorological condition from the weather data such as the wind direction, wind velocity, and solar radiation is also available.

#### 10.4.2 Estimating the short-term average concentration

The numerical simulation models, described in section 2, are used for the estimation of the concentration under specific meteorological conditions and of the diffusion under complex topography along with the plume and puff models. A numerical simulation model is used often for the diffusion calculation for sea-land breeze condition. Shown in Fig. 10.4.5 is a computation example of a simulation of the sea-land breeze on the Kanto Plain using the numerical simulation model. This displays the vector of the wind direction and the wind velocity near ground level. This numerical model is considering the latent heat flux and a sensible heat flux produced between the ground surface and the atmosphere by the solar radiation.

It becomes possible with the plume and puff models to estimate the short-term diffusion concentration in an actual situation by introducing the wind direction and velocity and, also, the change in the height of the plume width, etc. In addition, the box model, the limited length plume model and the floating puff model are used for diffusion calculation when there is a change in the field of diffusion during the time. Shown in Fig. 10.4.6 is the concept figure of a floating puff model and a limited length plume model.

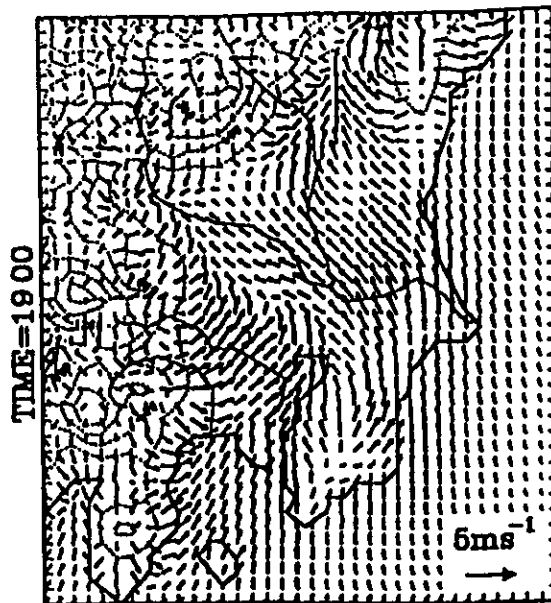


Fig.10.4.5 Distribution of surface winds on the Kanto Plain in the case of sea-land breeze from a numerical simulation model

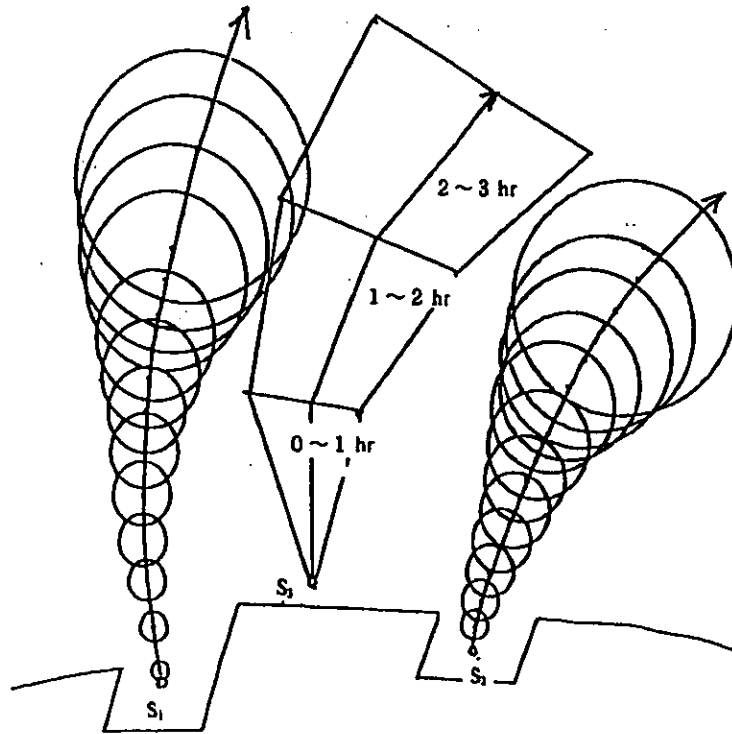


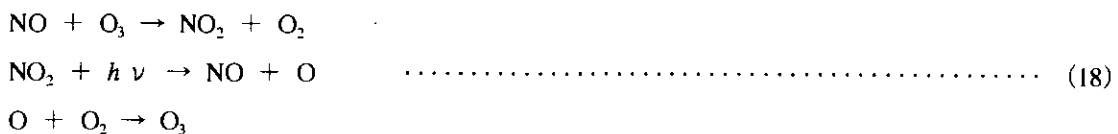
Fig. 10.4.6 The concept figure of a floating puff model and a limited-length plume model

### 10.4.3 Estimating chemically reactive substances

The environmental concentration simulation of the material which reacts chemically in the atmosphere, the process of the chemical change should be simulated along with the diffusion process. The photochemical reaction is involved in the chemical change in most cases. The representative reactions are oxidants generated through photochemical reactions, nitrogen oxides formed into nitrogen dioxide, sulfates and acid substances formed from sulfur dioxide, particulate matter formed from nitrogen oxides and hydrocarbon species and destruction of the ozone in the stratosphere.

Several tens of chemical substances and twice or more that of the reaction formulas are considered in the simulation of photochemical oxidants. The representative chemical substances are non-methane hydrocarbon, nitrogen oxides etc. The non-methane hydrocarbon is classified into ethylene, aromatic and aliphatic hydrocarbons, excluding ethylene and a carbonyl compound excluding formaldehyde.

Regarding the reaction of nitrogen oxides ( $\text{NO} \rightarrow \text{NO}_2$ ), in the case like the diffusion of a short distance, the reaction of  $\text{NO}$  and  $\text{O}_3$  is the quickest reaction and the photochemical reaction can be disregarded. Then,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{O}_3$ ,  $\text{O}_2$  and the intensity of ultraviolet radiation are applied. A reaction formula is shown in Eq. (18) but the details of the reaction have been described in Chapter 4 section 2.



For the total emission control for nitrogen oxides in Japan, the method (exponential function model) of Eq. (19) which simplified this reaction system is used.

$$\frac{\text{NO}_2}{\text{NO}_x} = 1 - \frac{\alpha}{1 + \beta} \{ \exp(-kt) + \beta \} \dots\dots\dots (19)$$

Here,  $\alpha$  is the rate of NO at the emission source,  $\beta$  is the numerical value related to the rate of NO when its reaction reaches a equilibrium, and  $k$  is a reactive constant. Also, the value of these coefficients are as follows:

$$k = 0.0062 \text{ UO}_{3B} \text{ (stationary sources, vessels)}$$

$$= 0.208 \text{ UO}_{3B} \text{ (automobiles, houses)}$$

$$\beta = 0.3$$

$$\alpha = 0.9$$

Here, U is velocity and  $\text{O}_{3B}$  is concentration of Ozone.

Other than the method above, there is another method using Eq. (20) (statistical model) in which the ratio is statistically processed from the observed data of nitrogen oxides.

$$\text{NO}_2 = a [ \text{NO}_x ]^b \dots\dots\dots (20)$$

Here, the coefficients a and b are determined through the analysis of observed data:

In the estimate of environment concentration, an exponential function model is used when the contribution of each stationary source are required and the statistical model is used when computing the influence of automobiles.

#### 10.4.4 Estimating from wind tunnel tests

Wind tunnel tests are utilized in conjunction with numerical models in the simulation of diffusion when the so-called down-washes and down-drafts, affect the diffusion due topography or buildings, for example, effects from the wake in the lee of buildings and smoke stacks. An example of wind tunnel equipment is shown in Fig.10.4.7.

### Atmospheric Air flow and Diffusion Research Wind Tunnel

- |                      |                            |                |
|----------------------|----------------------------|----------------|
| 1 Fan                | 6 screen                   | 11 Moving cart |
| 2 Mixing chamber     | 7 Test section             | 12 3rd corner  |
| 3 1st corner         | 8 Probe carriage           | 13 4th corner  |
| 4 2nd corner         | 9 floor lift               |                |
| 5 Air heater/chiller | 10 Heating/chilling system |                |

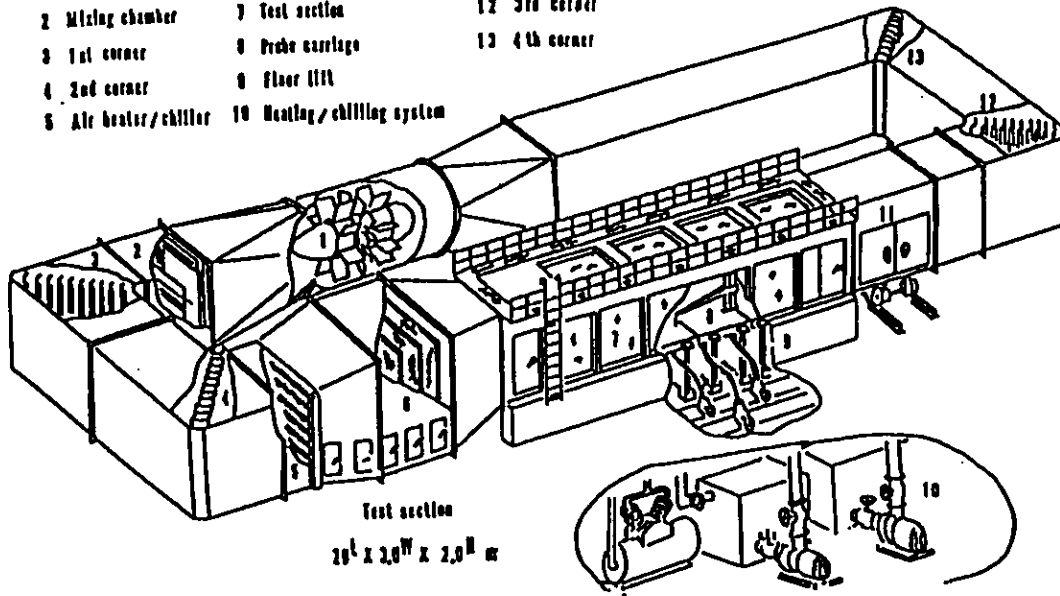


Fig.10.4.7 Example of wind tunnel equipment

The wind tunnel consists of a blower, a flow control section, a test section and a re-circulation section. In the diffusion wind tunnel test, instead of smoke, a tracer gas flows from the model stack over a model topography or building which was built in the test section, then tracer gas is sampled in the downstream side and analyzed. To make it similar to the turbulent boundary layer of the atmosphere, a turbulence generator and a roughness element are placed in the upstream side of the test section. To develop a sufficient turbulent flow boundary layer, an approach distance of at least 2 to 5 meters is necessary and a test section length of more than 5 meters is required. Scale model of 1/100 to 1/1,000 in the case of buildings and for topographic model the scale of 1 to several hundredths to 1 to several thousandths is used.

As a tracer gas for the diffusion experiment, a kind of hydrocarbon such as ethylene or propane is used. The wind velocity in the wind tunnel test is the same or a little less than the actual velocity. Therefore, the range of the wind velocity which is required for the diffusion wind tunnel is approximately 1 to 10 m/s. Photo.10.4.1 shows the diffusion of smoke in a wind tunnel test.

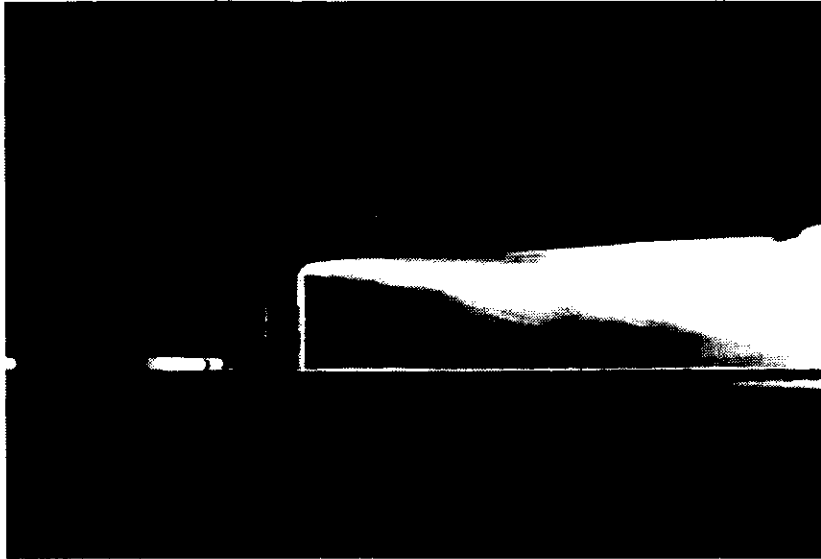


Photo.10.4.1 Smoke diffusion experiment with a diffusion wind tunnel

## 10.5 The Statistical Forecasting Model

A physical and chemical diffusion models are generally used for estimating air pollution. However, in the case of the pollution of which physical and scientific models have not established or when any input data for the physical pollution model can not be obtained or when it is only necessary to forecast the air pollution level for a few hours after then statistical methods to estimate the pollution using time series concentration data or the pollution and weather data are used.

The category classification method by weather charts is used for the forecasting of the air pollution level. This method estimates the pollution level by examining the concentration of the pollution for each pattern, classifying a pattern in the weather by season and the existence or non-existence of high atmospheric pressure, low atmospheric pressure, fronts and so on. This method is used to estimate the density level of photochemical oxidants and nitrogen oxides for the whole area. An example of a classification of a typical weather chart is shown in Table 10.5.1.

Table 10.5.1 An example of a weather chart classification for an air pollution estimate

Symbol	Explanation
PN	North high and south low type (Baiu type, Okhotsk high type, etc.)
PE	East high and west low type
PS	South high and north low type (Typical summer pressure type, the bright in May, etc.)
PW	West high, east low type (Strong northerly wind type in winter)
OW	Weak west high and east low type (Weak northerly wind type in winter)
H	Covered with a traveling high pressure. Covered with a long belt high.
F	Front (Influence from an active front).
T	Typhoon (Influence from a typhoon).
LF	Strong cyclone front
LC	Strong cyclone central area
LB	Strong cyclone rear area
LS	Strong cyclone warm area
OLF	Weak cyclone front
OLC	Weak cyclone central area
OLB	Weak cyclone rear area
OLS	Weak cyclone warm area
L <sub>2</sub>	Middle area of twin cyclone
OL <sub>2</sub>	Weak twin cyclone
MX	There are a low-atmospheric-pressure and a traveling high-atmospheric pressure which is strong within 500 km.
OX	Others

The auto-regression estimate method, which is called multiple regression analysis, a Karman filter or Wiener filter, is a way to make a quantitative estimate several hours after the concentration without using a physical diffusion model. The change of concentration is divided into the random component and the periodic component like the

transition of a day and a numerical value filter method is used for the random component.

In other words, the estimate object, which is the concentration  $C(t)$  at the time of  $t$  is divided into the diurnal change  $C_p(t)$  and an irregular component  $C'(t)$ , and past concentration data can be used for the day change. As for an irregular component, for example in the Wiener model,  $C_e(t)$ , which is the estimate model of  $C'(t)$  is estimated by the weight function  $w(\tau)$  as shown in Eq. (21).

$$C_e(t) = \int_0^{\infty} w(\tau) C'(t - \tau) d\tau \quad \dots \dots \dots (21)$$

Here,  $\tau$  is delay time and  $w(t)$  is sought in order to minimize the error of  $C'(t)$  and  $C_e(t)$ . Fig.10.5.1 shows an example of the estimate results of one hour ahead for  $SO_2$  using the Karman filter model.

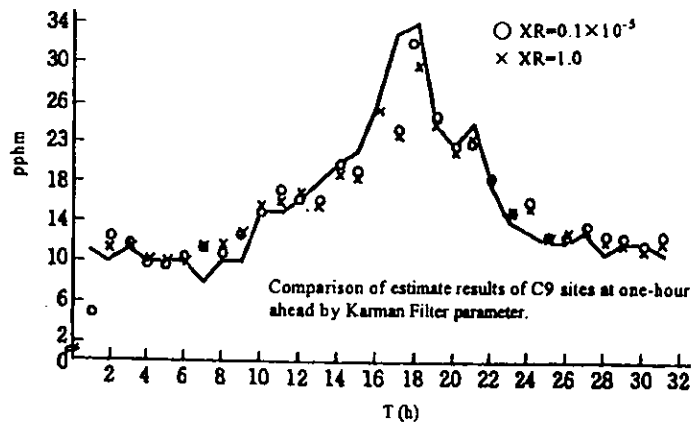


Fig. 10.5.1 An example of one-hour ahead estimate of  $SO_2$  by the Karman filter model



## 10.6 CMB Method

### 10.6.1 Overview

The supposition of simple mass conservation is adopted in the basic concept of the receptor model, which is used to determine the emission source of the suspended particulate matter in the atmosphere. That is, the emission source exists in the symbol  $p$  and supposes that there is not an interaction which would cause the removal and generation of a mass among the discharged particulate matter. Then, the material concentration  $C$  of the particle in the receptor (the measuring point) becomes the linear summation of  $S_j$  from each emission source as seen in Eq.(22).

$$C = \sum_{j=1}^p S_j \quad \dots\dots\dots (22)$$

In the same way, the mass concentration  $C_i$  of the component  $i$  of the particulate matter is as shown in Eq.(23).

$$C_i = \sum_{j=1}^p a_{ij} S_j \quad \dots\dots\dots (23)$$

Here,  $a_{ij}$  is the density containing component  $i$  in the particulate matter which comes from the emission source  $j$  to the receptor.

There are two receptor models, the CMB (Chemical Mass Balance) model which handles one piece of observation data and a multivariate model which handles numerous pieces of data. Outlined below is the typical CMB model method which is often used for the analysis of source contribution.

### 10.6.2 The CMB model

Fig. 10.6.1 shows the concentration range and geometric mean of various elements in the atmosphere gathered from the results of analyzing many particulate matter samples collected in city areas. Table 10.6.1 is an example of measurements of the component concentration of the particles discharged from the principal emission source in a city area. The basic concept of CMB is that the contribution concentration of the emission source is determined by obtaining much of the chemical components concentration from the samples which were gathered at the measurement station. Analysis is made with the observation on the characteristic features of a chemical component concentration pattern (emission source profile) of particles discharged from the emission source as shown in Table 10.6.1.

More than 20 kinds of chemical components are supposed to have been to be correctly grasped from the discharged particles of the principle emission source in the city area. Then, as for the concentration of the chemical component of the particulate matter which is observed in the environment, the concentration of the chemical components of these particles from the emission source superpose into a chemical component concentration pattern with the weight percentage of  $S_j$ . The weight percentage is  $S_j$  which is the contributed concentration of the particles discharged from the emission source. When the measured number  $n$  of the chemical components is equal to or

higher than the number of the emission sources, theoretically, by solving the simultaneous equation in Eq. (23), the mass contributed concentration of each emission source can be obtained. The tracer element method, linear programming method, the ordinary linear least-squares method, the effective variance least-squares method and so on, are used as the actual calculation techniques.

