C-1 Studies on the Development of Matrix for Air Pollutants Emission and Deposition and International Cooperative Field Survey in East Asia (Abstract of the Final Report)

Contact Person Kentaro Murano
Independent Senior Researcher
Atmospheric Environment Research Division
National Institute for Environmental Studies
Onogawa 16-2, Tsukuba, Ibaraki, 305-8506 Japan
Tel:+81-298-50-2537 Fax:+81-298-50-2579
E-mail: murano@nies.go.jp

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1. Introduction

A large amount of SO_2 and NO_x which were the precursor of acid rain is emitted in East Asia followed by European continent and Northern America continent. It was forecasted that the emission of SO_2 and NO_x may increase in the near future by the development of industry and increment of population. Therefore, the deposition of acidic component will increase in East Asia. It is anxious that the damage by acid rain expand in East Asia including Japan. Acid deposition is widely recognized as one of the most serious global atmospheric pollution problems. East Asian countries face a potential regional scale, international acid deposition problem, and have recently started to expand their monitoring activities called as "Acid Deposition Monitoring Network in East Asia(EANET)".

It is pointed out that wind pattern variations associated with synoptic scale pressure system changes are extremely important for the transboundary transport of pollutants. To understand the long-range transport phenomena, series of special aerosol observations (surface and airborne) should be conducted during the winter monsoon because high concentration of SO_2 and sulfate are usually observed.

2. Reseach Objectives

In order to clarify the transboundary air pollution from the Asian continent to Japan, we conducted international air pollutants observations, emission inventory compilation and model development.

It is necessary to monitor air pollutants in China, which is a large emission-source region of acid precursors. However, such data are yet insufficient.

In order to establish a proper method of quality evaluation for the EANET program, periodic and linear regression models were applied to nine-year long monitoring results in Japan to assess their performance and to detect trends of major ion concentrations.

To understand the transboundary air pollution to Japan from Asian continent, we conducted a field survey to monitor air pollutants concentration at remote site(Echizen Head and Toyooka city: January 2000, 2001 and 2002) and sub tropical area(Okinawa Island). Gas and aerosol measurements are conducted to know the scavenging coefficients of gas and aerosol at Happo.

Comprehensive air pollutants emission inventories should be developed covering East

Asia to use the atmospheric transport model run and analysis of air pollutants monitoring and measurements data.

The atmospheric transport model including air pollutants emission, chemical transformation, and deposition module should be applied to conduct detailed(50 areas) source-receptor analysis(emission-deposition matrix). The development of the transport model of sulfur oxide that covered the wide region of the EANET territory should be carried out.

3. Research Method

The observation of air pollutants on three sites in China (Fenghuang Mountain, Liaoning Pr.; Tianheng Island, Shandong Pr.; Shengsi Island, Zhejiang Pr.) was conducted. Besides ground-based observations in China aircraft observations of acid-rain precursors were carried out over the East China Sea. Moreover, aircraft observations in China were carried out for the first time as a cooperative research between China and foreign countries.

Advanced data- analysis techniques were developed in terms of data control chart, temporal trend analysis models and standardized procedures of precipitation monitoring in cold regions. Also discussed was requirements and constraints to produce contour maps of precipitation chemistry of Japan.

The ozone concentration is monitored with Dasibi model 1006-AHJ Ozone Monitor at Hedo. Hedo is located at the northern tip of Okinawa Island and considered as low pollution area. Long term monitoring of air pollutants with 4 stage filter pack method was conducted at Toyooka city which is a coastal area of Sea of Japan designated as low pollution area and Kobe city which was urban area. High resolution concentration measurements of air pollutants is conducted at Toyooka city in typical winter season, at that time the long range transport of air pollutants from Asian continent is expected. The field survey was conducted from 18 January through 1 February, 2000, at Echizen Head in Fukui prefecture which was faced to Sea of Japan and considered as low pollution area.

The filters are placed in a series of Fluoropore, Nylon, alkali impregnated and acid impregnated to collect particulate matter, gaseous nitric acid and part of sulfur dioxide, the rest of sulfur dioxide and ammonia. The collected compounds are extracted followed by ion chromatographic determination of corresponding ionic species.

Washout process of sulfur compounds on a mountainous area was studied. Observations of air pollutants and precipitation were made both at the ground station and at the mountain station simultaneously in the Chubu mountainous area. Elevated concentrations of sulfur dioxide (SO_2) exceeding the environmental standard were observed in Nagano Prefecture as well as in the Kanto and Tokai areas of Japan in August and September 2000. Measurements of gases and aerosols by a four-stage filter pack method were made at Nagano, Mt. Happo, and the village of Hakuba in the north of Nagano prefecture.

