B-2.5 Source Inventory of Industrial Facilities

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Abstract

The Source inventory of methane (CH_4) and nitrous oxide (N_2O) from industrial activities are studied. Coal mines in Japan have routine monitoring system for CH_4 contents based on the mining safety law. The emission factors of CH_4 from coal mines in Japan are ranging between 12 to 116 m3 per ton of coal production. The average emission factor is 27 m^3/ton. The annual total amount of CH_4 emission from coal mines in Japan is 0.22 Tg at 1989 fiscal year statistics and 0.15 TgCH_4 was discharged into atmosphere. CH_4 emission from Chinese coal mines was estimated by using emission factors based upon the data of Japanese coal mines and Chinese statistical data. Total coal product in China is 1,138 Mton in 1993 and 8.8 Tg of methane is estimated to discharged into atmosphere due to the coal mining.

A recent study on the contribution of the combustion of fossil fuels has shown that the earlier results are incorrect because N_2O was being artificially produced in the flask. The latest estimation of the global flux of N_2O from combustion sources is between 0.1 and 0.3 Tg-N per year. Updated estimation of annual nitrous oxide emissions from fossil fuel combustion in Japan shows that the total N_2O emission from coal, oil and natural gas is 2.6 Gg-N in conventional combustors and 2.3 Gg-N in fluidized bed combustors. The annual N_2O emission from power plants in Japan was estimated as 1 Gg-N/year by latest data of power plants.

Total annual emission of N_2O from fossil fuel combustion in China was estimated as 12 Gg-N on the assumption that 3% of coal is burned by fluidized bed combustion. Global annual N_2O emission from coal combustion was estimate as 90-210 Gg-N from world's annual coal consumption.

Key Words Industrial Facilities, Methane, Coal mining, Nitrous Oxide, Combustion

1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are one of the greenhouse gases and their concentration in the atmosphere is increasing year by year. The sources are natural and anthoropogenic. The contribution is estimated in many reports recently due to the importance for the roll in the climate change.

Methane emission

The sources of CH_4 are variety; from natural wetlands, rice paddies, animals, oceans and human activities (eg. coal mining, landfills and so on). The total amount of emission to the atmosphere is estimated to be 525 Tg (teragram = 10^{12} g) per year. The energy-related emission is 120 Tg and coal mining contributes only 35 Tg of the total on global scale. CH_4 emission from coal mining is estimated between 10 and 50 Tg per year. Coal mine is one of the sources and discharges methane into the atmosphere during the mining and handling on the ground. The emission factor varies due

to coal mines, that is coal rank, depth of pit and treatment of degasification. Japanese case was analyzed by Hayashi and Isei[5]. The coefficient changes more than 100 to 10 m^3/t at the mining site. The averaged discharge coefficient for total discharge is 33 m^3 per tons of the coal product and 26 m^3/tons is obtained after the degasification process. These values are larger compare to the estimations in the DOE[2] report and the same order in OECD[15] report.

The purpose of this study is to develop a source inventory of methane from coal mining in Japan and east Asia and obtain an emission factor. CH_4 in coal seams is created in the process of coalification and stored inside the coal and surrounding rock strata. The quantity of CH_4 mainly depends on the coal rank (coalification) and the depth of the mine. The amount of CH_4 increases directly with the coal rank or coalification. Deeper coal seams generally contain more CH_4 than shallow coal. The adsorptive capacity of CH_4 depends on the coal itself and the surrounding rock. The CH_4 seeps back into the atmosphere during coal mining.

Nitrous oxide emission

Once the total global production of N_2O of 11.2-16.1 Tg N per year was consistent with the amount needed to balance the rate of global destruction and its rate of accumulation of N_2O in the atmosphere (10.5-17.5 Tg-N/year). According to the new artifact-free estimate for N_2O production due to fossil fuel combustion and biomass burning, the estimated global production of N_2O is reduced from the above listed range to 7.5-12.6 Tg N. The production by fossil fuel combustion is estimated only between 0.1-0.3 Tg-N/year by IPCC report[7,8], rather than the earlier estimates of about 3.2 Tg-N/year.