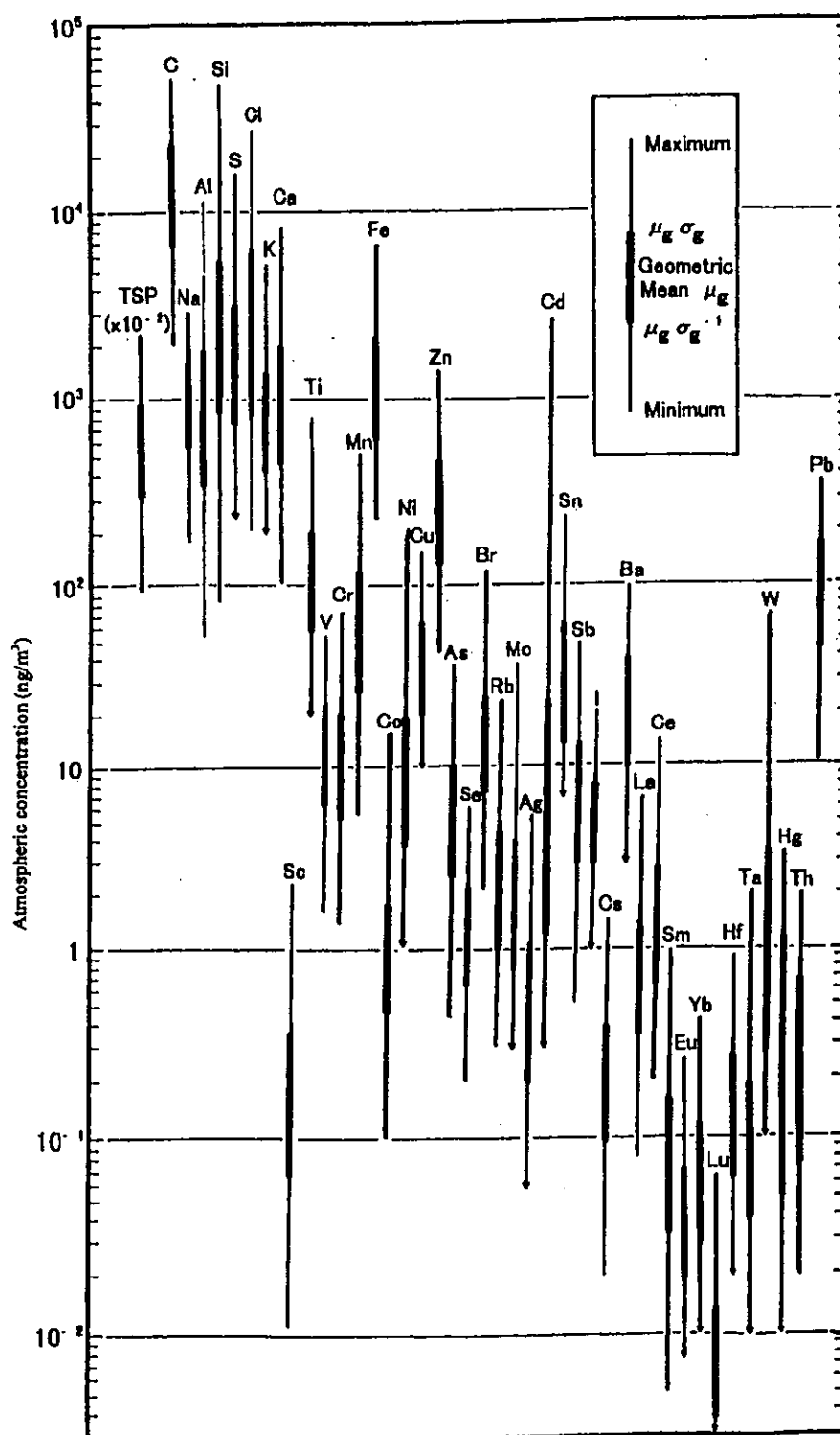


Fig.10.6.1 Concentration of each element in the suspended particulate matter in the atmosphere in a metropolitan area

Table 10.6.1 Component concentration of discharged particles from the main emission sources in the city area

(Unit:  $\mu\text{g/g}$  unless indicated otherwise)

Element	Soil	Sea salt	Iron/ steel Ind.	Trash burning	Petroleum burning	Auto exhaust gas	
						Leaded gasoline	Diesel
Na (%)	1.3	30.42	1.4	12	1.0	0.027	0.10
AL (%)	6.8	0.00003	1.0	0.42	0.21	0.15	0.24
S (%)	—	2.6	4.8	13	9.6	—	—
CL (%)	0.023	55.05	3.4	27	0.092	0.34	—
K (%)	1.6	1.1	1.3	20	0.085	0.12	0.090
Ca (%)	1.6	1.2	4.5	1.1	0.085	—	0.20
Sc	8.6	0.001	1.3	0.46	0.09	0.23	0.36
Ti	4200	0.029	1000	900	740	—	190
V	61	0.058	130	27	9200	2.4	9.6
Cr	46	0.001	3200	850	210	16	12
Mn	790	0.058	22000	330	120	36	44
Fe (%)	3.9	0.000029	15.7	0.62	0.46	0.41	0.11
Co	8.9	0.014	44	21	31	1.8	0.70
Ni	—	0.014	2900	—	4900	39	—
Cu	—	0.017	3700	3600	—	190	—
Zn	290	0.029	52000	26000	400	1500	470
As	18	0.029	100	150	23	—	2.8
Se	5.2	0.12	51	48	—	—	—
Br	1.3	1900	140	830	8.5	6200	38
Rb	86	3.5	77	260	—	—	1.9
Ag	—	0.087	54	150	—	—	—
Cd	—	0.002	250	500	240	—	—
Sn	—	0.087	2800	3000	—	—	—
Sb	37	0.014	90	610	6.9	2.1	4.7
Cs	3.2	0.029	—	12	—	—	0.21
Ba	730	0.86	—	390	920	640	20
La	21	0.009	9.8	7.7	—	—	1.1
Ce	49	0.012	69	170	—	—	2.1
Sm	4.2	—	0.21	0.49	7.6	—	0.13
W	2.5	0.003	47	—	—	—	—
Pb (%)	—	0.087	1.4	1.7	0.033	3.7	—
Th	9.3	0.020	—	—	—	—	0.11
V-C (%)	—	—	—	—	—	—	10.7
NV-C (%)	—	—	—	—	—	—	76.6
SO <sub>4</sub> <sup>2-</sup> (%)	—	—	—	—	—	—	7.7
NO <sub>3</sub> <sup>-</sup> (%)	—	—	—	—	—	—	0.78

(1) The tracer element method

The tracer element method is the simplest calculation process and it is assumed that, concerning the chemical component concentration, unique components (the index components) exist in each emission source that do not exist in other emission sources. The contributed concentration  $S_j$  of the emission source  $j$  which has the index component  $t$  can be given by Eq. (24) which is a transformation of Eq.(23).

$$S_j = \frac{C_t}{a_{ij}} \dots \dots \dots (24)$$

This method is effective only when, regarding the index element, ① the emission source particles  $a_{ij}$  in the receptor are well known and do not change between the emission source and receptor, ②  $C_i$  in the environment sample can be measured with correctness and high precision and ③ the concentration of component  $i$  in the receptor originates only from the emission source  $j$ .

Actually, the above conditions are not completely satisfied and limiting a model to one emission source with one index element means that the other important information contained in the aerosol is not used. This is effective to find the most approximate value or the upper limit value of the source contribution.

(2) The linear programming method

The linear-programming method seeks the combination which maximizes the summation of the source contribution under the conditions given by Eq. (25) and Eq. (26).

$$0 \leq S_j \leq M \quad \dots\dots\dots (25)$$

$$C_i + 3\sigma_{C_i} \geq \sum_{j=1}^p a_{ij} S_j \quad \dots\dots\dots (26)$$

Here,  $M$  is the concentration of the particulate matter and  $\sigma_{C_i}$  is the measurement error of  $C_i$ . These conditions are necessary conditions that source contribution should meet, but they are not sufficient and the physical meaning of the end result is not definite.

(3) The ordinary linear least squares method

In the least squares method, the most probable value of the source contribution  $S_j$  is sought in the case of  $n > p$  by minimizing  $x^2$  which is calculated in Eq. (27).

$$x^2 = \sum_{i=1}^n \frac{(C_i - \sum_{j=1}^p a_{ij} S_j)^2}{\sigma_{C_i}^2} \quad \dots\dots\dots (27)$$

Eq. (23) of CMB becomes Eq. (28) when expressed in a procession.

$$C = AS \quad \dots\dots\dots (28)$$

Here,  $C$  is the component concentration observation value vector of the environment particle of  $n \times 1$ ;  $A$  is the component concentration procession of the emission source particle  $n \times p$  and  $S$  is the emission source contribution concentration vector of  $p \times 1$ . The answer to the least-square method adopted for Eq. (28) is shown in Eq. (29).

$$S = (A'WA)^{-1} A'WC \quad \dots\dots\dots (29)$$

In this formula,  $W$  is a diagonal matrix and its diagonal component is  $\sigma_{C_i}^{-2}$ . Also, the  $t$  and the  $-1$  in the power positions show a transpose matrix and an inverse matrix, respectively. There is no error in the component concentration matrix of the source particle. And then, the estimated error of the emission source contribution which is calculated by the least-square method, supposing that there is an error only in the component concentration observation value of the environment particles is given as the diagonal component of the matrix in Eq. (30).

$$(A'WA)^{-1} \dots\dots\dots (30)$$

(4) The effective variance least-squares method

Since calculations are accompanied by an error in the emission source profile data as well as an error in the established data of the component concentration value of environmental particles, both of these errors must be considered. Effective variance least-squares method is solved in Eq. (23) by multiplying each chemical by the weight percentage which is proportional to the precision of the emission source profile data and the environment concentration measurement data of each chemical component. That is, repeat computations to seek  $S_j$  which minimizes the function  $x^2$  in Eq. (31).

$$x^2 = \sum_{i=1}^n \frac{(C_i - \sum_{j=1}^p a_{ij} S_j)^2}{\sigma_{C_i}^2 + \sum_{j=1}^p \sigma_{a_{ij}}^2 S_j^2} \dots\dots\dots (31)$$

Here,  $\sigma_{a_{ij}}$  is the error which accompanies the measurement of  $a_{ij}$  and the denominator on the right side of Eq. (31) is called the effective dispersion  $V_{eff,i}$ . The answer in Eq. (31) is expressed by the matrix in Eq.(32).

$$S = (A'VA)^{-1} A'VC \dots\dots\dots (32)$$

Here,  $V$  is the diagonal matrix which has  $V_{eff,i}^{-1}$  as the diagonal component. Also, the estimated error of the computed  $S_j$  is given as the diagonal component of the matrix in Eq. (33).

$$(A'VA)^{-1} \dots\dots\dots (33)$$

10.6.3 Index elements

In CMB, the choice of the index component determines the result. Index components are selected after considering ① that they are specific to the emission source and distinguishable from other emission sources, ② that they are measured with high precision and ③ the component concentration does not change in the atmosphere. The components that meet condition number ② do not always have a high concentration because sensitivity and precision depend on the analysis method used. Components which escape from particles in the atmosphere or those that were released in gas and form particles in the atmosphere do not satisfy condition ③. Actually, among the

optional components that satisfy condition (1), the best component which meets conditions (2) and (3) most is chosen.

The components which are often used from the past as the indexes are Al, Sc, Ti, Th (soil burning and coal combustion), Na (sea salt), V, Ni (fuel oil combustion), Fe, Mn, Zn (iron/steel industry), K, Zn (refuse incineration), Pb, Br, C (car exhaust), and so on. These components are used independently or in combination.

#### (10.1~5) References

- 1) The Ministry of International Trade and Industry, Environmental Site Bureau Supervisory; The technique of pollution control and the law (atmosphere edition), The Industrial Environment Management Association (1996)
- 2) About the air pollution real-time supervisory control system, industrial pollution 11, (9) 43-52 (1975)
- 3) Ito, T., Izumikawa, S., Jinnari, H., Ohara, T., Tonooka, Y., Uno, I., Wakamatsu, S.; The building of a photochemical oxidant and simulation model and verification, J. Jpn. Soc. Atom. Environ. 32, (1) 6-28 (1997)
- 4) Kondo, H.; The Thermally Induced Local Wind and Surface Inversion over the Kanto Plain on Calm Winter Nights, J. of Applied Meteorology, 34, (6) 1439-1448 (1995)
- 5) Yamada, Y. and S. Bunker; Development of a nested Grid, Second Moment Turbulence Closure Model and Application to the 1982 ASCOT Brush Creek Data Simulation, J. of Applied Meteorology, 27 (5) 562-578 (1988)

#### (10.6) References

- 1) Mizohata, A.; The Present Situation of the Receptor Model, J. Jpn. Soc. Atom. Environ. 26, (2) 59-71 (1991)
- 2) Environmental Agency, Air Quality Bureau, Air Pollution Regulation Division; The Analysis and Estimate of the Suspended Particulate Matter Pollution, pp.303-314 (1987)

## Chapter 11 System of Air Quality Conservation Laws

### 11:1 Overview

The Basic Environment Law forms the foundation of the system of environmental laws in Japan. Led by the Air Pollution Control Law, the Law Concerning Special Measures to Reduce the Total Mass of Nitrogen Oxides Discharged by Motor Vehicles in Specified Areas (The Motor Vehicle NO<sub>x</sub> Law), the Mine Safety Law, the Electricity Utility Industry Law, the Gas Utility Industry Law, the Road Traffic Law, the Road Transport and Motor Vehicle Law, and the “pollution control ordinances” issued by local governments make up the system of air quality conservation laws that control both the emission of air pollutants such as smoke, soot, injurious gases and dust arising from industrial plants or business establishments and motor vehicle exhaust gases. The government has furthermore been promoting offensive odor control regulations with the Offensive Odor Control Law. (Table 11.1.1)

In keeping with the increasing importance of the global environment issue, Japan has entered a number of international conventions and protocols for the prevention of global warming and the depletion of the ozone layer and has enacted domestic laws to enforce these measures. As can be seen here, the Japanese system of air quality conservation laws is complex. This chapter thus begins with the introduction of these basic items.

Table 11.1.1 Air Pollution Control Statutes and Contents

Law	Regulation Contents
○ Air Pollution Control Law (Law 97, 1968)	Regulates smoke, soot, injurious gas and dust emissions (emissions standards, etc.), sets maximum limits on vehicle exhaust emissions and fuels.
○ Motor Vehicle NO <sub>x</sub> Law (Law 70, 1992)	Regulates the types of vehicles that can be used in specified areas.
○ Electricity Utility Industry Law (Law 170, 1964)	Sets technical standards for electric installations and issues orders to conform to technical standards.
○ Gas Utility Industry Law (Law 51, 1954)	Sets technical standards for gas installations and issues orders to conform to technical standards.
○ Mine Safety Law (Law 70, 1949)	Prevents mine pollution from gas, dust and smoke (smoke emissions standards, safety orders, etc.).
○ Quarry Law (Law 291, 1950)	Approves quarry plans and sets up preventative measures against disasters quarrying suspension orders, etc..
○ Road Transport and Motor Vehicle Law (Law 185, 1951)	Sets up vehicle exhaust emission controls (sets safety standards for road transportation vehicles).
○ Road Traffic Law (Law 105, 1960)	Sets up controls on poorly maintained vehicles (driving suspension orders and traffic control like road blockage, traffic signals, etc.).
○ Law Concerning Rational Use of Energy (Law 49, 1979)	Prevents gas emissions with a more efficient use of energy.



### 11.1.1 Laws and Ordinances

A “law” is a form of national legislation which is enacted, in principle, on passage by both Houses of the Diet, and it has the highest force after the Constitution and international conventions. An “ordinance” is local legislation enacted by a resolution of the local assembly for the purpose of managing the local governments administrative affairs based on the right of enactment guaranteed by the Constitution. An ordinance represents the fundamental administrative act of a local governments based on its own judgment.

### 11.1.2 Conventions, Protocols, and Declarations

A “convention”, in a broad sense, refers to a written agreement between states or international organizations, and it may be called a “charter”, a “convention”, an “agreement” (or arrangement), or a “protocol”. An agreement by convention can only be concluded between bodies with the power to sign a convention. In Japan, the Cabinet is vested with this power, and it is required to obtain the approval of the Diet. A “convention” takes precedence over domestic laws, and a domestic law is usually enacted in order to implement a convention.

A “protocol”, in a broad sense, is also a convention. The term “protocol” usually refers to an existing convention which has been modified or supplemented. An “agreement”, or “arrangement”, is often used to refer to a relatively technical or insignificant arrangement between states. A “declaration” refers to a document which has been prepared between international organizations, or to a written statement or mutual pact which has been signed between countries.

## 11.2 The Basic Environment Law

### 11.2.1 Overview

Basic Environment Law sets down the fundamental requirements for environmental administration, and it was enacted in 1993 in consideration of the importance of environmental pollution control, protection of the natural environment, and international cooperation. This law sets forth basic principles of environmental conservation and articulates the responsibilities to be borne by the state, the local governments, the enterprises and the nation's citizens. Its objectives are to determine the fundamental policies for environmental conservation measures, to comprehensively and systematically promote those policies to ensure healthy, secure and culturally rich living for the present and future generations of people, and thereby to contribute to the well-being of mankind. The Law defines "environmental impact", "global environmental conservation", and "environmental pollution" and calls for the needs to ensure that people can enjoy the blessings of the environment and pass these to future generations and to form a society with low environmental impact so that sustainable development is ensured, and to conserve the global environment through international cooperation. The Law designates an "Environment Day", makes provisions for the "basic environment plan", "environmental quality standards", "environmental pollution control programs", "environmental impact assessment", "environmental pollution controls", "protection of the natural environment", "economic measures", "improving and developing facilities", "environmental education and study", "provision of information", "implementation of surveys and investigations", "establishment of surveillance and monitoring systems", "promotion of science and technology", "settlement of environmental pollution disputes and relief to victims", "international cooperation for the environment", "measures for the local governments and private organizations", "measures by the local governments", and for the "bearing of costs and financial measures". The following sections outline specific provisions that affect air quality conservation.

### 11.2.2 Environmental Quality Standards

The provisions in the Basic Law on environmental quality standards read: "1. With regard to environmental conditions related to air pollution, water pollution, soil contamination and noise, the government shall respectively establish environmental quality standards which is desirable to maintain for the protection of human health and the conservation of the living environment, 2. (omitted), 3. With regard to the standards set forth in Paragraph 1, due scientific consideration shall always be given and such standards shall be revised whenever necessary, 4. The government shall make efforts to attain the standard provided for in Paragraph 1, by comprehensively and effectively implementing policies concerning environmental pollution control, which are set forth in this chapter (hereinafter referred to as "environmental pollution control policies)." Environmental quality standards serve as the target to be achieved by the environmental administration. The standards for sulfur dioxide (daily average of hourly values below 0.04 ppm) and nitrogen dioxide (daily average of hourly values between the range of 0.04 and 0.06 ppm) are set as the environmental quality targets in the regulation of total emission that will be touched on later and treated as key elements in pollution control programs and the environmental impact assessment system.