Data base for emission factor and emission source compilation was conducted with main focus on China which is the biggest emission source in East Asia. As for NH_3, emission amounts from livestock, fertilizer application, biomass burning, human sweat were estimated in China, Taiwan, South Korea, North Korea and Mongolia. In GEIA(Global Emission Inventory Activity), 1 degree resolution gridded dataset with base year of 1990 was established already, however, we provided emission data with 1 degree resolution base year 1995 upon the introduction of change of agricultural activities. Concerning South Korea and Taiwan, we used government estimation as base information, however, energy consumption and activities on industrial production and new information on emission factors are utilized for to estimate NH_3 emission amount.

The atmospheric transport model including air pollutants emission, chemical transformation, and deposition module was applied to conduct detailed(50 areas) source-receptor analysis(emission-deposition matrix).

The transport model is constituted by two parts; the meteorological model which predicts meteorological field and diffusion model. The regional spectral model (RSM) was applied to the meteorological model as 40km of horizontal resolution, and the Lagrangian particle method was adopted in advection and diffusion model. Though this method has adifficult point in order to express chemical reaction of the air pollutants, it is a convenient method in order to obtain the contribution rate from the emission sources. The gas phase and liquid phase oxidation of SO_2 course by the cloud were included for the transport model, and it is expressed by the transformation coefficient from SO_2 to SO_4^{2-}. And, not only dry deposition but also wet deposition course by the precipitation was adopted.

4. Results and Discussion

We started to observe air pollutants on three sites in China (Fenghuang Mountain, Liaoning Pr.; Tianheng Island, Shandong Pr.; Shengsi Island, Zhejiang Pr.). The experiments were carried out from January 15 to 30, 2000. During this experimental period, concentrations of SO_2 and particles became high on the same day at Fenghuang Mountain and Tianheng Island. It showed that both the sites were covered with the same heavily polluted air mass. The first aircraft experiments with co-operation of Japanese scientists was performed in March 2002. Pollutants of very high concentration confined to lower altitude were observed in both the experiments when high-pressure system covered the observation sites.

Ionic concentrations were shown to obey log-normal distribution rather than conventional normal distribution, based on which a control chart was proposed to identify outliers for further data analysis. Two types of models were studied to provide temporal trends of precipitation chemistry in Japanese. One model employing non-transformed concentrations divulges that sulfate concentrations are generally decreasing whereas nitrate is increasing. Ammonium and calcium ions were changing although both increasing and decreasing trends were discerned in Japan. The other model with logarithmically transformed concentrations clearly displayed similar trends of deposition and concentrations of sulfate and nitrate. The results were further discussed in terms of potential sources both domestic and overseas affecting precipitation chemistry in Japan. Kriging method was discussed to apply datasets at 44 stations in Japan. The number of the monitoring stations was found to be insufficient to produce appropriate contour maps, which strongly suggests datasets for more stations should be included in the datasets. Monitoring techniques in Russia was developed and standardized for practical operations. HPLC Determination of hydrogen carbonate ion was developed to apply actual samples, which successfully decreased unacceptable samples in terms of ion balance criteria from 58% to 6.6% of the samples collected in 17 months.

The ozone monitoring results collected in Okinawa exhibited the typical variation pattern of subtropical region. The variation of ozone concentration was well elucidated by the movement of Pacific high, Continental high, Migratory high pressure system and Low pressure system. From December to February the range of ozone concentration is 24 ~ 65 ppbv (average 45.3 ppbv). The ozone concentration is mainly governed by the Continental high pressure system, so the air pollutants originated from Asian continent is transported to Okinawa. From March to May, the variation of ozone concentration is the largest and it ranged from < 3 to 84 ppbv. The short cycle of pressure system and the location of the front is the main cause of this large variation of ozone. From June to August, the range is big (< 3 ppbv ~ 82 ppbv); however, the average concentration is the lowest (16.4 ppbv) indicating the

coverage of Pacific high pressure system. From September to November, the variation is big (< 5.0 ~ 84.4 ppbv) and the average concentration is larger compared with that of summer season. The transport of Continental high pressure system brought this high concentration of ozone.

