B-14,4,2 A Study on Emission Analysis of Greenhouse Gases and Related Compound from Daily Life Facilities.

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Abstract

The sampling and analytical methods for greenhouse gases(GHGs) and related compounds such as CO_2 , CH_4 , N_2O or others were developed. Field test studies on GHGs emission were done by using these methods at municipal solid waste (MSW) incinerators, MSW landfill sites and night soil treatment plants (NSTP). Emission rates of CO_2 , CH_4 , N_2O , halocarbons from MSW incinerators (partly large stationary combustion sources) in Japan (or worldwide) were estimated roughly. Utilizable energy volume emitted from waste treatments and the energy amounts used for operation of MSW incinerators and NSTP in Japan were also calculated.

Key Words Global warming, Emission analysis, Greenhouse gas, Waste treatment

1. Introduction

The contribution of the trace greenhouse gases (GHGs), such as Methane (CH $_{\rm a}$), nitrous oxide(N $_{\rm 2}$ 0), halocarbons(CFCs), ozone(O $_{\rm 3}$), to the global warming is considered to be increasing numerically and relatively during the last two decades. In addition, the emission of these trace GHGs can be easily reduced, compared with CO $_{\rm 2}$ emission which is directly connecting the energy uses, if the sources and the emissions of them could be clear. They, however, are obscure in detail parts, especially from daily life facilities.

2. Development of sampling and analytical methods for greenhouse gases and related compounds.

The field surveys at the sources of trace GHGs and the developments of the sampling and analytical methods for the surveys are the objectives in the first stage of this project. The sampling train at waste combustion facilities is composed of, a glass probe with glass fiber filter, a 1 N sodium carbonate solution (20 mL) impinger cooled with ice, a glass wool cooled impinger, a sample bottle (volume: ca. 1 L) with teflon stop valves at each end, and a suction pump. More than 99.8% of sulfur dioxide and condensed

water in flue gas which produce nitrous oxide as an artifact are removed by the two impingers.

A single separation column is used for nitrogen, oxygen, carbon dioxide, and carbon monoxide for easy handling under different conditions and detectors shown in Table 1. Carbon monoxide at ppm level is analyzed by a flame ionization detector after the conversion to methane with a nickel catalyzer tube (17cm*6mm 0.D.) at 350°C.

 CO_2 , coexisting in flue gas at high concentration, seriously affects the N_2O sensitivity in GC-ECD, as well as O_2 . Thus, the GC-ECD method with carrier gas doping technique were developed for N_2O in flue gas. In this method, the sensitivity of ECD to N_2O was increased to about five times, and the influence of CO_2 or O_2 concentration in the sample gas to the determination of N_2O could be minimized.

Analytical methods by gas chromatography for methane, sulfur dioxide, lower hydrocarbons are also developed, and are shown in Table 1.

Analytical methods using gas chromatographs were improved in introduction techniques. A gas sampling valve attached a loop, which connected vacuum line, is used for introduction of a constant volume of sample gas in a bottle. Thus, the analytical precision of nitrogen, oxygen, and carbon dioxide rise less than 2% of relative standard deviation from 10% with syringe introduction.

Analytical techniques for GHGs in field studies with potable GC, "Microsensor Gas Analyzer Model 200", for field study were also developed and was used at landfill in Bangkok.

compounds	Introd. method	Separ. column	GC oper. cond.	Det.
N2,02	loop	Mo.Si.5A, 1.8m	80°C, He:40ml/min	TCD
CO ²	loop	ditto	250°C, He:40ml/min	TCD
CO	loop	ditto	80°C, He:40, H ₂ :40	FID
N20	loop	Porapak Q, 3m	36°C, N2:47, CO2:0.4	ECD
S02	loop	10%TCP/C545,2m	80°C, N ₂ :43ml/min	FPD
CH⊿,NMHCs	loop	Porapak N, 3m	60°C, N ₂ :25ml/min	FID
C ₂ ·HCs	liq. 02 cryog.	Porapak N, 3m	70°C, N≥:25ml/min	FID
C3-C5·HCs	liq. 02 cryog.	10%DMS/C22,11m	0°C, N ₂ :40ml/min	FID

Table 1. Analytical conditions

In this method, a special made wet test gas meter, which generate 50 or 500 electric pulses with one revolution of the measuring drums (volume: 500 mL), a digital flow indicator and a mini-computer are used. The measurement range of the method is very wide, as 5 L/min to less than 1 mL/min, and

^{3.} Development of automated continuous measurement method of gas volume emitted from garbage decomposition.

it permit the monitoring of small volume of gas emitted from garbage decomposition. The gas contains methane or carbon dioxide at high concentration, and it is regarded as one of main sources of methane.

4. Field studies on GHGs emission.

(1) MSW incinerators

The data concerned MSW in Japan was compiled from the databook and the amounts of MSW treated in incinerators were separately calculated with four types as shown in Table 2.

Table 2 Incineration plants and treatment amounts of MSW in Japan (in FY 1987)

Type of furnace	No.	Total cap.	Treat. amount	Ratio
	plants	(K·ton/y)	(K·ton/y)	(%)
Fully continuous	409	4,241	2,263	69.4
combustion				
Semi continuous	230	642	249	7.6
combustion				
Mechanized batch	970	990	523	16.0
combustion				
Fixed grate-bar	284	90	47	1.4
batch combust				
Others			180	5.5
Total	1,893	5,963	3,262	

Field test studies were done at various, type MSW incineration plans by using the developed methods and emission rates for each MSW incinerator was estimated as following.