### 11.2.3 Environmental Pollution Control Programs

The Basic Law provides: "1. The Prime Minister shall instruct the prefectural governors concerned to formulate programs on measures for environmental pollution control (hereinafter referred to as the "Environmental Pollution Control Program"); concerning areas specified as either of the following categories; by presenting the basic directions of the environmental pollution control policies to be implemented in those areas: 1) areas where environmental pollution is currently serious and where it is recognized that it is very difficult to prevent environmental pollution unless the environmental pollution control policies are comprehensively implemented; 2) areas where environmental pollution is likely to become serious due to the rapidly increasing concentrations of population, industry, etc., and where it is recognized that it will be very difficult to prevent environmental pollution unless the environmental pollution control policies are comprehensively implemented, 2. The basic directions provided for in the preceding Paragraph shall be formulated on the basis of the Basic Environment Plan, 3. When the prefectural governor concerned has received the instruction provided for in Paragraph 1, he/she shall formulate an Environmental Pollution Control Program in accordance with the basic directions provided for in the same Paragraph and shall submit it to the Prime Minister for his/her approval, 4. Prior to issuing the instruction under Paragraph 1 or giving the approval required under the preceding Paragraph, the Prime Minister shall ask the Conference on Environmental Pollution Control for its decision, 5. Prior to issuing the instruction under Paragraph 1, the Prime Minister shall hear the opinions of the prefectural governors concerned." "The State and local governments shall make efforts to take measures necessary for the attainment of the Environmental Pollution Control Programs." Thirty-four regions across the country have formulated, or are reviewing, their respective environmental pollution control programs at present.

### 11.2.4 International Cooperation

The Basic Law provides that the State shall endeavor to attain international cooperation for global environmental conservation, take measures necessary for promoting international cooperation, and support environmental conservation in developing countries. The State is required to obtain international cooperation for surveillance; monitoring and observation, take steps to promote international cooperation conducted by local governments and private organizations, endeavor to take global environmental conservation into account whenever it engages in international cooperation activities, and provide information to enterprises and take other measures necessary for them to give proper consideration to global environmental conservation in their business activities that are carried out abroad.

## 11.3 Air Pollution Control Law

### 11.3.1 Overview

The Air Pollution Control Law was established in 1968, based on the Smoke and Soot Regulation Law, which was passed by the 40th session on the Diet in May 1962, and it has been amended several times since its enactment.

The Air Pollution Control Law was enacted to promote the public health and preserve the living environment with respect to air pollution through regulating the emission of smoke and soot (sulfur oxides, soot and dust, and other toxic substances) arising from industrial plants or business establishments as a result of industrial activities, and establishing the maximum permissible limits of motor vehicle exhausts.

Subsequently, against the background of the progress of air pollution brought about by the increased consumption of petroleum-based fuels, photochemical smog, air contamination caused by toxic substances such as hydrogen fluoride, cadmium and lead, and the problem of fumigation by automobile exhaust gas, efforts were made to tighten control. These efforts included the introduction of the system of absolute liability of enterprises and regulation of total emission on sulfur oxides and nitrogen oxides, and a series of emission standards revises and addition of controlled substances.

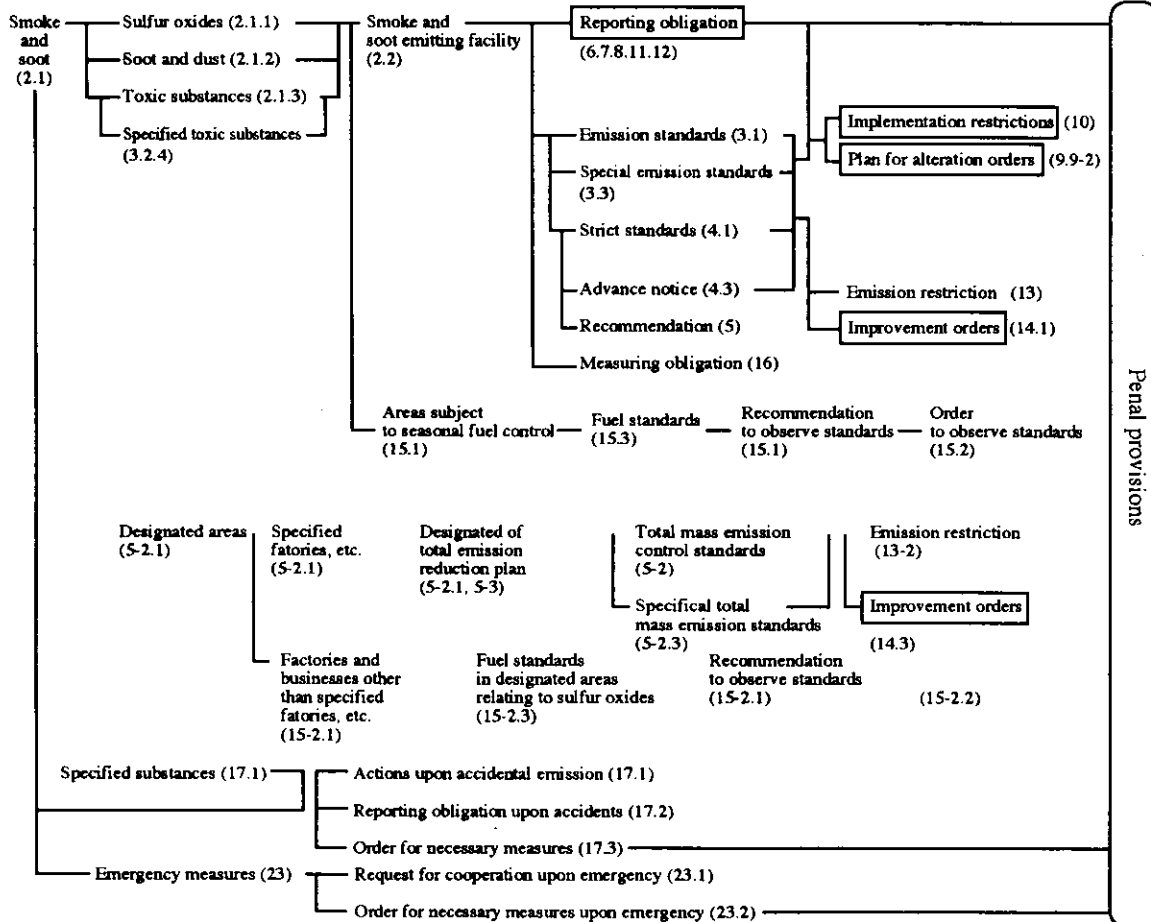
The Law was further revised in 1996 to include provisions that require implementation of measures necessary for the prevention of undesirable effects on people's health, with respect to benzene and other harmful air pollutants that are feared to adversely affect human health by long-term exposure to low concentrations. The structure of the Air Pollution Control Law is given in Table 11.3.1.

**Purpose**

To promote public health and preserve the living environment with respect to air pollution through:  
 1) Regulating the emission of soot, smoke and dust arising from industrial plants/businesses as a result of industrial activity or the demolition of buildings.  
 2) Promoting implementation of measures to control harmful air pollutants.  
 3) Establishing the maximum limits on vehicle exhaust gas  
 4) Protecting victims by making provisions for the liability of enterprises to compensate for damages where such enterprises cause air pollution which damages human health.

**Smoke and Soot Control**

**<Chapter II> #See Chapter 4 for emergency measures.**



**Dust**

**Dust <Chapter II-2>**

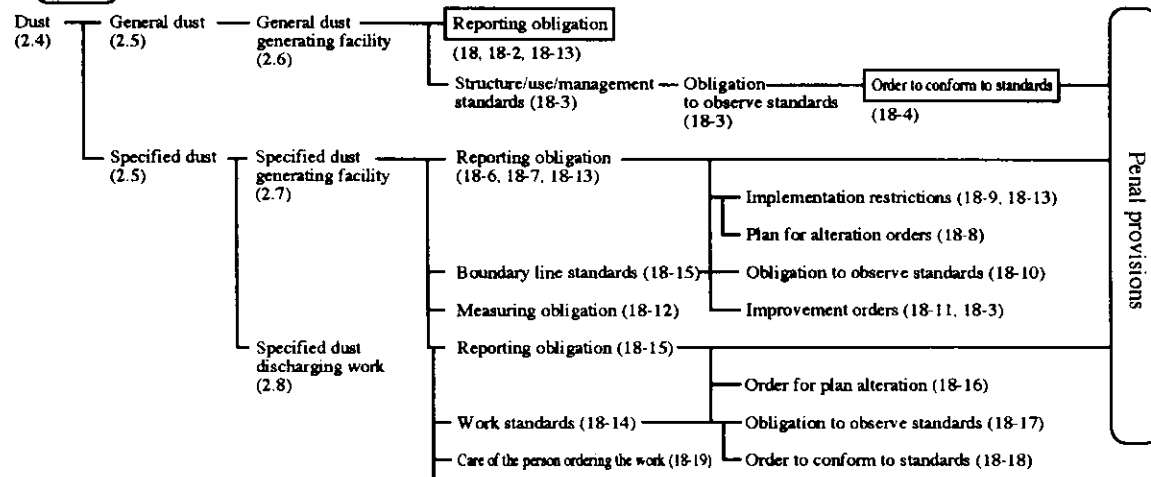


Fig.11.3.1 (1) Structure of the Air Pollution Control Law Purpose

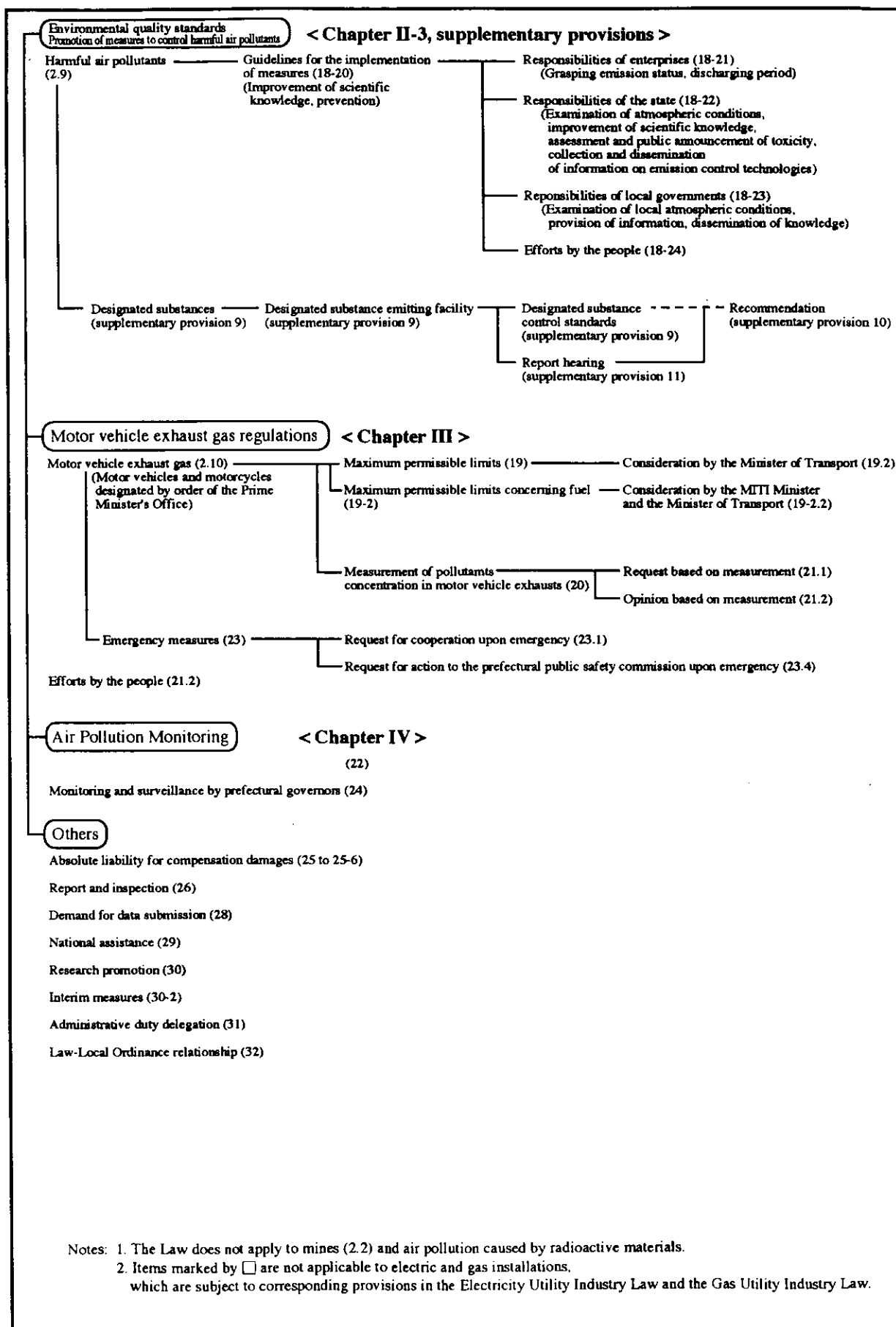


Fig.11.3.1 (2) Structure of the Air Pollution Control Law Purpose

### 11.3.2 Smoke and Soot

#### (I) Smoke and Soot and Smoke and Soot Emitting Facilities

In the Air Pollution Control Law, "smoke and soot" means (i) sulfur oxides, (ii) soot and dust, and (iii) toxic substances designated by Cabinet Order (cadmium and its compounds; chlorine and hydrogen chloride; fluorine and its compounds; lead and its compounds; and nitrogen oxides), that are generated as a result of combustion of fuel and other materials. "Smoke and soot emitting facilities" mean any facility that meets the following three conditions: (i) that it is installed in an industrial plant or a business establishment (excluding mines); (ii) that it generates and emits smoke and soot; and (iii) that the smoke and soot so emitted cause air pollution. More specifically, boilers, heating furnaces, roasting furnaces, blast furnaces, drying furnaces, waste incinerator, gas turbines, diesel engines, gasoline engines and so on that are over a certain size are designated as such facilities by Cabinet Order (Table 11.3.1).

Table 11.3.1 (1) Smoke and Soot Emitting Facilities Targeted by the Air Pollution Control Law

	Facility	Size
1	Boiler	<ul style="list-style-type: none"> <li>· Heating area of 10 square meters and over</li> <li>· Combustion ability of 50 liters/hour and over</li> </ul>
2	Gas generating furnace, heating furnace	<ul style="list-style-type: none"> <li>· Raw material treatment ability of 20 tons/day</li> <li>· Combustion ability of 50 liters/hour and over</li> </ul>
3	Roasting furnace, sintering furnace (machine)	<ul style="list-style-type: none"> <li>· Raw material treatment ability of 1 ton/day and Over</li> </ul>
4	(Metal refining use) Blast furnace, converter, open-hearth furnace	
5	(Metal refining and casting use) Smelting furnace	<ul style="list-style-type: none"> <li>· Fire grid area of 1 square meter and over</li> <li>· Tuyere area of 0.5 square meters and over</li> <li>· Combustion ability of 50 liters/hour and over</li> <li>· Transformer nominal capacity of 200 kva and Over</li> </ul>
6	(Metal forging, rolling and heat treatment use) Heating furnace	
7	(Petroleum product, petrochemical product, and coal tar product manufacturing use) Heating furnace	
8	(Petroleum refining use) Fluid contact disintegration device- catalytic converter	<ul style="list-style-type: none"> <li>· Ability to heat the carbon adhering to the catalyst of 200 kg/hour and over</li> </ul>
8-2	Sulfur collecting facility attached to the oil gas cleansing device	<ul style="list-style-type: none"> <li>· Combustion ability of 6 liters /hour and over</li> </ul>
9	(Ceramics manufacturing use) Kiln, smelting furnace	<ul style="list-style-type: none"> <li>· Fire grid area of 1 square meter and over</li> <li>· Combustion ability of 50 liters/hour and over</li> <li>· Transformer nominal capacity of 200 kva and over</li> </ul>
10	(Inorganic chemical industry product and foodstuff industry use) Reaction furnace (including carbon black manufacturing fuel burning device), direct heating furnace	
11	Drying furnace	
12	(Iron, steel, alloyed iron, carbide manufacturing use) Electric furnace	<ul style="list-style-type: none"> <li>· Transformer nominal capacity of 1,000 kva and over</li> </ul>
13	Waste incinerator	<ul style="list-style-type: none"> <li>· Fire grid area of 2 square meters and over</li> <li>· Incineration ability of 200 kg/hour and over</li> </ul>

Table 11.3.1 (2) Smoke and Soot Emitting Facilities Targeted by the Air Pollution Control Law

	Facility	Size
14	(Copper, lead, zinc refining use) Roasting furnace, sintering furnace (including pellet sintering furnace), blast furnace, converter, drying furnace	<ul style="list-style-type: none"> <li>· Raw material treatment ability of 0.5 tons/hour and over</li> <li>· Fire grid area of 0.5 square meters and over</li> <li>· Tuyere area of 0.2 square meters and over</li> <li>· Combustion ability of 20 liters /hour and over</li> </ul>
15	(Cadmium-based pigments and cadmium carbonate manufacturing use) Drying facilities	<ul style="list-style-type: none"> <li>· Capacity of 0.1 cubic meters and over</li> </ul>
16	(Chlorinated ethylene manufacturing use) Chlorine quick cooling facility	<ul style="list-style-type: none"> <li>· Chlorine treatment ability of 50 kg/hour and over</li> </ul>
17	(Ferric chloride manufacturing use) Smelting furnace	
18	(Activated charcoal [limited to the method which uses zinc chloride] manufacturing use) Reaction furnace	<ul style="list-style-type: none"> <li>· Combustion ability of 3 liters /hour and over</li> </ul>
19	(Chemical product manufacturing use) Chlorine reaction facilities, hydrogen chloride reaction facilities, hydrogen chloride absorbing facilities	<ul style="list-style-type: none"> <li>· Chlorine treatment ability of 50 kg/hour and over</li> </ul>
20	(Aluminum refining use) electrolytic furnace	<ul style="list-style-type: none"> <li>· Electric current capacity of 30 ka and over</li> </ul>
21	(Phosphorus, phosphoric acid, phosphoric acid fertilizer, compound fertilizer [products which use phosphorus as a raw material] manufacturing use) Reaction furnace, enrichment facilities, Kiln, smelting furnace	<ul style="list-style-type: none"> <li>· Mineral phosphate treatment ability of 80 kg/hour and over</li> <li>· Combustion ability of 50 liters/hour and over</li> <li>· Transformer nominal capacity of 200 kva and over</li> </ul>
22	(Hydrofluoric acid manufacturing use) Enrichment facilities, absorption facilities, distillation facilities	<ul style="list-style-type: none"> <li>· Heating area of 10 square meters and over</li> <li>· Pumping power of 1kw and over</li> </ul>
23	Sodium tripoli phosphate manufacturing use [products which use phosphorus as a raw material]) Reaction furnace, drying furnace, Kiln	<ul style="list-style-type: none"> <li>· Raw material treatment ability of 80 kg/hour and over</li> <li>· Fire grid area of 1 square meter and over</li> <li>· Combustion ability of 50 liters/hour and over</li> </ul>
24	(Secondary lead refining [lead alloy manufacturing], lead piping, sheeting and line manufacturing use) Smelting furnace	<ul style="list-style-type: none"> <li>· Combustion ability of 10 liters hour and over</li> <li>· Transformer nominal capacity of 40 kva and over</li> </ul>
25	(Lead storage battery manufacturing use) Smelting furnace	<ul style="list-style-type: none"> <li>· Combustion ability of 4 liters /hour and over</li> <li>· Transformer nominal capacity of 20 kva and over</li> </ul>
26	(Lead-based pigments manufacturing use) Smelting furnace, reverberatory furnace, reaction furnace, drying facilities	<ul style="list-style-type: none"> <li>· Capacity of 0.1 cubic meters and over</li> <li>· Combustion ability of 4 liters /hour and over</li> <li>· Transformer nominal capacity of 20 kva and over</li> </ul>
27	(Nitric acid manufacturing use) Absorption facilities, bleaching facilities, enrichment facilities	<ul style="list-style-type: none"> <li>· Nitric acid synthesis, bleaching, enrichment abilities of 100 kg/hour and over</li> </ul>



Table 11.3.1 (3) Smoke and Soot Emitting Facilities Targeted by the Air Pollution Control Law

	Facility	Size
28	Cokes oven	· Raw material treatment ability of 20 tons/ hour and over
29	Gas turbine	· Combustion ability of 50 liters/hour and over
30	Diesel engine	
31	Gas engine	· Combustion ability of 35 liters /hour and over
32	Gasoline engine	

Note: · When the facilities are of a number of different sizes, each one can be categorized appropriately as a smoke and soot generating facility.