In this study, N_2O emission from industrial activities in Japan was estimate. N_2O emission in China and globe were estimated with forcused on the coal combustion facilities.

2. Methane emission from coal mining

2.1 Research methods

Methane is used as trace gas to study the mechanism of the atmospheric behavior, physical and chemical, and the interaction between atmosphere and earth surface using numerical models. The simulation calculation requests the source inventory in grid scale as same as the model. Fung et al. estimate the methane flux from coal mine to atmosphere by the location of coal mine and coal product in 1 degree by 1 degree mesh. Taylor et al.[17] estimate the methane flux is proportional to the coal consumption, i.e. CO_2 emission. The methane emission in China is estimated more precisely in this investigation employing the statistics of coal industry.

To answer the request from modelers, the atmospheric chemists organized an international group to set up the database of the flux of the trace gases. The movement is called Global Emissions Inventories Activity, GEIA, as a component of the International Global Atmospheric Chemistry Project, IGAC (Graedel et al.[4]). This investigation was carried out along the guideline of this activity.

Coal mines in Japan have a routine monitoring system for CH_4 contents based on the mining safety law. CH_4 concentration is measured at the outlet of the air ventilation shaft. Degasification by boreholes is employed in most coal mines in Japan. CH_4 concentration is also measured at boreholes. The total emission of CH_4 during coal mining is the summation of the degasification and the emission at the mining face and wall. The recovered CH_4 is used as fuel for boilers as a domestic energy sources, thereby reducing its atmospheric discharge.

Methane emission inventory is estimated for Chinese coal mining. The methane emission rate is largely concerned with the coal mining product. The coal productivity is cited in the 'Annual

Report of Coal Industry in China'[1]. The latest report was published in 1990 when we started this work. The total amount of coal product in China was 1054.15 mega tons (Mt) in 1989, 458.3 Mt from major coal mining called "Tong-Pei" and remaining 595.85 Mt from local coal mining. Major coal mining sites are listed in the report. Most of each summation is consistent in each table, but there are some exceptions. For example, the total coal product in the Ryan-nyang province in the summarized table was 49.8 Mt but 46.7 Mt in the individual tables. The statistics mostly depended on the individual tables. The individual tables list up the major mines; names, products and so on. The location is identified by the local name on map, Atlas of the People's Republic of China, published by the China Cartographic Pub. House, 1989. Most of major mining sites and a few of minor local mining site can be seen on the map. It amounts 454.63 Mt (99.2%) for major and only 66.6 Mt (11.2%) for local mines. The coal ore is found in a restricted area in geological, the undefined mines and their products are distributed according to the known mine and products.

2.2 Results

Figure 1 shows the relationship between CH_4 emission and mining depth. The open circle shows the total emission and the solid circle shows the direct emission into the atmosphere after degasification. It may say that the emission rate is relatively proportional to the depth.

The emission factor for CH_4 from coal mines in Japan ranges from 12 to 116 m^3 per ton of coal production. The mean emission factor is 27 m^3/ton after the reduction process. The annual total amount of the CH_4 emission from coal mines in Japan was 0.22 Tg in fiscal year1989; 0.15 Tg CH_4 was discharged into the atmosphere.

The emission factor for CH_4 from coal mining depends on the depth of working face and the coal rank. Bituminous coal is the main product in Japan. The obtained emission factors are larger than the estimation by Barns and Edmonds for United States, 7.3 m^3/ton, or for world estimation, 8 m^3/ton. The emission factor for deep mining depth is comparable to the estimation for long wall coal mining, 56 m^3/ton. The original emission values (open circles in Fig.1) can be reduced by degasification and the recovery of CH_4, which is used as fuel and converted to CO_2. CH_4 has a larger radiative force to the atmosphere than CO_2. Therefore, the application of the enhanced degasification system for highly gaseous coal seams through boreholes prior to mining is useful for energy efficiency and mine safety. The reduced emission factor in Japan is the same level as that cited in OECD report[15].