The amount of MSW incinerated in furnaces varies continuously and the emission values of CH_4 and N_2O in sample bottles are resulted from the instantaneous burning of MSW. Thus, the weight of MSW feeding into the hopper of a furnace for certain period is not appropriate for the calculation of emission factor (EF). We introduced the following equation for the EF at the MSW incinerators, in which the CO_2 concentration in sample gas is regarded as the incinerating MSW, on the assumption that the organic carbon in MSW is completely changed into CO_2 after incineration;

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EF(g/ton)=44·C<sub>N2O</sub>(ppm)·Rc·100/C<sub>CO2</sub>(%)/12

[Where; 44: Molecular weight of N<sub>2</sub>O, C<sub>N2O</sub> and C<sub>CO2</sub>: N<sub>2</sub>O and CO<sub>2</sub>]

[concentration in sample, 12: Atomic weight of carbon]

Rc: Organic carbon content in MSW (20-25% in Japan)]
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About 150 samples were taken at 13 furnaces or various types of incineration plants. The emission factors (EFs) of CH₄ and N₂O were calculated from the measurement data. EFs of CH₄ were low (-1 \sim -2 g/ton) at big fully con-

tinuous combustion types, and high (40 \sim 2300 g/ton) at small mechanized batch combustion types. EFs of N₂O (30 \sim 300 g/ton) were not depending on the types so much.

(2) MSW landfill sites

CH₄, N₂O, CO₂ and others were measured at two landfill sites in Japan. 12-38% of CH₄ in the gas removal pipes was observed in one sites. N₂O was emitted at 0 - 7.4 ppm (sample no.=17) and 3.3 - 5.0 ppm (no.=4) at the each of two MSW landfill sites

The field study in the landfill sites in Thailand was carried out at the wet and the dry season for the determination of the emission rates at tropical countries, which is essential to estimate the emission rates in the world. The ratios of CH_4 to (CH_4+CO_2) in the samples gases are about 55%, close to those obtained in Japan.

(3) Night soil treatment plants

The CH₄ and N₂O were measured at four facilities of three types, that is, one NSTP with conventional denitrification process (CDP), two NSTPs with high loading denitrification process (HLDP), and one combined septic tank (CST). Their concentrations in the ducts of the tested facilities were changed as $3 \sim 600$ ppm(CH₄) or $0.5 \sim 3400$ ppm(N₂O). N₂O from HLDP and CH₄ from CST were especially high and further field studies at different facilities or for other seasons are needed.

5. Estimates of carbon dioxide from municipal solid waste incinerators.

The three different estimation methods are developed for the purpose on the basis of the carbon content of waste materials, components of waste materials with the carbon content of each component, and the emission factor of carbon dioxide. We calculated the values of the carbon content, the composition rate and the emission factor from our original data or already published data for 1988. The emission amounts obtained by the three methods are 8.20, 8.19 and 7.71 Tg, respectively. Thus, we currently estimate about 8 Tg of carbon dioxide emitted from municipal solid waste incinerators in Japan.

6. Estimates of CH_4 and N_2O from municipal solid waste incinerators.

The emission rates of CH₄ and N₂O were determined roughly to be ca.7 $Gg(CH_4)/year$ and ca. 5 $Gg(N_2O)/y$ in Japan, and ca. 8 $Gg(CH_4)/y$ and ca. 11 $Gg(N_2O)/y$ in OECD countries, respectively. Thus, the magnitude of the emissions from MSW incinerators is found to be not considerable.

7. Estimates of methane emission from large stationary combustion sources.

The emission factors of methane for nine categories of large stationary sources working continuously with various fuels were estimated from the

data published in Japan, on the assumption that the feeding air contained 1.8 ppm of methane.

Boilers, heating furnaces, metal furnaces and glass melting furnaces were found to have minus emission factors (-0.0645 -0.505 ug(CH₄)/kJ), and as a result, they are regarded as the sinks of atmospheric methane. The maximum removal rate of methane at electric power plants is determined roughly to be ca. 1.0 Gg/year in Japan, ca. 18 Gg/year in the world. On the contrary, coke ovens and sintering furnaces were considered to be significant industrial sources, and the minimum emission rate is determined roughly to be ca. 20 Gg/year in Japan, ca. 120 Gg/year in the world, on the basis of modernized facilities with efficiently controlled operations. Therefore, the magnitude of the methane emission from large stationary sources is not considerable if they are operated in similar conditions to those in Japan.

Some facilities, however, such as coke ovens or hot blast furnaces which generate coke oven gas or blast furnace gas which contain methane at high concentration, are shown to have a higher potential possibility of methane emission into the air.

8. Estimates of utilizable energy emitted from waste treatments.

The heat of combustion (Hc) of MSW were was separately estimated to be 1895 kcal/kg for a large city type and 1580 for other types from the compositions and the water content of MSW. UE from MSW incinerators is determined roughly to be ca. 54 Pcal/year in Japan (41 from continuous type, and 13 from others). The magnitude of UE from MSW incinerators is 2.75 % of the UE from crude oil (ca.1950 Pcal/y in 1988) currently consumed in Japan. The incineration of industrial waste and agricultural waste, which have higher heating values, are out of the scope of this estimation. Thus, total UE from waste would amount to several percent and is considerable.

UE by the combustion of CH_{4} emitted from night soil plants with anaerobic digestion treatment in Japan is also estimated to be ca. 500 Tcal/y, which is not much, compared to the UE from solid waste or crude oil.

9. The energy used for operation of MSW incinerators and night soil treatment plants in Japan.

They were estimated from the database of ca. 1700 incinerators for FY 1987 and 1250 night soil plans for FY 1988. The total energy estimated to be ca. 4460 Pcal and ca. 2900 Pcal, respectively.