· “Combustion ability” refers to the combustion capacity per hour when calculating fuel on the basis of heavy oil.

## (2) Smoke and Soot Emission Control

Means and methods of control applicable to smoke and soot can be roughly divided into the following four categories:

### 1) General emission standards

Emission standards established by the State as the national minimum according to the type of smoke and soot emitting facility.

### 2) Special emission standards

These special emission standards, which are stricter than general one, are applicable to new smoke and soot emitting facilities established in areas where location of smoke and soot emitting facilities, which emit smoke and soot containing sulfur oxides, soot and dust or specific toxic substances, are concentrated (an area with a heavy concentration of facilities).

### 3) Strict emission standards

These are stringent emission standards which are established by prefectures according to their respective ordinances, to take the place of general emission standards, when it is recognized that existing general or special emission standards are inadequate to protect public health or conserve the living environment in an area under their jurisdiction. Prefectures are authorized to issue strict standards for the emission of toxic substances other than soot and dust and sulfur oxides. Twenty-one prefectures have these standards at present.

### 4) Regulation of total emission standard

These standards are applicable to specified factories emitting designated smoke and soot in designated areas where it is recognized to be difficult to attain the prescribed environmental quality standards solely with the emission standards referred to from clauses 1 to 3 above.

Unlike emission standards, the regulation of total emission standards are not standards which are applied to each unit of a facility, but they are concerned with the total mass emission of designated smoke and soot generated at all the smoke and soot emitting facilities at specified factories. The governor of the prefecture prescribes this standard

based on relevant total mass emissions reduction plan. Sulfur oxides and nitrogen oxides are designated smoke and soot substances at present, and regulation of total emission standards are prescribed for these substances.

Twenty-four areas (accounting for about 33 percent of the total population and about 56 percent of the national fuel consumption) are designated by the regulation of total emission standards for sulfur oxides, and these have been subject to full control since May 1978. Standards for nitrogen oxides have been designated for three areas (Tokyo, Osaka, and Kanagawa Prefectures) and have been in effect since 1982.

In an area where emission standards alone do not provide adequate control over sulfur oxides, the governor of the prefecture may prescribe a fuel standard for the sulfur contents of fuels used in such an area and may recommend or order enterprises and other persons in the area to observe the said fuel standard. Fuel standards may be divided into those applicable for a prescribed period to cope with air pollution caused by things like the heating of buildings in city centers and those applicable to any person who establishes a factory or business, other than a specified factory, within an area designated for sulfur oxides.

Regarding actions at the time of an accident, the person who installed a smoke and soot emitting facility or specified facility (a facility which generates any of the 28 specified substances, including ammonia and benzene, which are designated by Cabinet Order as substances liable to adversely affect human health or the living environment, and is not a smoke and soot emitting facility) is required to take emergency measures promptly to restore normal conditions upon mechanical failure or any other accident that causes discharge of a large quantity of smoke and soot or specified substances, and notify the governor of the prefecture of the occurrence.

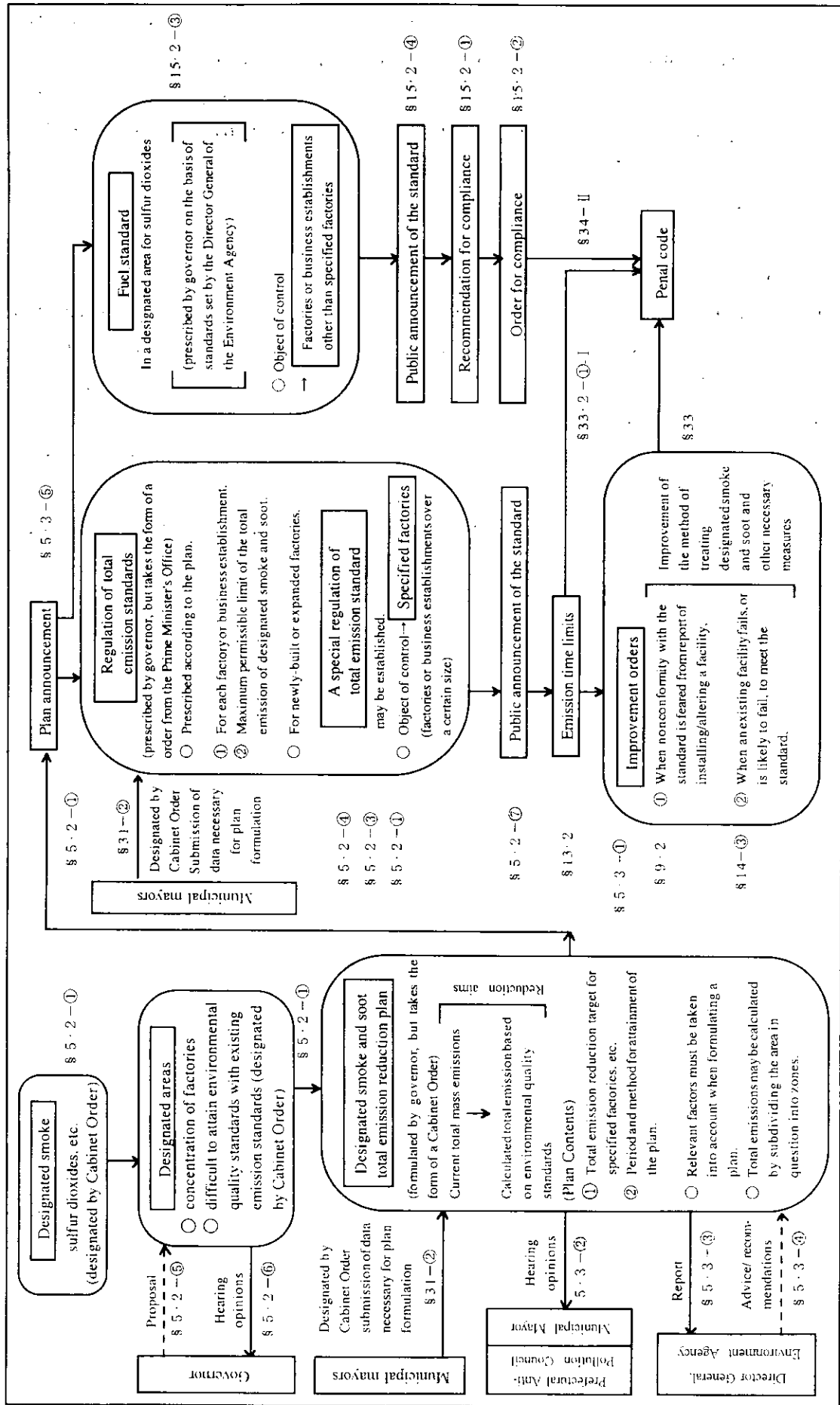


Fig. 11.3.2 Regulation of Total Emission Structure Outline

### 11.3.3 Particulate dust

In the Air Pollution Control Law, the term “particulate dust” means any substance discharged or scattered as a result of crushing, sorting or any other mechanical treatment of materials, or the piling up of materials. Regulations on general particulate dust are prescribed in the form of standards for the structure, use and management of general particulate dust emitting facilities. Asbestos is designated as specified particulate dust liable to adversely affect human health, and the maximum permissible level of its concentration in the ambient air along the boundary line of the premises of a factory or business establishment is prescribed. Other control measures are also in place such as work standards designed to prevent the scattering of asbestos from building demolition sites.

### 11.3.4 Motor Vehicle Exhausts

With the rapid progress of motorization from the late 1950s, motor vehicles have become an important means of transportation for economic activity as well as in people’s daily lives. On the other hand, they have also caused serious air and noise pollution, vibration and other environmental pollution and have raised major social problems. In response to this, regulations on motor vehicle exhausts were introduced from the late 1960s, and these have been strengthened gradually.

To control motor vehicle exhaust gas in Japan, the Director General of the Environment Agency establishes the maximum permissible amounts of substances causing air pollution discharged by motor vehicles based on the results of air pollution monitoring and future prospects for technologies to reduce motor vehicle exhausts. The Minister of Transport in turn establishes necessary measures to control motor vehicle exhausts, in the form of safety standards, under the Road Transport and Motor Vehicle Law, to ensure that the said maximum permissible limits are maintained. Controls on new-model and existing-model vehicles are secured by vehicle inspection. The maximum permissible limits were formerly established for standard-, small-, and mini-sized motor vehicles with respect to carbon monoxide, hydro-carbons, nitrogen oxides, particulates (for diesel-powered vehicles only) and diesel smoke. Motorcycles will be subject to control starting from 1998.

### 11.3.5 Controls on Harmful Air Pollutants

The amendment of the Air Pollution Control Law in May 1996 called for the implementation of measures necessary for the prevention of undesirable effects on human health with respect to harmful air pollutants that are feared to adversely affect human health by long-term exposure to low concentrations.

Of the harmful air pollutants, benzene, trichloroethylene and tetrachloroethylene have been specified as designated substances whose emission or scattering must be prevented immediately. Facilities that discharge or scatter these substances are designated as designated substance discharging facilities and are subject to substance control standards established for each facility. (Fig.11.3.3)

watasi

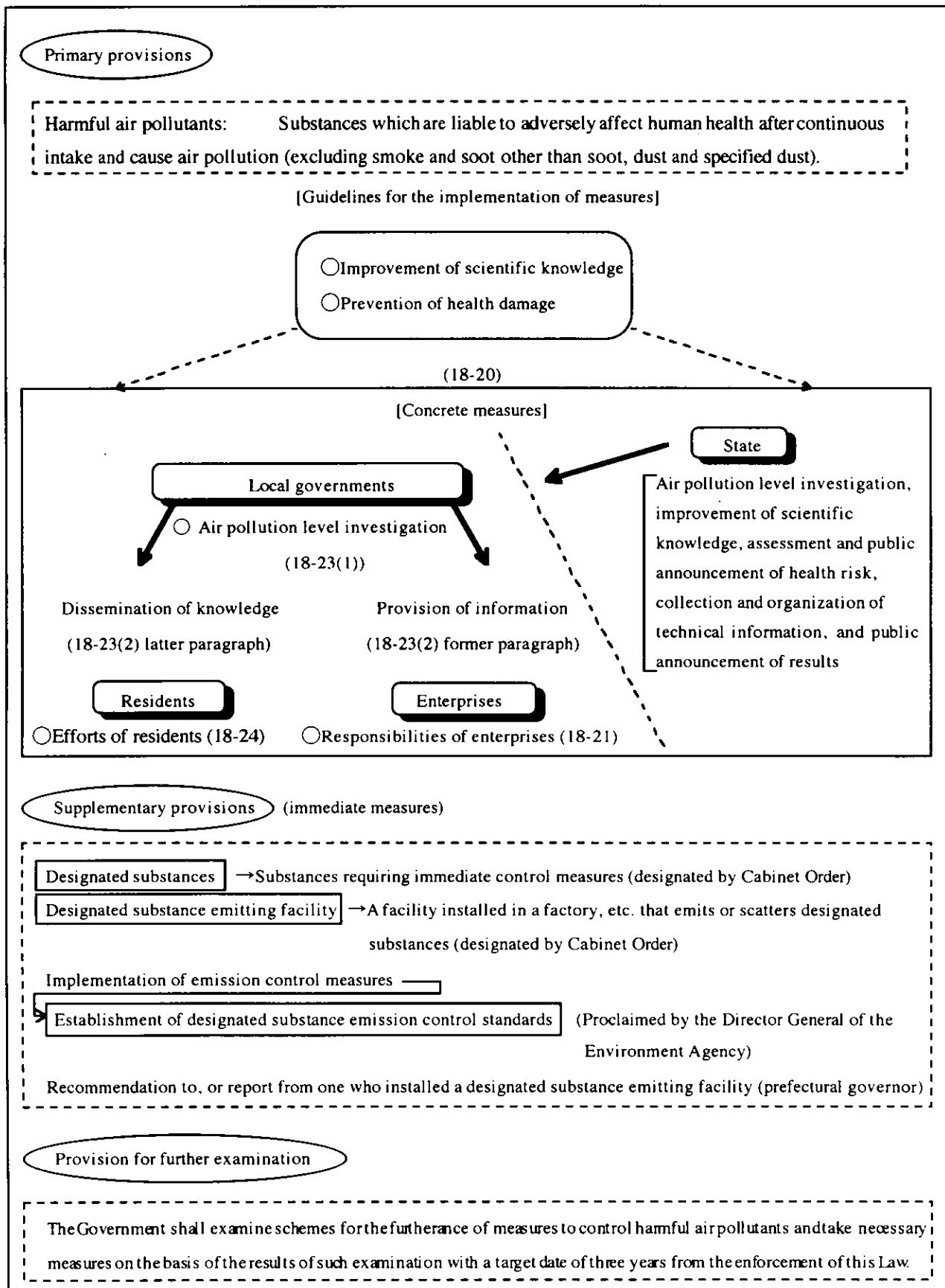


Fig.11.3.3 Outlines of measures to control harmful air pollutants under the revised Air Pollution Control Law

## 11.4 Law Concerning Special Measures to Reduce The Total Mass of Nitrogen Oxides Discharged by Motor Vehicles in Specified Areas (The Motor Vehicle NOx Law)

### 11.4.1 Background

To cope with the problem of air pollution from nitrogen oxides, the Air Pollution Control Law has been amended to intensify control over factories, business establishments and motor vehicle exhausts (single source control). Nevertheless, air pollution due to nitrogen oxides remains serious in metropolitan districts, and polluted areas have expanded. Load factors, as the source of nitrogen oxide from emission motor vehicles, have remained extremely high in recent years, and these stood at 71 percent in Tokyo and 53 percent in Osaka in 1990.

To reduce area-wide total load of nitrogen oxides discharged from motor vehicles in areas where it is expected to be difficult to attain the prescribed environmental quality standard for nitrogen dioxide solely with the reinforcement of single source control, the Law Concerning Special Measures to Reduce the Total Mass of Nitrogen Oxides Discharged by Motor Vehicles in Specified Areas was enacted in 1992.

### 11.4.2 Overview

A total of 196 cities, wards, towns and villages in Tokyo metropolitan, Saitama, Chiba, Kanagawa, Osaka and Hyogo Prefectures were designated under the Law as specified areas with significant air pollution due to nitrogen oxides exhausted from motor vehicles. The following measures are being carried out with a view to roughly achieving the prescribed environmental quality standard for nitrogen dioxide by the end of fiscal 2000 (Fig.11.4.1):

#### (1) Basic plan for total mass emissions reduction and the total mass emissions reduction program

The State prescribes the basic plan for total mass emissions reduction as the master plan under the Law. As specific measures to achieve total mass emission reduction in the basic plan, the State is required to tighten single source control over motor vehicles, implement control by vehicle categories, encourage the spread of pollution-free automobiles, promote measures to facilitate the flow of goods, people and traffic, promote localized pollution control, and promote activities to disseminate knowledge and raise awareness. Each prefecture on its part formulates a total mass emissions reduction program in accordance with the basic plan and enforces necessary measures.

#### (2) Emission Standards for specified motor vehicles

Emission standards are established for specified motor vehicles such as cargo trucks and buses that are operated within a specified area as the home service area. Vehicles that fail to meet the prescribed standards are subject to various measures such as non-issuance of official car inspection certificates.

#### (3) Guidelines for the rational use of motor vehicles

The competent minister overseeing business establishments draws up guidelines for facilitating the rational

use of motor vehicles and provides the enterprises with necessary directions and advice to help reduce the amounts of nitrogen oxide emissions.

(Promulgated on June 3, 1992. "Regulations by vehicle categories" enforced as of December 1, 1993)

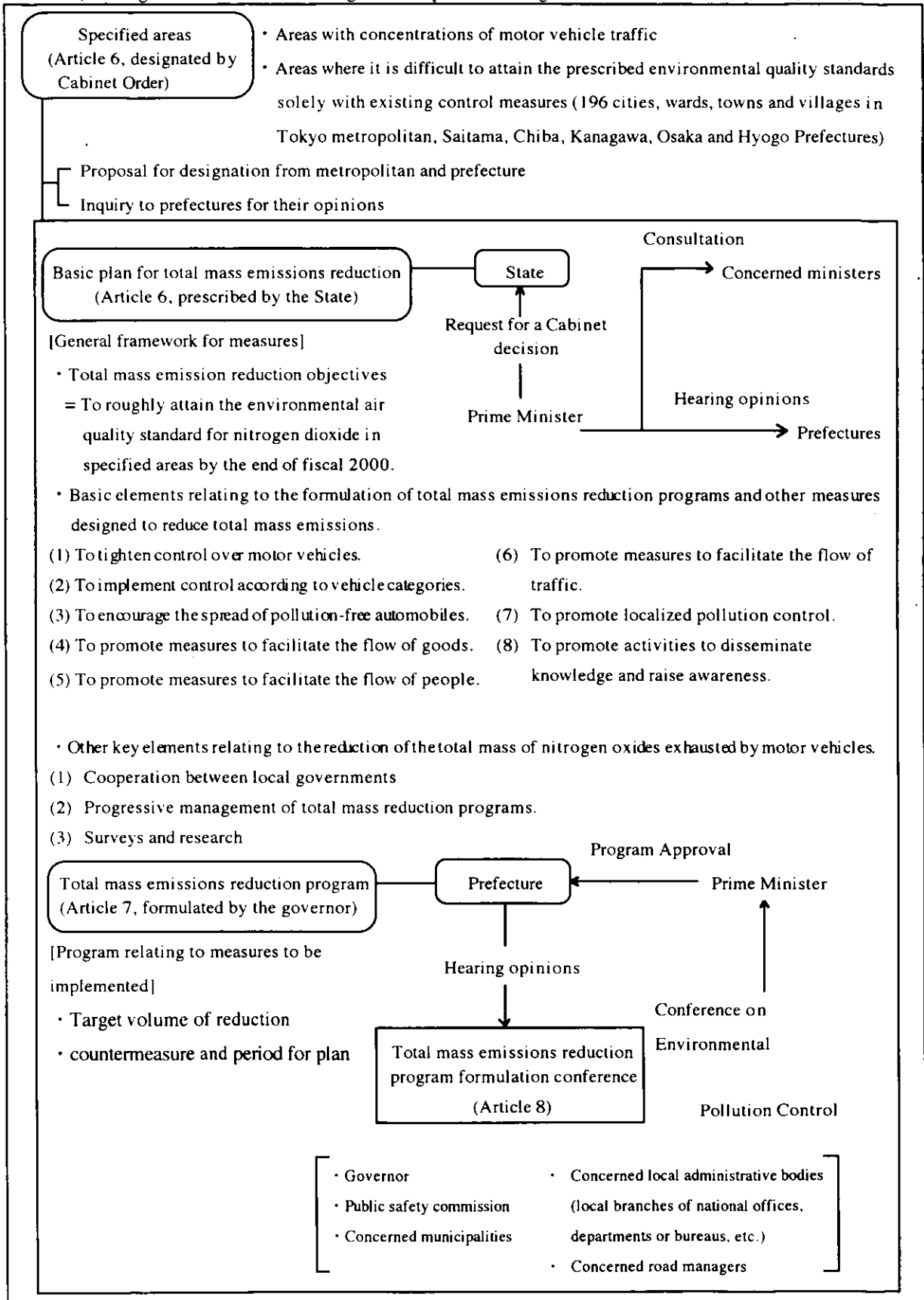


Fig.11.4.1 (1) Structure of the Motor Vehicle NOx Law



(Promulgated on June 3, 1992. "Regulations by vehicle categories" enforced as of December 1, 1993)

State assistance (Article 15)

Application of emission standards to specified motor vehicles (control according to vehicle categories) (Articles 10-12)

- Cargo trucks, buses and other special motor vehicles that are operated within a specified area as the home service area and which fail to meet the emission standards for specified motor vehicles (table below) will not be permitted to operate.