In Toyooka city, occasional high concentration peaks are observed for SO_2 and NH_3 concentrations, however, the peak time is different indicating the different source. The concentrations of HCl and HNO_3 are low during the observation period. As for particulate matter the concentration of Cl^-, NO_3^-, SO_4^{2-}, NH_4^+ and Na^+ showed occasional peaks. The high concentrations of Na^+ and Cl^- are introduced by sea salt particles. The high concentrations of NO_3^-, SO_4^{2-} and NH_4^+ are attributed to the anthropogenic origin.

In Echizen Head typical winter weather pattern were observed at 20 ~ 21, 25 ~ 27 and 30 ~ 31 January. As for gasous compounds, SO_2 showed the highest average concentration. Occasional high consentration peaks of SO_2 are observed at 19th and 30 ~ 31st, January, reaching 80 and 135 nmol m^{-3}, respectively. The variation of NH_4^+ and SO_4^{2-} concentrations correlated very well. The SO_4^{2-} concentration reached 100 nmol m^{-3} at 18:0, 30th. From back trajectory analysis, the air mass traveled over at the foot of Shandong peninsula, Shanghai and Chugoku district in Japan.

Scavenging ratios of SO_4^{2-} were ranged from 65 to 1827 and 416 for geometrical average, which was about the same value as 450 at Northwestern Kyushu region. The concentration of SO_4^{2-} in aerosols was extremely high at 44.4 μg/m^3 in Nagano on September 14, and on September 15 it exceeded 30 μg/m^3 at both Mt. Happo and Hakuba. On the basis of the assumed ion balance of SO_4^{2-} and NH_4^+, unusually high levels of sulfuric acid (H_2SO_4) and high molar concentration ratios for H_2SO_4/SO_4^{2-} were estimated for each of the 3 locations: H_2SO_4 concentrations were 21~33 (Nagano), 11~21 (Mt. Happo), and 6.8~18 μg/m^3 (Hakuba); molar concentration ratios were 46%~72%, 34%~65%, and 22%~58%.

Gridded emission amounts of SO_2, NOx, NMVOC(non methane volatile organic compounds) and NH_3 from anthropogenic and natural sources are compiled with satisfactory accuracy. NMVOC emission amounts were compiled for China, Korea, Japan and Taiwan. China is the major contributor for East Asian NMVOC emission. Fossil fuel combustion is the biggest followed by biomass burning by sector contribution. The high emission areas are located at the seashore line of central China. Emission amount comparison between our data and those of obtained from European researcher was conducted. Finally we compiled next mentioned emission inventories as CD-ROM for the convenience to use for model run and air pollutants monitoring and measurements activities.

Target Compounds: SO_2, NOx, NMVOC, NH_3
Target Areas: China, Taiwan, South Korea, North Korea, Mongolia, Japan
Horizontal Resolution: 1 degree
Time Resolution: Annual Value(As for biogenic NMVOC monthly and noon time night time resolution)
Base Year: 1995

The simulation model run performed during the period from January 15 to February 15, 1999, showed that about 78% of the sulfur deposition in Japan was originated from foreign countries. The most heavy contributor to Japanese sulfur deposition is three times bigger than that of lighter contribution area.

According to the source–receptor analysis of air pollutants in East Asia, the domestic including active volcano fraction to total sulfur deposition is sbout 64% and 20% in July and December, respectively. In July, the contribution of volcano is especially large in western

part of Japan. The main contributor is eastern China(around Shanghai and Qingdao) to Japanese deposition. On the other hand, in December the domestic contribution is small and Chinese contribution is large and major contribution area are around Dahlian and Harbin. The contribution from (around Peking and Taiyuan) are large in both July and December. The coastal area of Sea of Japan which are supposed to suffer from continental contribution, the deposition of sulfur did not differ in July and December. However, the fraction of contribution area is quite different.

Transport simulation of SO_2 and SO_4^{2-} of the EANET region were carried out on January and July 2000 by using transport model. The anthropogenic and volcanic emission sources data set was made, and each was placed in the point source of 120 sites for anthropogenic emission and 20 sites for volcanic emission. From simulation result, the transport between China, Korea and Japan was main in the EANET region, and it was found that the transport from the source in low latitude such as Indonesia did not reach to middle latitude. And, SO_2 concentration and wet deposition amount of SO_4^{2-} reproduced by transport simulation was compared with the measured value at several observation station in our country. On January and July, though the SO_2 concentration agreed with the observed value comparatively well. both of January and July were results of the underestimation on SO_4^{2-} wet deposition amount. The contribution from the emission source in Korea was large in January for the wet deposition amount in Oki receptor, and the contribution from Sakurajima and Osaka was also observed.