Vehicle classification by total weight	Emission standards for specified motor vehicles
1.7 t or less	Equivalent to gasoline or LPG-fueled vehicles under 1988 registration
Over 1.7t - 2.5t or less	Equivalent to gasoline or LPG-fueled vehicles under 1989 registration
Over 2.5t - 5.0t or less	Equivalent to accessory cell diesel-powered vehicles under 1989 regulation (*)
Over 5.0t	Equivalent to direct injection diesel-powered vehicles under 1989 regulation (*)

\* Note: Vehicles produced in or after September 1995 are subject to standards equivalent to those under 1994 regulations.

Category	Number of years from initial registration	Classification number on number plate
Ordinary-sized cargo vehicle	9	1 and 10-19
Small-sized cargo vehicle	8	4 and 40-49, 6 and 60-69
Large bus (seating capacity of 30 or more persons)	12	2 and 20-29
Microbus (seating capacity of 11 or more but under 30 persons)	10	2 and 20-29 (5 and 50-59, and 7 and 70-79 for some)
Special vehicle (based on a cargo truck or bus)	10	8 and 80-89

\* Control according to vehicle categories is secured by the Road Transport and Motor Vehicle Law (An official car inspection certificate will not be issued to a non-conforming vehicle).

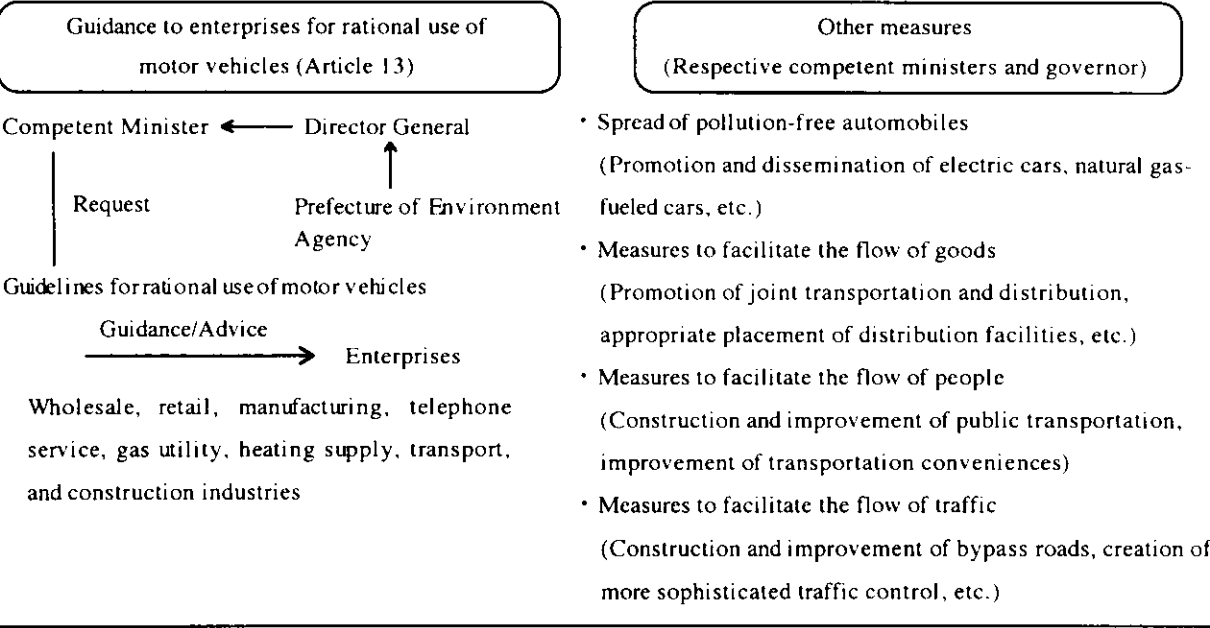


Fig. 11.4.1 (2) Structure of the Motor Vehicle NOx Law

## 11.5 Offensive Odor Control Law

Offensive odors are typical sensorial pollution. Offensive odor claims or complaints are often registered against, among other facilities, fish gut and bone processing plants, slaughter houses and oil refineries. An actual example of such a complaint was the problem of offensive odors which were emitted from waste matter containing mercaptan which was dumped illegally in the harbor south of Kawasaki City in August and September of 1963, and which affected a wide area extending to the vicinity of Kita-Ku, Tokyo and Naka-Ku, Yokohama City.

Nevertheless, due to the stagnation in the development of effective preventative technologies and the difficulty of making quantitative assessments of offensive odors, regulatory actions were delayed until 1971 when the Offensive Odor Control Law was enacted. The Law aims at preserving the living environment and contributing to the protection of people's health by regulating the emissions of offensive odorous substances arising from the activities of factories and business establishments. Areas to be regulated are designated by the governor of the prefecture who also regulates offensive odors by establishing control standards for the 22 offensive odorous substances in terms of atmospheric concentrations along the boundary lines of factory sites or in terms of discharged gas concentrations and concentrations in exhaust water, within ranges prescribed by Cabinet Order. The Olfactory Sense Test was adopted as a regulatory method in 1996.

## 11.6 Law Concerning the Improvement of Pollution Prevention Systems in Specific Factories

The basic framework of the system of environmental pollution control laws was completed with the enactment of the Air Pollution Control Law and the Noise Pollution Control Law in 1968, and the Vibration Regulation Law in 1970. Although education and training of the technical staff of government agencies involved with environmental pollution administration has been conducted by the Institute of Public Health, Ministry of Health and Public Welfare and the Agency of Industrial Science and Technology, Ministry of International Trade and Industry, since the mid-1950s, industrial enterprises that carry out actual implementation of environmental pollution controls could not follow these developments in terms of the organizational set up and were particularly behind in the area of training qualified technical staff.

The Law Concerning the Improvement of Pollution Prevention Systems in Specific Factories was thus enacted in 1971. The Law places enterprises under obligation to establish pollution prevention systems at their own factories and business establishments which emit environmental pollutants. The purpose of the Law is to develop pollution prevention systems at specified factories by introducing the system of pollution control management and creating a framework for organization-wide efforts to prevent environmental pollution. Manufacturers, and electric power, gas or heating utility suppliers over a certain size that have subject facilities are designated as "specific factories", or business establishments, subject to the Law's provisions. Each specific factory is required to make organization-wide efforts to prevent environmental pollution by appointing a pollution control supervisor, a chief manager of pollution control, and a pollution control manager, whose identities must be registered with the governor of the prefecture.

Pollution control managers and the chief managers of pollution control must be qualified individuals who have passed national examinations. Pollution control managers are categorized as follows: Class 1-4 Air: Specified Dust, Class 1-4 Water Quality, Noise, and Vibration.

## **11.7 Environmental Impact Assessment Law**

The Environmental Impact Assessment Law was enacted on June 9, 1997. Considerable importance is attached to air quality conservation in the system prescribed under this law. This law and the environmental impact assessment system will be outlined in Chapter 12.

## **11.8 Law Concerning Rational Use of Energy (Energy Saving Law)**

Most of the global energy needs had been met by the use of oil from the late 1950s, but the oil shortage emerged as a new reality after the onset of the first oil shock in 1978. Japan was forced to change from its former energy supply structure, characterized by a continued heavy reliance on oil. It had to pursue energy saving policies and promote the development of energy sources other than oil. In consideration of Japan's energy situation, with its heavy dependence on foreign sources, a Bill Concerning Rational Use of Energy (which became the Energy Saving Law) was introduced before the Diet in 1978 for the purpose of promoting the efficient use of fuel resources and implementing measures to ensure the rational use energy in factories, buildings and machinery. The Energy Saving Law was passed in 1979, abolishing the former Heat Control Law. To meet energy demands amid the growing awareness of global environmental problems, the Energy Saving Law, the Law Concerning Promotion of the Development and Introduction of Alternative Energies, the Law on Special Accounts for Measures Concerning Coal, Oil and Alternative Energies, were revised, and the Law on Temporary Measures to Promote the Rational Use of Energy was enacted in 1993 in order to further promote energy saving and recycling and to rationalize the use of designated chlorofluorocarbons (CFCs).

## 11.9 Conventions, Protocols and Associated Laws

In keeping with the globalization of environmental problems, Japan has concluded a number of international conventions and protocols and promulgated domestic laws to implement them. The subsequent sections give an overview and descriptions of the principal conventions, protocols and domestic laws relating to air quality conservation.

### 11.9.1 Overview

The United Nations Conference on the Human Environment, held in Stockholm, Sweden in June 1972, was the first international forum in which Japan took a serious part in the field of environmental administration. Subsequently, Japan played leading roles in the Nairobi Conference held in Kenya in 1982 and in the United Nations Conference on Environment and Development held in Rio de Janeiro, Brazil in 1992.

### 11.9.2 Conventions and Protocols

The Japanese government continued to make a range of efforts to promote international cooperation for the environment after signing the Environmental Protection Cooperation Agreement between the Japanese Government and the Government of the United States of America in 1975. Of these efforts, the signing of the Vienna Convention and Montreal Protocols for the Protection of the Ozone Layer (1988) and the United Nations Framework Convention on Climate Change (1994) that was basically agreed upon at the United Nations Conference on the Environment and Development in Rio de Janeiro, has had a direct bearing on air quality conservation.

### 11.9.3 Promulgation of Domestic Laws

The Japanese government enacted the Law Concerning the Protection of the Ozone Layer by Regulating Certain Substances in 1988 and proceeded with diverse measures such as regulating the production of specified substances (CFC). It is also endeavoring to protect the ozone layer by observing the ozone layer conditions and measuring the atmospheric concentrations of specified substances (CFC). Efforts to cope with climate change associated with global warming are also underway, through the Action Program to Prevent Global Warming, adopted by the Cabinet, and the Law Concerning Rational Use of Energy (Energy Saving Law).

**(11.1~11.9) References**

- 1) Environmental Law Study Group, Environment Agency of Japan, eds., Environmental Laws, Chuo Hoki (1997)
- 2) Planning and Coordination Bureau, Environment Agency of Japan, Environmental Impact Assessment Law (1997)
- 3) Suzuki, Toshio, Introduction to Environmental Laws, Diamond Sha (1997)
- 4) Action Program to Prevent Global Warming, The Industrial Pollution Prevention Association (1990)

## Chapter 12 Administrative Outline of Air Quality Conservation

### 12.1 Air Quality Conservation Administrative Framework and Enforcement System

The administration of air quality conservation in Japan is carried out by competent local governments under the direction of the Air Quality Bureau of the Environment Agency. The Environment Agency Air Quality Bureau, the structure and functions of which are shown in Fig. 12.1.1, consists of 4 divisions and 2 sections under the supervision of the Bureau Director, including the Planning Division, the Air Pollution Control Division, the Motor Vehicle-Related Environment Measures Divisions 1 and 2, the Air Living Environment Section and the Wide Scale Air Quality Control Section, and carries out administrative measures to regulate air pollution, noise, and vibrations, offensive odors, protection of the ozone layer and acid rain.

Also, the Planning and co-ordination Bureau carries out work related to pollution control programs and systems for compensating air pollution victims and preventing air pollution-related health damage and coordinates measures carried out by other government agencies, including research on air pollution control measures, fuel supply improvements, and energy saving policies.

The administration of air quality conservation is apportioned among the national agencies and local governments chiefly as follows. The national agencies establish the national and area-wide standards and promote projects to assist the local governments, and the local governments are responsible for, among other things, carrying out regulations and supervision.

Regarding air pollutant emissions control, according to the Air Pollution Control Law, for example, the national government conducts investigations into the actual conditions of environment and air pollutants in order to establish controlled facilities and emissions standards, and it also examines the state of air pollutant emitting facilities and emission prevention technologies. The local governments are responsible for administrative activities like carrying out notification inspections and competence inspections of smoke and soot emitting sites as well as regular supervision, guidance and orders for site improvements.

Furthermore, regarding regulations of total emission of designated smoke and soot emissions, the national government establishes controlled areas and controlled substances and determines the content of total-volume regulations manuals and pollutant substances measuring technologies, and the local governments conduct numerous technological investigations, determine the total-volume emission regulation standards due to the total-volume emission reduction programs which are the basis of total-volume emission controls, issue improvement orders, and promote the work of enforcing regulation of total emissions.

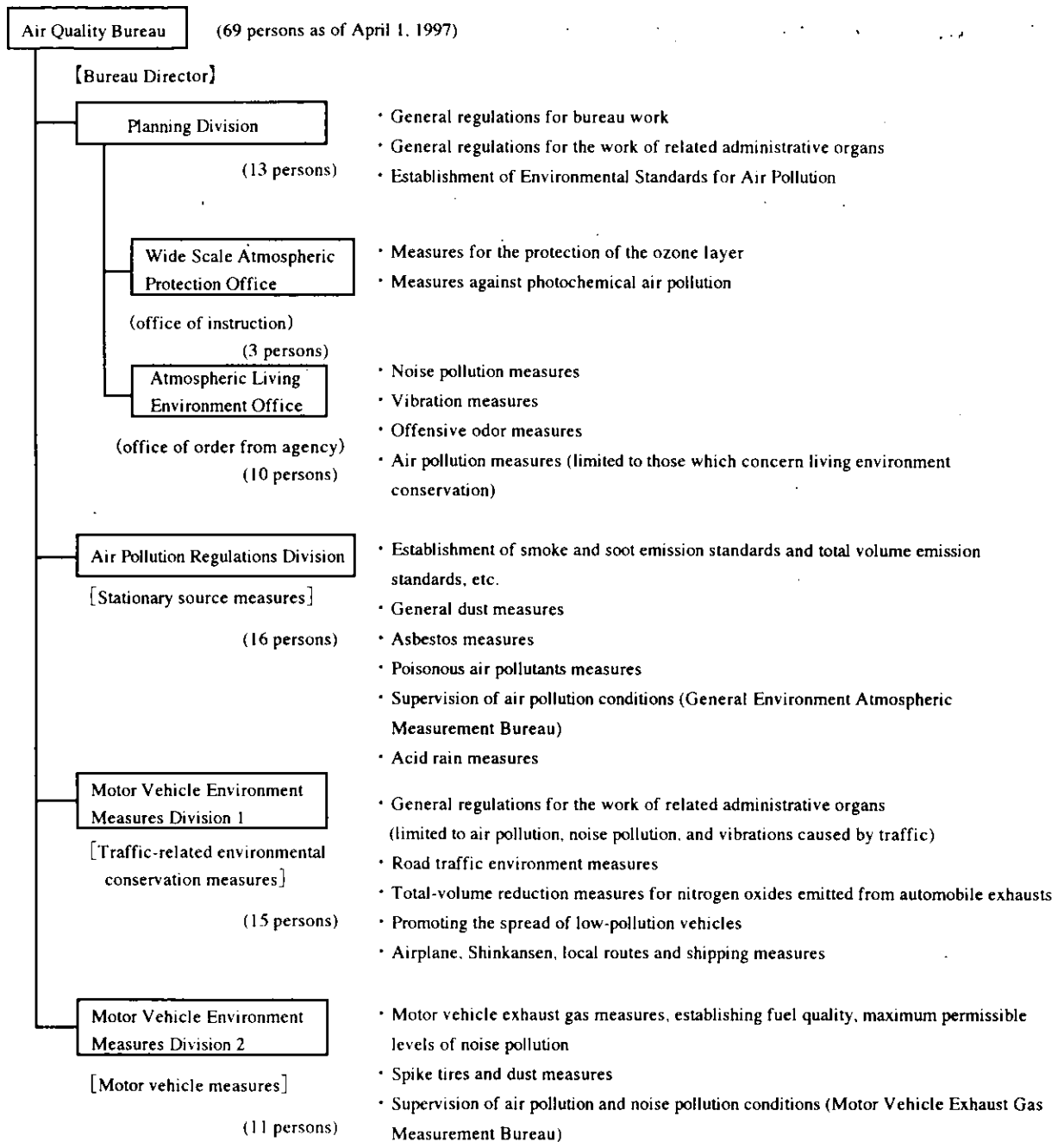
The national government agencies regulate automobile exhaust gases by establishing the maximum permissible levels of automobile exhaust gases and motor vehicle fuel quality, and the local governments measure the concentrations of automobile exhaust gases in the vicinity of roads and issue requests and opinions concerning necessary particulars to the relevant administrative organs based upon their findings. Moreover, local governments are to maintain and operate the air pollution monitoring systems established due to the Air Pollution Control Law, and the data from these monitoring systems on air pollution conditions is gathered nationally by the Air Quality

Conservation Bureau, which aims to spread knowledge about this data by compiling the “Environment White Paper” and the “Report on the Measurement Results of the Air Quality Monitoring Station”.

Furthermore, in regard to the Special Measures Law concerning the reduction in total-volume emissions in areas designated for automobile emissions of nitrogen oxides, the national government formulates the basic policies for the reduction of total-volume emissions, designates the controlled areas, and establishes the specified motor vehicle emissions standards, and the local governments set up concrete plans to carry out total-volume emission reductions based on these basic policies for the reduction of total-volume emissions.

The Air Quality Conservation Bureau also establishes the standards and names the offensive odorous substances according to the Offensive Odor Control Law and designates the measurement methods for that law.

Besides these undertakings, the Air Quality Conservation Bureau also establishes the environmental quality standards (related to air quality) according to the Basic Environment Law, compiles measurement guidelines and administrative manuals, executes pollution control programs, and works positively to promote administrative activities related to international cooperation and the protection of the ozone layer and other related global environment issues.



\* Air Quality Conservation Bureau Related Laws

- Air Pollution Control Law
- Law Regarding Special Measures for the Total-Volume Reduction of Nitrogen Oxides Emissions From Motor Vehicles in Specified Areas
- Spike Tire and Dust Emission Control Law
- Law Regarding the Regulation of Specified Substances For the Protection of the Ozone Layer
- Noise Control Law
- Vibration Control Law
- Offensive Odor Control Law

Fig.12.1.1 Environment Agency Air Quality Conservation Bureau Structure and Related Laws



## 12.2 Atmospheric Environment Monitoring System

### 12.2.1 Overview

According to Article 22 of the Air Pollution Control Law, continuous monitoring of air pollution conditions is the responsibility of the local government. The local government bodies maintain and operate continuous air pollution monitoring systems with the aim of getting a grasp on the conditions of environmental atmospheric pollution and promoting measures to control air pollution.

The concrete aims in monitoring air pollution are: (1) for the local government bodies to get a grasp on pollution conditions throughout their regions, make the enforcement of the emergency measures provided for in Article 23 of the Air Pollution Control Law run more smoothly, be of more help in judging the suitability of environmental standards and be able to apply this to judging the necessity of strengthening regulation effects and regulation standards, and (2) to promote the environmental impact assessment system, to set regional pollution control plans and total volume emissions reductions and apply knowledge about environmental data.

### 12.2.2 Information Flow in the Atmospheric Environment Monitoring System

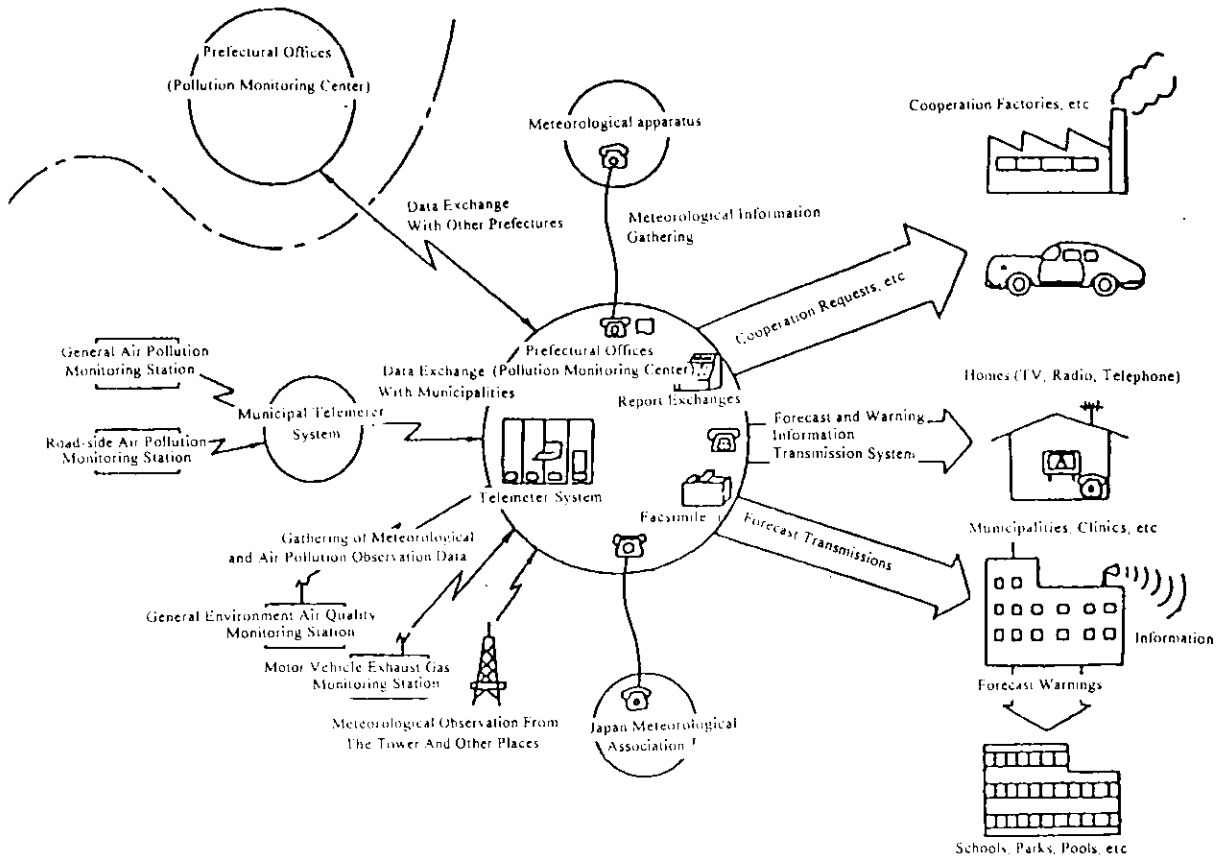
The information flow in the atmospheric environment monitoring system is laid out in Fig.12.2.1.

In this way, with the telemeter system, the measurement result data on the concentrations of air pollutants and meteorological elements is sent in real time to the monitoring centers run and operated by the local government bodies, there managed by computers and then dispatched to the nearby local government bodies where it is used for emergency measures. The telemeter system is also set up to exchange data with the meteorological agency.

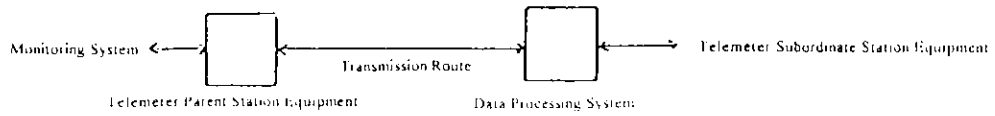
### 12.2.3 Atmospheric Environment Monitoring System Applications

#### (1) Environmental Quality Standards and Data Evaluation

Whenever measurement figures which are judged to indicate a condition of abnormality exist after the actual air pollution conditions and hourly fluctuations in concentration levels in the area being monitored are considered, the maintenance and control of the air pollution measuring apparatus, meteorological conditions and the conditions of air pollution sources are examined carefully. In this case, if it is recognized that the abnormal measurement figures exist as a result of a malfunction in the maintenance and operation of the measuring apparatus and that the regional air pollution conditions are not being accurately reflected in the apparatus, then the measurement figures are of course excluded from the figures to be assessed. Further, whenever evaluations are made on a daily basis, the average hourly measurement data are excluded from the figures to be evaluated if they are the lack of a period of over 4 hours in a 24-hour period. Also, in long-term assessments, data from the monitoring stations which do not add up to a 6,000-hour period per year is excluded from the evaluation. Environmental quality standards necessary for both short-term and long-term evaluations are listed in Table 12.2.1.



The Information Flow in the Atmospheric Environment Continuous Monitoring System



The Data Transmission System Diagram

Fig.12.2.1 The Information Flow and the Data Transmission System Diagram in the Atmospheric Environment Continuous Monitoring System

Table 12.2.1 Short-Term and Long-Term Evaluations for Environmental Quality Standards

Substances	Short-Term Evaluation (evaluations compared to the below environmental standards)	Long-Term Assessment
Sulfur Dioxide	One-hour mean value for a day should be 0.04 ppm or less, and one hour value should be 0.1 ppm or less	Evaluated by removing 2% from the highest daily average value However, when there are two days or more when the environmental standard is exceeded consecutively, it will be regarded as non conformity.
Nitrogen Dioxide	One-hour mean value for a day should be within the zone between 0.04 ppm and 0.06 ppm or less	Evaluated by removing 98% from the lowest daily average value However, when there are two days or more when the environmental standard is exceeded consecutively, it will be regarded as non conformity.
Carbon Monoxide	One-hour mean value for a day should be 10 ppm or less, and one-hour mean value for 8-hour should be 20 ppm or less	Same as SO <sub>2</sub>
Suspended Particulate Matter	One-hour mean value for a day should be 0.10 mg/m <sup>3</sup> or less and one hour value should be 0.20 mg/m <sup>3</sup> or less	Same as SO <sub>2</sub>
Photochemical Oxidants	One hour value should be 0.06 ppm or less	None

(2) Measures for Coping with Air Pollution Emergencies

Whenever there is a marked increase in air pollution or any danger that human health or living environment may be adversely affected, or whenever any condition arises that has been provided for by Cabinet Order, Article 23 of the Air Pollution Control Law provides that prefectural governors must inform the general public of the actual conditions and ask for the cooperation of smoke and soot emitting facilities or motor vehicle operators and drivers to reduce smoke and soot emissions and/or exercise independent restraint in operating motor vehicles. Furthermore, whenever pollution worsens and a situation of grave emergency, which is defined in Article 23 of the Air Pollution Control Law, that same law allows the local governments to order necessary measures for the reduction of smoke and soot concentrations in the atmosphere and for the restricted operation of smoke and soot emitting facilities. Penal regulations are also provided for in the case of violations of these orders. Conditions for the official announcement of such states of emergency are set down in Table 12.2.2, and the atmospheric environment monitoring system is also utilized in these official announcements. Furthermore, in such cases of emergency, the technological contents like the scale, reductions, and changes in operation of smoke and soot emitting facilities need to be examined and practical agreements need to be reached.

Table 12.2.2 Air Pollution Emergency Conditions

Substances	General Emergencies	Critical Emergencies
Sulfur Oxides	Over 0.2 ppm for 3 continuous hours Over 0.3 ppm for 2 continuous hours, Over 0.5 ppm, 48-hour average value of over 0.15 ppm	0.5 ppm for 3 continuous hours 0.7 ppm for 2 continuous hours
Suspended Particulate Matter	2 mg/m <sup>3</sup> for 2 continuous hours	3 mg/m <sup>3</sup> for 3 continuous hours
Carbon Monoxide	Over 30 ppm	Over 50 ppm
Nitrogen Dioxide	Over 0.5 ppm	Over 1 ppm
Oxidants	Over 0.12 ppm	Over 0.4 ppm

(All measurement values are based on hourly units)

### (3) Other Applications

Data from the air pollution monitoring system is important in judging the effects of reductions in the emissions of pollutants provided for in emission source regulations. In other words, it is an important issue for the administrative judgment to compare air pollution monitoring system data with the environmental quality standards it aims for and apply this to judging the necessity of strengthening regulations on the air pollutant emission levels of emission sources to ascertain the advisability of attaining such standard values.

Also, highly reliable, long-term atmospheric environment data is essential for implementing environmental impact assessment. In other words, the local governments can, based on this principle data, consider the increases in air pollutant emission levels which accompany new development projects and, by estimating their effect on the atmospheric environment, evaluate such new development projects and examine measures to cope with them.

In establishing pollution control programs, based on current air pollution monitoring data, the local governments consider the new factories, development areas, and roads planned for the region and plans necessary measures based on estimates of the air pollution conditions of the area in the future. The diffusion simulation model making is necessary for arriving at such estimates, but air pollution monitoring data is also essential because the calculated values need to compare with actual measurement values.

## 12.3 Factory Inspections

### 12.3.1 Overview

Article 26 of the Air Pollution Control Law requires smoke and soot emitting facilities to submit reports to the prefectural governor and also allows prefectural governors to conduct inspections of smoke and soot emitting facilities.

More precisely, necessary restrictions to enforce the Air Pollution Control Law provided to the prefectural governor in the same Article allow the governor to send his officials into factories and business establishments and conduct inspections of smoke and soot emitting facilities and smoke and soot controlling facilities. The enforcement regulations set forth in the Air Pollution Control Law are powers granted to the prefectural governors in order to guarantee that they are able to carry out such inspections, and competence inspections are conducted strictly as administrative measures necessary to see that this law is enforced. Judicial police authority to conduct criminal investigations is not recognized. Authority to conduct inspections is also granted in certain cases to municipal mayors, by the Cabinet Order set forth in Article 31 of the Air Pollution Control Law.

### 12.3.2 Items Targeted for Inspection

Items targeted for such inspections are listed as follows:

- ① Inspections, in the form of related reports requested from those who have established smoke and soot emitting facilities, of boilers, heating furnaces and other smoke and soot emitting facilities, dust collector, exhaust gas desulfurization equipment and other smoke and soot treatment facilities, and other related facilities, and inspections of fuel, raw materials and related account books utilized at the smoke and soot emitting facilities.
- ② Inspections, in the form of reports on the structure, operations, and management of the general dust emitting facilities requested from those who have established general dust emitting facilities, of the general dust emitting facilities and other related facilities and related account books.
- ③ Inspections, in the form of reports on the operations, management methods, dust scattering control methods, etc. used in specified dust emitting facilities requested from those who have established specified dust emitting facilities, of specified dust emitting and other related facilities and the raw materials and related account books used at specified dust emitting facilities.
- ④ Inspections, in the form of reports on accidents and measures to cope with accidents at specified facilities requested from those who have established specified facilities, of the specified facilities, other related facilities and related account books.

Furthermore, officials who carry out the above inspections must carry with them the form of personal identification shown in Fig. 12.3.1 and produce this to the competent persons.

Front

12 centimeters

No. \_\_\_\_\_

Personal Identification Card Stipulated in Article 26, Paragraph 2 of the Air Pollution Control Law

Job Title or Name

Date of Birth: (y/m/d)

Issue Date: (y/m/d)

Prefectural Governor / Mayor

Seal/Signature

8 centimeters

Back

**Air Pollution Control Law Excerpt**

Article 26: The local governments may, to the extent necessary for the implementation of this law and in accordance with the provisions of the Cabinet Order, require a smoke and soot emitting facility, an owner/operator of a specified facility, a person installing a general dust discharging facility or a specified dust emitting facility to report necessary matters, such as the conditions surrounding accidents at the specified facility, conditions of the general dust emitting facility, or conditions of the specified dust emitting facility, or he may have a competent official or officials of the prefectural government conduct an on-site inspection of an industrial plant or smoke and soot emitting business establishment, a specified facility or a general dust or specified dust emitting facility respectively to examine the said smoke and soot emitting facility, smoke and soot treating facility, specified facility, general dust or specified dust emitting facility or other related equipment.

2. The prefectural government official who conducts the inspection, under the provisions of the preceding paragraph, shall bring a personal identification card with him and present it to the personnel concerned in the industrial plant or business establishment.
3. Authority for on-site inspections, provided for in Paragraph 1, shall not be interpreted as that which has been recognized for the purposes of a criminal investigation.

Article 31: Administrative duties belonging to the governor of the prefecture, under the provisions of this law, may be delegated to the mayors of municipalities designated by the Cabinet Order in accordance with the provisions of the Cabinet Order.

Article 35: Any person who falls under any one of the following items shall be subject to a fine of not more than 100,000 yen.

3. Any person who fails to submit a report, under the provisions of Article 26, Paragraph 1, or who has filed a false report, or who refuses, obstructs or evades an inspection conducted under the provisions of the same paragraph.

Fig.12.3.1 Personal Identification Card Stipulated in Article 26, Paragraph 2 of the Air Pollution Control Law

### 12.3.3 Inspections

Inspections are normally carried out by two officials. These officials first gain an understanding of the general overview of the factory by confirming the manufacturing process and work contents, from the introduction of raw materials to the finished product, from the flow sheet. Then, on-site inspections are conducted on the basis of reports submitted by those who established the smoke and soot emitting facility. Items which need to be confirmed upon inspection are listed as follows:

- ① In the case of a facility targeted for inspection which contains boilers and other furnaces used in factories, the items to be inspected include the types, compositions, and volumes of use of fuel and raw materials, the types, constructions, sizes and abilities of furnaces, the organization, abilities, exhaust gas levels, concentrations of air pollutants within the exhaust gases from burners, the compositions of exhaust gases, the quality of materials used to build the exhaust stacks, stack heights, and in the case of exhaust gas treatment equipment, the treatment methods, the concentrations of air pollutants contained within the inlet pipes, the concentrations of air pollutants contained within the outlet pipes, the rate of air pollutant eliminations, waste treatment methods, etc. Moreover, in order to inspect the air pollutant emissions standards and the degree of conformity with fuel-use standards, inspectors need to measure the sulfur oxides, nitrogen oxides and smoke and soot concentrations within the exhaust gases and gather samples for fuel inspections. Inspecting officials do their utmost to ensure safety while these measurements are being carried out.
- ② In the case of a facility which is conducting work related to chemical reactions like the combination and disintegration of chemical substances, items to be inspected include the structures of the reaction vessels, reaction framework (reaction methods), reaction temperatures and pressures, the concentrations in the inlet and outlet pipes of poisonous gas treatment equipment, and the waste treatment methods.
- ③ In factories targeted for total emissions control, which have apparatus for measuring the concentrations of air pollutants in exhaust gases, these measurement figures are confirmed. Also, measurements are confirmed in the same way when the similar steps are required by conventions or agreements.
- ④ Inspectors listen to explanations regarding measures for times of accident or emergency, measures and policies to cope with and ensure safety within the factory, and other systems to enforce these measures and confirm the overall pollution control structure.

## 12.4 Environmental Impact Assessment System

### 12.4.1 The Process of Establishing the Environment Impact Assessment System

The “Environmental Impact Assessment System” is a system whereby project proponents investigate, estimate and assess the possible effects on the environment of their development projects which may have significant impacts on the environment before they implement the projects and make appropriate considerations concerning environmental conservation based on these results of the assessment. This system is a very important measure to prevent environmental destruction before it occurs and to build a sustainable society, and ever since it became systematized in the United States of America in 1969, it has become progressively systematized in various countries all over the world.

In the process of putting this system into effect in Japan, a bill was submitted in 1981, after the Cabinet agreement in 1967 on “Mitigation Measures Concerning the Various Public Works”, but the bill was rejected in 1983. After that, the environmental impact assessment system has been operated on the basis of the “Implementation Scheme For Implementing Environmental Impact Assessment” (hereafter referred to as the “The 1984 Cabinet Decision Guideline”) which was fixed upon in 1984, individual laws like the Public Water Body Reclamation Law, individual administrative guidances like MITI’s departmental meeting decisions regarding the sites for power stations, and local governments’ ordinances and guidelines. The enactment of the Environment Basic Law in 1993 provided the opportunity to begin to reassess the system and after deliberations by the Central Council for Environmental Pollution Control, the Environmental Impact Assessment Bill was drawn up by Cabinet Decision in March 1997 and was submitted to the 140th session of the Diet where it was passed on June 9th and put into effect as Law No.81.

### 12.4.2 Environmental Impact Assessment Law and An Overview of the System

Article 20 of the Environment Basic Law requires the national government to take measures toward the environmental impact assessment (hereafter referred to as EIA), and the Environmental Impact Assessment Law stipulates concrete procedures based on this requirement.

Chapter 1 contains the general provisions of the law. Chapter 1, “the Aims”, stipulates that this law aims to recognize the importance of the environment impact assessment system, identify the responsibilities of the national government for the smooth running of the system, make proper considerations on environmental conservation by reflecting results of EIA, and contribute to securing healthy, cultural lives for the people. In this way, with the enactment of this law, the operation of the system, according to the administrative guidance of the Cabinet Decision Guidelines, was put into practice with authority and with highly reliable rules. Article 2, “the Definition”, defines EIA as project proponents’ actions to investigate, estimate and assess the possible environmental impacts caused by the proposed projects, according to the items which are related to environmental elements, identify mitigation measures during the process, and assess the environmental impacts comprehensively if the mitigation measures are implemented. This law defines those targeted projects as “projects which are required



to enter procedures for EIA". Further, it introduces a system of screening whereby the necessity of carrying out assessment is decided on an individual basis, classifying the target projects into "Class-1 Projects" which are of a certain size or larger and must carry out EIA, and "Class-2 Projects" which are smaller in scale than these. In other words, "Class-2 Projects" are judged necessary to carry out EIA and they are considered target projects. The details of "Class-1 and Class-2 Projects" are set down concretely by the government ordinances. Power stations were also included among those targeted projects by law and the screening system was renewed. Article 3, "the Responsibilities of the National Government and Others", stipulates that the national government, local governments, project proponents and the people should recognize the importance of EIA and make appropriate considerations so that the procedures can be implemented smoothly and the environmental burden due to the execution of the projects can be avoided or decreased. In Chapter 2, which sets down the procedures which must be carried out before the preparation of a draft of the environmental impact statements (hereafter referred to as Draft EIS), Article 4 defines the screening process for environmental impact assessment (hereafter referred to as EIS), and Articles 5-10 stipulate the procedures for preparing Scoping Documents for EIA study. More precisely, project proponents prepare Scoping Documents for EIA study regarding the methodology of investigations, estimates, and assessments of the target project and send them to the prefectural governors and municipal mayors. Furthermore, the Scoping Documents for EIA study are also published and offered for public review, and the public opinions on these documents are compiled and sent again to the governors and mayors. Then the prefectural governors, after listening to the opinions from the mayors, submit their opinions to the project proponents. These procedures, which have been newly introduced, are called "scoping". Articles 11-13 concern the implementation of EIA and stipulate that the project proponents select the items of EIA study and methodology of investigations, estimates and assessments, based on the above opinions, and that they may receive technological support and assistance, where necessary, from the Competent Ministers. Also, the basic items for these guidelines are defined by the Director General of the Environment Agency, and the Competent Ministers meet with the Director General to establish regulations of Ministries.

In Chapter 3, "the Draft EIS", Articles 14-20 concern the procedures regarding preparation of Draft EISs and stipulate that the project proponent must prepare a Draft EIS, send it to the prefectural governors and municipal mayors, publish and circulate it, hold public meetings, gather opinions regarding environmental conservation, and then send a summary of these opinions, along with their views on the opinions, to the governors and mayors. Then the governors, after hearing the views of the mayors, inform the project proponents of their views on the content of the Draft EIS. The previous restriction, by the Cabinet Decision Guidelines, of those who may submit their opinions to only those local residents who are affected has been removed, and the chances for residents to submit their views have been augmented. In Chapter 4, "the EIS", Articles 21-27 regulate the procedures regarding preparing the EISs. More precisely, the project proponent, based on the above procedures, prepares an EIS and transmits it to the licensing authority in charge. When necessary, the Director General of the Environment Agency may also submit his opinions to the licensing authority in charge, and based on these procedures, the licensing authority then submit its views to the project proponent. Then, the project proponent reexamines the projects, revises the EIS, issues the final EISs and offers it for public review. Chapter 5 concerns the revisions on the contents of the target projects. Precisely, Articles 28-30 stipulate the revisions of the contents of the target projects and so on, and point out the

necessity for those project proponents, except for the projects whose revisions are minor or which do not fall under the categories designated by the government ordinances, to redo the entire procedure, if they need to revise the contents of their projects, during the period from their public notice regarding Scoping Documents for EIA study to their public notice regarding the Final EIS. Chapter 6 concerns the procedures for after the public notices and circulation of the EISs, and Article 31 forbids the target project proponents to take any actions before the Final EIS is issued and published. Article 32 regulates the re-implementation of EIA after the Final EIS is published, and Articles 33-37 concern the environmental review relating to the approvals and permissions of the proposed projects. It is stipulated that the licensing authorities may conduct environmental reviews, based on the EISs and the opinions toward the EISs, and they may refuse to grant approvals and permissions or append conditions to the granting of them. Article 38 stipulates the considerations which the project proponents must make concerning environmental conservation. In Chapter 7, "the Special Provisions for EIA", Articles 39-46 describe the special cases where those with the power to make city planning decisions may conduct the assessments in place of the project proponents, and Articles 47-48 describe the special cases regarding port and bay area planning. In Chapter 8, "Miscellaneous", Articles 49-50 stipulate communication with local governments, Article 51 concerns technological development, Article 52 stipulates exclusions, Articles 53-56 stipulate procedural measures, Article 57 stipulates matters which are entrusted to the government ordinances, Article 58 concern regulations regarding the Competent Ministers and Article 59 concerns the regulations regarding power stations. Articles 60-61 concerns the relationship with local governments and it stipulates that this law does not interfere with the assessment procedures for those project proponents which do not fall under the categories of the target projects or with local governments' ordinances which regulate the procedures to be conducted in the local governments, which are stipulated in this law. Finally, this law is considered to be effective for a two-year period starting from June 13, 1997.

The flow chart of the EIS process is shown in Fig. 12.4.1. In the operation of this system, attention will be paid to those local governments' ordinances and guidelines.

#### 12.4.3 Assessment concerning Air Quality Conservation

This section generally discusses EIS regarding air quality conservation because the Environment Impact Assessment Law has not yet been put into force. There are many types of pollutants which cause air pollution, but in the EIA of individual projects, according to the contents of the target project, pollutants to be targeted for assessment are selected according to the types and levels of effect of those pollutants which can be emitted in the course of project operations, and considered as items to be investigated, estimated and assessed. Generally, according to the characteristics of those target projects, those substances which are specified by environmental standards, such as sulfur oxide, nitrogen oxide, carbon monoxide and suspended particulate matter, are selected. In environment impact assessment air pollution becomes the target of investigations, estimates and assessments because the atmospheric concentrations of pollutants increase and have a harmful effect on the environment due to the air pollutants being emitted by target projects in the course of their operations. Air pollutants have extremely wide-ranging effects on the environment. They cause not only damage to people's health but also plant and crop failure and corrosion damage to buildings, and smoke, soot, dust, and offensive odors which interfere with people's

daily living environments.

#### (1) Investigations

Air quality investigations (present condition investigations) for environment impact assessment aim to obtain the acquisition of necessary data to recognize the present conditions of air pollution and estimate and assess future air pollution in areas which are thought to be affected by the operations of target projects. When the atmosphere has become polluted, the first item investigated is the present concentrations of air pollutants. Other items selected for investigation are those necessary for atmospheric diffusion estimates such as meteorological and topographical statistics, data on natural objects and landmarks, land use, the situations with nearby sources of air pollutants and other data necessary in considering the related laws and ordinances.

For example, sulfur oxides, nitrogen oxides, smoke and soot would be chosen as the types of air pollutants which are related to the fuel burning process. Also, in addition to the aforementioned substances, hydrogen chloride, mercury, lead and other heavy metals would be chosen according to the properties of incinerated waste matter in the waste incineration process.

#### (2) Estimates

Air pollution estimate items are chosen from among those air pollutants considered items for investigation in current condition investigations, with reference to the estimated figures for total air pollutant emission levels, and they are necessary for estimating and considering the level of environment burden caused by the target projects. Other items examined include the possible causes for the impact and the distribution of the impact. Technological guidelines provide many different types of such examples. Sometimes basic estimate items are listed according to the type of target enterprise and other times conditions are indicated for the selection of estimate items. Based on this type of idea, if you look at examples of environment impact assessment and pay attention to what sorts of items were selected as a result, you can see that the items chosen for estimates are the majority of the time, those which are stipulated by general environmental standards, namely, nitrogen oxide, and after that, sulfur oxide, carbon monoxide, dust, and suspended particulate matter.

Pollution estimate methods in environment impact assessment are broadly classified into quantitative methods to estimate air pollutant concentration by methods like diffusion calculations and qualitative methods which do not make quantitative estimates of the concentrations but refer to analogous examples to make qualitative estimates.

##### (Quantitative Methods)

Quantitative methods include (1) diffusion calculation methods, (2) statistical methods, (3) wind-tunnel experiment methods, and (4) outdoor experiment methods. Sometimes two methods may be combined, but in this case, other methods may be adopted to perfect normal diffusion calculations.

##### (Qualitative Methods)

Qualitative methods include (1) analogical methods using similar examples, (2) pollutant emission level examination methods, (3) environmental conservation measure examination methods, and (4) meteorological factor (wind speed, wind direction) examination methods.

### (3) Assessments

Assessments are usually conducted on items to be estimated on the basis of the scope, period and time. When quantitative estimates are not made, assessments are based on qualitative estimates.

Environment impact in Japan are conducted by identifying mitigation measures for expected environmental impact and indicating project proponents views. When quantitative estimates are made, projects proponents indicate their views on the expected environmental impacts while referring to the uniform national environmental standards, the guideline figures and guidelines set down by the government, and the target figures and guideline figures, based on ordinances and plans set down by local governments reflecting particulate local characteristics.

When there are no administrative target figures, projects proponents indicate their views by establishing environmental target figures based on scientific information regarding pollutant concentrations in the environment, people's health and living environments, and vegetation.

When emission standards and total-volume regulations based on the Air Pollution Control Law and other ordinances are being put into effect, assessments are conducted by first ascertaining whether estimated pollutant emission levels have reached the standards level.

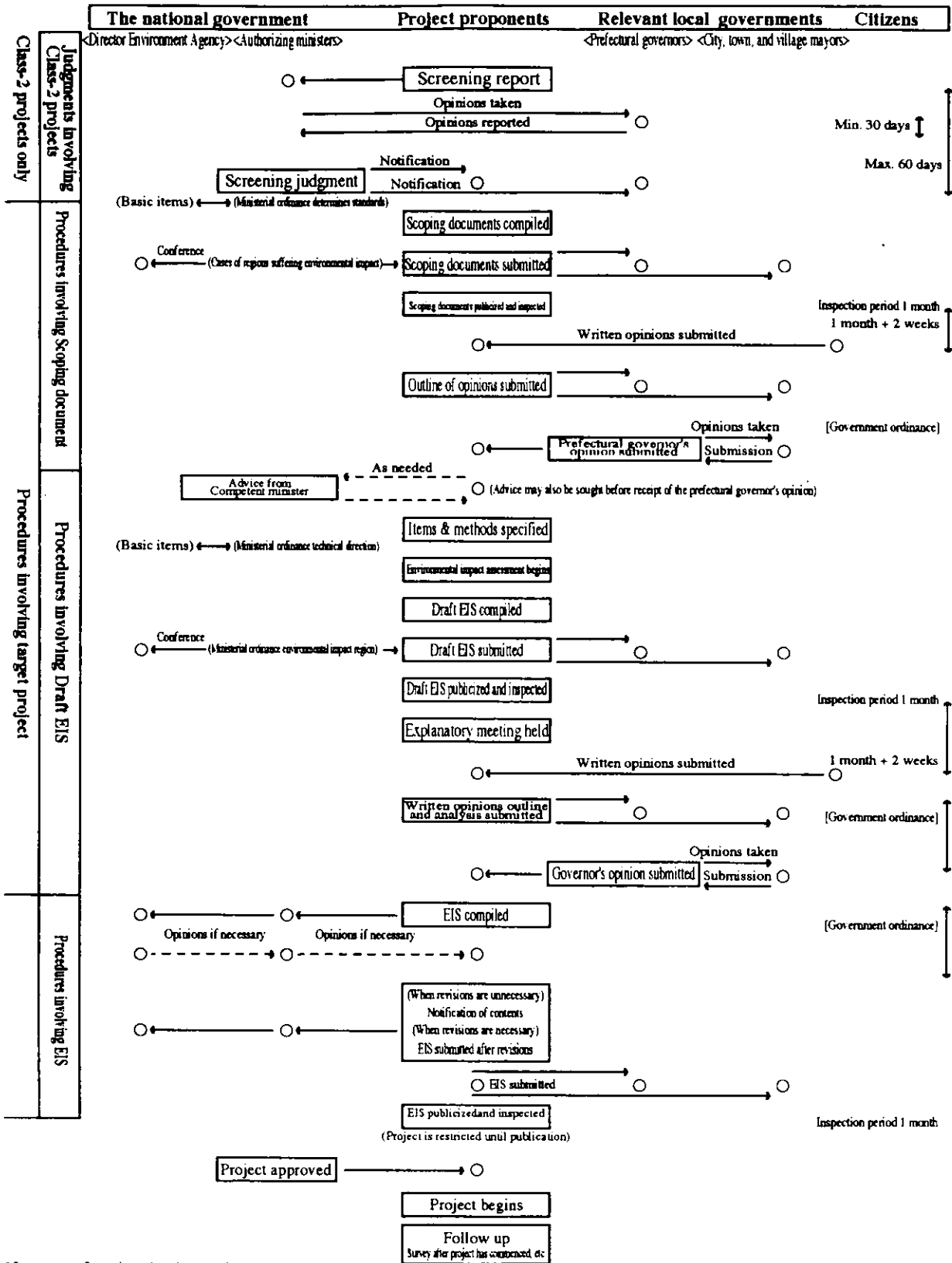
Conditions applied to assessment methods, as shown in technological guidelines, show that, in principle, it is normal to conduct assessments by making comparisons with environmental conservation target figures, and items which have set environmental standards make these environmental standards their environmental conservation targets. Also, when quantitative estimates are difficult to make and for items which have no set environmental standards, many technological guidelines set target figures by referring to qualitative targets and scientific information.

### (4) Follow-Up Investigations

In the environmental impact assessment system, estimates and assessments are generally conducted for project plans drawn up by the projects proponents, but after these plans are already initiated, various fluctuations may be possible in external factors.

For example, as for related facilities, planned transportation levels and moving speeds may change due to changes in the social and economic conditions before the annual year which the estimate is set for, or there may be some uncertainties left in the accuracy or set conditions of estimation methods in the estimation technology for the current environmental impact assessments.

Here, fellow-up investigations are to be conducted regarding environmental impact and project operating circumstances, even after project operations have already commenced. In this case, additional measures can be taken according to these results. Moreover, continuous monitoring of environmental load and impact can be implemented, and if analysis is possible after the accumulation of this data, it may be possible to further add to the scientific knowledge which will become the foundation for environmental impact assessment.



If nature of project is changed  
 [Slight changes... Conduct additional surveys as needed, continue procedures.  
 Major changes to project content... Return to preparation of Scoping documents and reimplement procedures.]

Fig.12.4.1 Procedures for the Environment Impact Assessment Law

## 12.5 System of Pollution Control Manager, etc.

### 12.5.1 Background Circumstances

The 64th session of the Diet in 1970 made some headway in the legal preparations for establishing the Air Pollution Control Law, Water Pollution Control Law, and Noise pollution regulations, and regulations regarding pollution were greatly expanded and strengthened. In order to translate these strengthened laws into sound practice, it became necessary to implement a suitable pollution control system on the side of the enterprises as well. In order to do this, those factories which were the sources of industrial pollution were required to set up pollution control systems, and with the aim of making enterprises set up pollution control systems, the "Law Regarding the Provision of Pollution Control Systems in Specified Factories" was enacted in 1971.

### 12.5.2 Specified Factories

Specified factories, which are the object of this law, include (1) manufacturing industries (including commodities processing industries), (2) electricity utility industries, (3) gas utility industries, and (4) heat utility industries, and specified factories related to air pollution are those which emit over a certain amount of air pollutants such as hazardous substances (cadmium and its compounds, chloride and hydrogen chloride, fluorine, hydrogen fluoride, silicon fluoride, lead and its compounds, and nitrogen compounds), sulfuric compounds, soot and dust, general dust, and specified dust.

Also, these specified factories must also set up pollution control systems including the pollution control managers (those with the proper qualifications) and supervisors of pollution control to oversee and manage these systems. However, those enterprises which have less than 20 employees are not required to provide these managers. (Enterprises over a certain size are required to have chief managers in charge of pollution control.)

### 12.5.3 Qualification Acquisition

There are 4 separate classifications from class 1 to class 4 for managers in charge of air quality-related pollution control, and managers in charge of dust-related pollution control are classified into general managers and specified managers. In order to acquire these qualifications, candidates must either pass the national examinations which are held once a year or must be specified by the school career or by the qualifications and also have completed a short-term course for accreditation conducted by the related Ministries.

### 12.5.4 Job Specifications

#### (1) Supervisor of Pollution Control

The jobs of supervisors of pollution control are to take necessary measures for the adequate and smooth implementation of pollution control operations and to supervise the conditions of the implementation. Specific

duties required of supervisor of pollution control are as follows:

#### Factories Which Have Smoke and Soot Emitting Facilities

- ① Supervising the operations of smoke and soot emitting facilities, and maintaining and running smoke and soot treatment facilities and other attached facilities
- ② Monitoring and recording the concentrations and emission volume of smoke and soot within exhaust gas
- ③ Setting up measures for accidents and emergencies at specified facilities

#### Specified Particulate Dust Emitting Facilities

- ① Supervising the operations of specified particulate dust emitting facilities, and maintaining facilities to manage the scattering of emissions from these facilities and other attached facilities
- ② Monitoring and recording the concentrations of specified particulate dust in the atmosphere around the boundaries of specified factory sites

#### General particulate Dust Emitting Facilities

- (1) Supervising the operations of general particulate dust emitting facilities, and maintaining and running facilities to manage the scattering of emissions from these facilities and other attached facilities

#### (2) Jobs of Pollution Control Managers

The jobs of pollution control managers, who take directions from the supervisor of pollution control, are to cope with the expert skill side of pollution control activities at the factory. Specific duties required of pollution control managers are as follows:

#### Factories Which Have Smoke and Soot Emitting Facilities

- ① Inspecting use of fuel and raw materials
- ② Inspecting smoke and soot emitting facilities
- ③ Operating, inspecting and maintaining treatment facilities and other facilities attached to specified smoke and soot emitting facilities
- ④ Monitoring and recording the concentrations and emission volume of specified smoke and soot in exhaust gas
- ⑤ Inspecting and maintaining of monitoring apparatus
- ⑥ Implementing emergency measures in case of accidents at specified factories
- ⑦ Implementing necessary measures and for reducing the smoke and soot volumes and concentrations, and for restricting the use of smoke and soot emitting facilities, and other measures in case of emergency

#### Factories Which Have Specified Dust Emitting Facilities

- ① Inspecting raw materials
- ② Inspecting of specified dust emitting facilities
- ③ Operating, inspecting and maintaining facilities for the treatment of emissions and the scattering of specified

dust from specified dust emitting facilities and other attached facilities

- ④ Monitoring and recording the concentrations of specified dust
- ⑤ Inspecting and maintaining monitoring apparatus

**Factories Which Have General Dust Emitting Facilities**

- ① Inspecting use of raw materials
- ② Inspecting general dust emitting facilities
- ③ Operating, inspecting and maintaining facilities for the treatment of emissions and the scattering of general dust from general dust emitting facilities and other attached facilities

**12.5.5 Other Systems Related to Air Quality Conservation**

Other systems for air quality pollution control besides the system of pollution control manager which have been enacted and are in use are shown in Table 12.5.1.

**Table 12.5.1 Other Systems Related to Air Quality Conservation**

Systems	Jurisdiction
(1) Environment Counselor (civil section, business section)	Environment Agency
(2) International Environment Adviser (classified according to fields of specialization)	Ministry of International Trade and Industry
(3) Consulting engineers (environment section)	Science and Technology Agency
(4) Certified Measurer	Ministry of International Trade and Industry
(5) Working environment measurement expert	Ministry of Labor



## 12.6 Supporting System of Pollution Control Measures

### 12.6.1 Governmental Supports for Commercial Pollution Control Investment

With the basic principles of the "polluter pays principle" (PPP), the government has provided various supportive measures such as public, long-term, and low-interest funding, favorable tax measures, technical guidance and technical development to promote pollution reduction and emission. The details are as follows.

#### (1) Funding and Technical Support

The Japan Environment Public Corporation (formerly called the Pollution Control Service Corporation) was established first as a professional organization for the provision of long-term and low-interest funding and technical support. This organization undertakes the construction and transfer of new pollution prevent facilities like green buffer zones and facilities necessary to relocate factories of small and medium-sized enterprises which cause pollution, and it also implements funding programs to provide enterprises with the funds to establish industrial pollution control facilities. Additional systems to finance the creation of pollution control facilities have also been established within the public organizations which have been providing the business funds and equipment investments.

Local government have inaugurated financing systems to compensate for private financial institution to provide funds for pollution control facilities to be established in regional small and medium enterprises. This system fully met the roles of the local government charge of pollution control regulations and has produced great results.

Compared to the private financial institutions' equipment investment funds, these funds offer cheaper and more remunerative terms, with longer term of redemption and lower rates of interest on the level of only 1-2 percent than private funds.

#### (2) Favorable Tax Measures For Pollution Control Measures Expenditures

In order to lighten the burden of pollution control measures expenditures on the management of enterprises and promote pollution control measures, favorable national and local tax measures have been established, such as tax reductions and tax exemptions for the purchasing land necessary to construct pollution control facilities.

##### (a) National Tax

- ① Special redemption system for specified pollution control-related facilities
- ② Tax exemption on reserve funds to promote the use of recycled resources
- ③ Life period curtailment measures for pollution control facilities
- ④ Specific examples of taxation when purchasing specified assets

##### (b) Local Tax

- ① Tax exemption from fixed asset taxes
- ② Tax exemption from special land taxes
- ③ Specific examples of business taxes

A favorable tax system has also been set up to help enterprises when they move from areas densely populated by factories into the suburbs.

### 12.6.2 Effects of Supporting Measures

Japanese enterprises have a very low rate of self-financing and are in an extremely difficult position to invest in unproductive pollution control. Private financial institutions are also unwilling to provide the funds and invest in high risk, low production facilities like pollution control facilities.

Public funds are structured to stimulate the interest of enterprises in investing on their own with the public financing toward their high-risk pollution control facilities and those production facilities with low-pollution and pollution prevention functions. The private financial institutions saw the results of this arrangement and began to act positively as well, and as a result, private funds have come to account for 60-70% of the gross total of investments by enterprises in pollution control.

From these fact, we can see the role and effects of supporting measures in pollution control, as follows:

- (1) Have been effective in inducing private funding of high-risk pollution control facilities.
- (2) Have arisen the interests of enterprises in pollution control measures by both modernizing and rationalizing their production facilities and in implementing pollution control measures at the same time. Technical guidance and information provision on proper pollution control, linked with regulations, was also conducted.

Fig.12.6.1 shows a transition of pollution control investments and Public Funds.

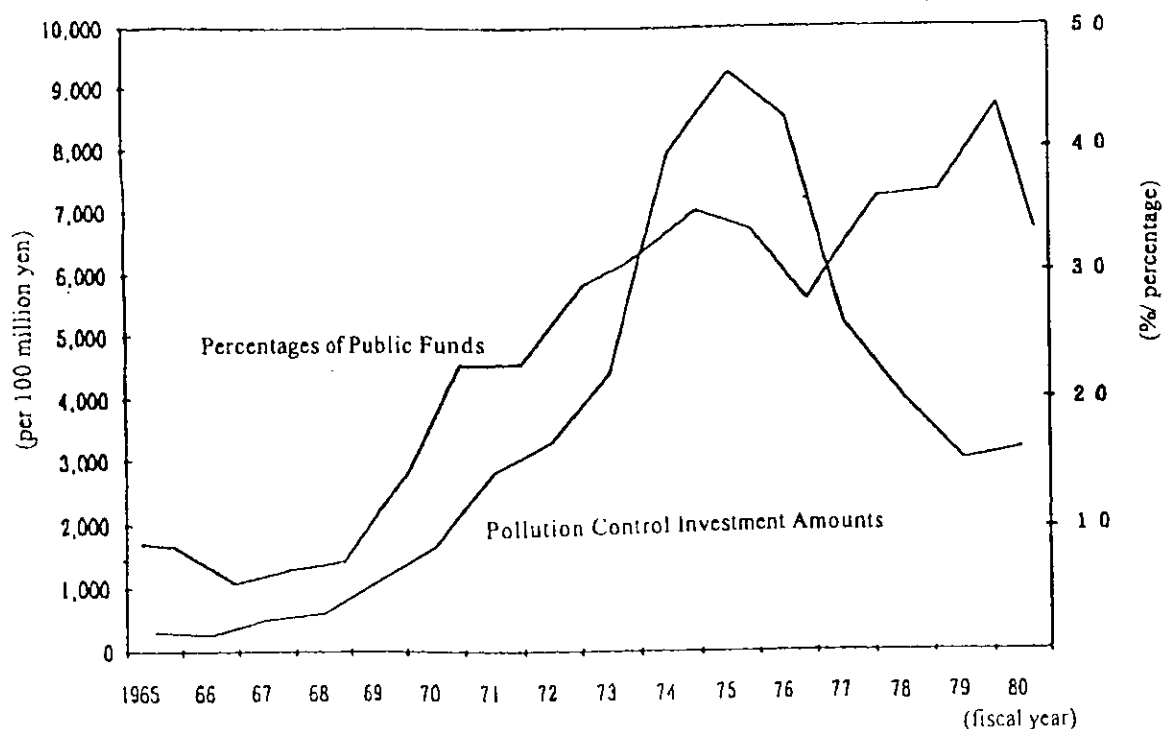


Fig.12.6.1 Transition of Pollution Control Investment and Public Funds

Investment on pollution control facilities also have brought increased demand in the industries which supply these facilities. In other words, pollution control investment has both important factors of reducing and increasing the substantial gross national products.

From these experiences, a supporting system in the environmental cooperation with developing countries are proposed as follows.

- ① A structure for inducing private funding is essential. In Japan's case, almost all public funds are furnished from financial investment and loan. These are funds which cannot be seen in other countries, and developed countries must rely on private institutions for necessary funds.
- ② In addition to providing the pollution control technology which Japan possesses, support to foster environmental industries is essential in order to curtail reduction of the gross national product due to pollution control investments.

One example would be to introduce a credit supplementing system into the two-step loan method for environmental cooperation. This is a structure whereby special pollution control organizations, which have technical guidance and inspection, information provision and credit guarantee functions, are established so that private financial institutions can then provide funds based on the credit guarantees of these organizations.

Fig. 12.6.2 shows pollution control cooperation based on the credit guarantee function.

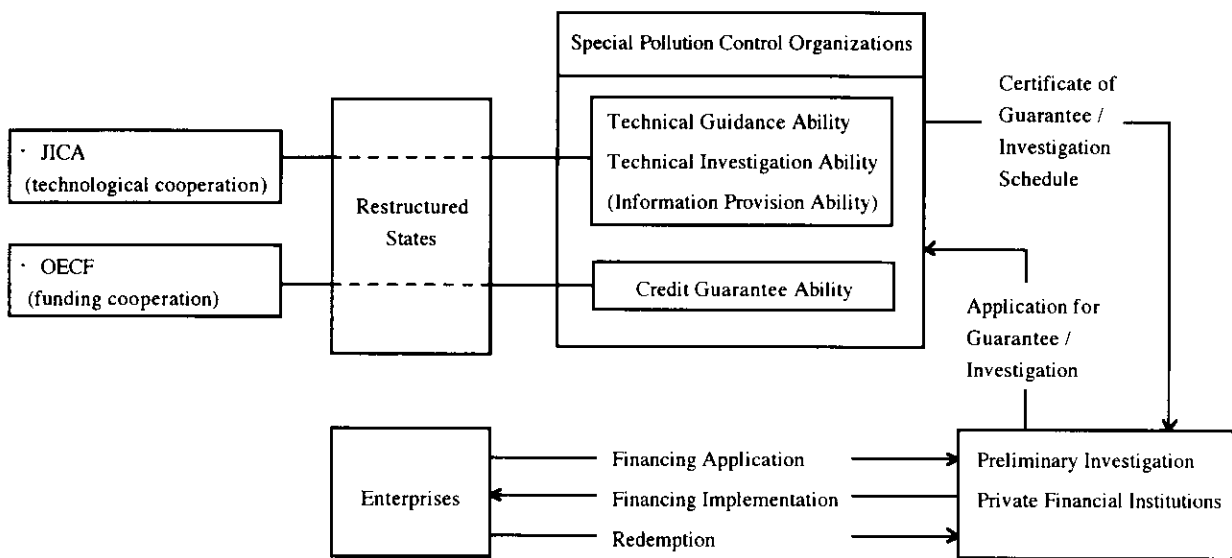


Fig. 12.6.2 Pollution Measures Cooperation in the Form of the Credit Guarantee Function

## 12.7 Systems of Compensation and Prevention for Pollution-Related Health Damage

### 12.7.1 Frequent Occurrence of Health Damage Followed by the Yokkaichi Pollution Lawsuit, and the Establishment of the System for Pollution-Related Health Damage Compensation

Looking back air pollution problems in Japan and counter measures for them, we can not forget the fact that there have been many health damage-related tragedies. From the early stages of the high economic growth period, many residents in industrial cities all over Japan, including Yokkaichi, began suffering from respiratory illnesses thought to have been brought about by air pollution, and the rates of outbreaks in such cities was as much as, and sometimes over, 2 to 3 times that in unpolluted areas. One of the actions taken by residents who suffered from illnesses related to air pollution was the Yokkaichi Lawsuit, which was brought before the courts in September 1967. The decision of the court in July 1972 rejected the plea of the accused enterprise that they had “employed prevention measures based on the latest technology at the time”, and stated that the enterprise could have known that its emission of air pollutants would be harmful to the lives and bodies of human beings. The decision further stated that “the best technology and knowledge in the world must be mobilized to devise prevention measures, without regard to the economical efficiency”, that “if such measures are neglected, then there will be no way to avoid accidents (no-fault liability for compensation)”, and that “the causal relationship is clear from the results of epidemiological examinations.”

As a result of this decision, a civil law system of compensation for damages was set up, and the Pollution-Related Health Damage Compensation Law put into effect in June 1973 as a law to provide a radical system of compensation for victims.

### 12.7.2 Recognition of the victims and Compensation, and the Expense Bearing

This law framed a system of understanding of exactly who can be recognized as a victim in such cases, stating that when those who have lived in or commuted to work in areas which have been designated (designated areas) to have frequent occurrences of illnesses related to marked air pollution for over a certain amount of time have been exposed to the air pollution (conditions of exposure), have contracted chronic bronchitis, bronchial asthma, asthmatic bronchitis, emphysema or have had recurring bouts of such illnesses (designated illnesses), then there can be said to be a causal relationship between the person’s illness and the air pollution.

A report from the Central Council on Pollution Control Measures, set down the necessary requisites for “designated areas” as those areas which have a “marked amount of air pollution” and “where there have been frequent outbreaks of related illnesses”. One typical example is when the level of air pollution amounts to an average yearly value for sulfur dioxide of over 0.05 ppm and when the prevalence of the illness is over 2 to 3 times the natural prevalence of the illness.

Health damage victims who have been recognized by the prefectural governor, according to the law, may be presented with indemnity to compensate for medical treatment expenses and for the earnings they lost due to

contracting the illness. The Compensation Law of Pollution-Related Damage also stipulates that the pollution-related health and welfare programs necessary to promote the welfare of these victims be implemented in order to help those whose health has been damaged by designated illnesses to recover, maintain and promote their health and well-being.

The expenses necessary to implement this system of compensation to victims whose health has been damaged by air pollution are to be determined by enterprises' rates of contributing to the emissions of air pollutants, and the ratio of expense bearing between stationary sources and mobile sources in factories and enterprises is to be 8:2 considering the national amounts of sulfur oxides and nitrogen oxides among all air pollutants. Expenses for stationary sources, which take up 80 percent of the overall total are to be born by all the nation's factories and establishments over a certain size, and individual factories and establishments are to bear the expenses according to their own emissions levels, using sulfur oxides as an index.

There were only 12 areas designated for the implementation of this system in the beginning, but this number gradually expanded to include 41 areas in 1978, and the number of recognized sufferers reached over 100,000 persons in 1988. Consequently, the total amount of compensation benefits on a yearly basis has exceeded 100 billion yen.

### 12.7.3 System Reform Accompanying Improved State of Pollution

Advances were made in air pollution control measures after the inauguration of this compensation system and there was such a remarkable improvement in the amount of pollution caused by sulfur dioxide, which had been used as one of the main indices of pollution for this system, that environmental standards were attained in almost all areas. A change can be seen to have occurred in the mode of air pollution, on the other hand, as the attainment of environmental standards for air pollution caused by nitrogen oxides and suspended particulate matter, has remained low, however, and has shown no marked fluctuations. More specifically, if you look at the changes in the state of pollution in yearly averages according to the continuous monitoring stations, you can see that the figures for sulfur dioxide have gone from 0.030 ppm (1973) to 0.010 ppm (1987), the figures for nitrogen dioxide have gone from 0.025 ppm (1973) to 0.028 ppm (1987), and the figures for suspended particulate matter have gone from 0.059 mg/m<sup>3</sup> (1974) to 0.041 mg/m<sup>3</sup> (1987).

After the final area was designated in 1978, a period of pure increase in numbers ensued and for every year period since then there have been roughly 9,000 newly-recognized patients and 6,000 persons leaving the system, leaving a yearly increase of 3,000 persons becoming involved with the system.

Under these conditions, the Central Council on Pollution Control Measures initiated an investigation of the management of the current system in the autumn of 1983, and issued a report in October 1986, based on the following 3 foundations: (1) that the designations of all previously designated areas be removed, (2) that recognized sufferers continue to receive compensation payments, and (3) that integrated environmental protection measures be promoted. As a consequence, the Compensation Law of Pollution-Related Damage was reformed in 1987, and all previous designations of pollution areas were removed in March 1988.

Accompanying the removal of these designations, the 14 years-long (1974-1988) recognition of new designated area was terminated. 110,000 patients were recognized at the time of this removal, and the total 180,000

(approx). Had been recognized as patients by the time of removal of designation. The total amount of compensation benefits, from the inauguration of the system until its dissolution in 1987, was one trillion yen. Even after the dissolution of the area designations, compensation benefits continued for those who had already been recognized as sufferers, and while they were receiving treatment for the designated illness. In the year of 1995, there were roughly 74,000 recognized patients, and the expenses for their compensation summed up to 89 billion yen. The cumulative expenses until that time was roughly 1 trillion and 800 billion yen.

#### 12.7.4 From Individual Patient Compensations to Health Damage Prevention Programs Directed at Entire Areas

No new pollution patients has been recognized after the dissolution of the area designations. The government, based on the consideration that they cannot deny the possible influence of present air pollution on chronic lung disease patients, determined to strengthen and promote the policies to prevent outbreaks of health damage due to air pollution, to advance research and investigations on the impact of air pollution on people's health, and to construct environmental protection surveillance systems. The government also decided that the expenses for the implementation of these health damage prevention programs were paid from the working profits of an endowment fund (50 billion yen) reserved by the government, the enterprises who construct the facilities which emit air-polluting substances and those which carry the air-pollution relating activities during 1988-1994.

### (12.1) References

- 1) Environment Law Research Group, Environment Agency, eds., Environmental Laws (1997)

### (12.2) References

- 1) Environment Law Research Group, Environment Agency, eds., Environmental Laws (1997)
- 2) Air Quality Conservation Regulations Division, Air Quality Conservation Bureau, Environment Agency, eds., Air Pollution Continuous Monitoring System Manual (1987)

### (12.3) References

- 1) Environment Law Research Group, Environment Agency, eds. Environmental Laws (1997)
- 2) Habitat Pollution Bureau, MITI, eds., 4th Revised Pollution Control Technology and Laws [Air Quality Edition]. Industrial Pollution Control Agreement (1992)
- 3) Japan Standards Association, JIS Handbook [Environment Measurements] (1992)
- 4) Air Quality Conservation Regulations Division, Air Quality Conservation Bureau, Environment Agency, eds., Smoke and Soot Emitting Facilities Manual. Gyosei (1981)
- 5) Air Quality Conservation Regulations Division, Air Quality Conservation Bureau, Environment Agency, eds., Dust Emitting Facilities Manual. Gyosei (1985)

### (12.4) References

- 1) Environment Law Research Group, Environment Agency, eds., Environmental Laws (1997)
- 2) Environment Impact Assessment Law (1997)
- 3) Air Quality Conservation Regulations Division, Air Quality Conservation Bureau, Environment Agency, Nitrogen Oxide Total-Volume Regulations Manual (Revised Edition). (1993)
- 4) Habitat Pollution Bureau, MITI, Environment Concentrations Estimation Method Manual For The Atmosphere In General Preliminary Investigations Of Industrial Pollution (1985)
- 5) Air Quality Conservation Regulations Division, Air Quality Conservation Bureau, Environment Agency, Suspended Particulate Matter Analysis and Estimation (1987)
- 6) Air Quality Conservation Bureau, Environment Agency, eds., Basic Atmosphere Related Information (1994)
- 7) Habitat Pollution Bureau, MITI, eds., 4th Revised Pollution Control Technology and Laws [Air Quality Edition] Industrial Pollution Control Agreement (1992)
- 8) Regional Planning Office, Water Environment Division, Ministry of Public Welfare, Environment Impact Manual For Final Waste Material Disposal Sites (1986)
- 9) Japan Roads Association, Road Environment Maintenance Manual (1989)
- 10) Planning and Regulating Bureau, Environment Agency, Detailed Environment Assessment (1992)
- 11) Japan Standards Association, JIS Handbook [Environment Measurements] (1992)
- 12) Nagayuki Yokoyama, Atmospheric Environment Simulation; Atmospheric Flow and Diffusion (1992)
- 13) Kondo, Jiro, Air Pollution; Analyzing the Phenomena and Creating Models (1975)

**(12.5) References**

- 1) Habitat Pollution Bureau, MITI, eds., 4th Revised Pollution Control Technology and Laws [Air Quality Edition] Industrial Pollution Control Agreement (1992)
- 2) Air Quality Conservation Bureau, Environment Agency, eds., Basic Atmosphere Related Information (1994)

**(12.6) References**

- 1) Environment Law Research Group, Environment Agency, eds., Environmental Laws (1997)