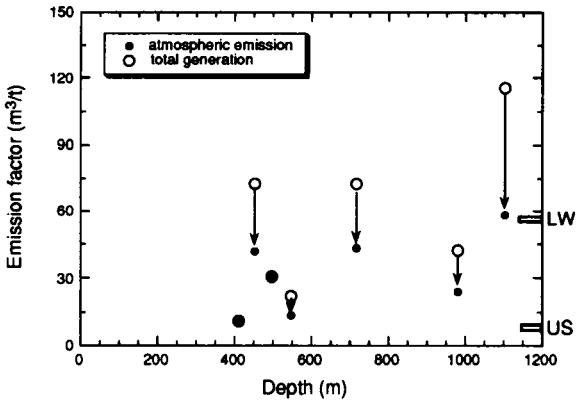

Fig. 1 CH_4 emission factor.

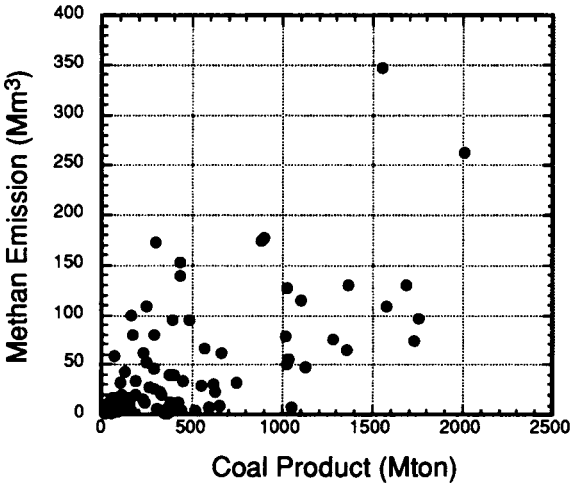

Fig. 2 CH_4 emission with coal product.

Figure 2 shows the relationships between the coal product and methane emission in China. The methane emission from Datong Mining Bureau. The methane emission there is extreamly smaller than other area. This is supposed to that methane emission is not monitoerd all mines of this bureau or is after the degasification. The emission factor is 11 m^3/ton. This is a little smaller than the

values reported past.

The methane emission can be estimated by multiplying coal product by the emission coefficient. Each report shows the different emission coefficient. The DOE report gives the coal-methane release factor, 8.4 m^3/ton for the world average and 6.1 for Japan, Australia and New Zealand. This region is categorized by other reason, geopolitic characterized region. The coal mining situation in Japan is different from Australia. These values were derived based on the adsorptive capacity of coal. The methane is released from coal at the pit, wall and gob. The release factor, 56 m^3/ton, in the report is more realistic for on-going coal mines. The recent OECD report recommended 27.1 m^3/ ton for the average emission coefficient for ventilation-related releases from underground mines. This is expressed as

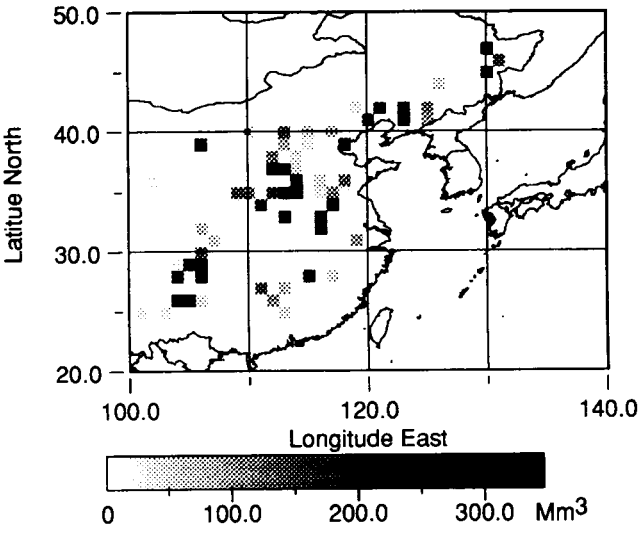

Fig. 3 CH_4 emission from major coal mining in China.

CH_4 emission coefficient (m^3 CH_4/ton coal mined)

$=2.04 \times C + 8.16$

where C is in-situ methane content in a unit of m^3 CH_4/ton of coal. Fig. 3 gives the coal product on a 1 degree by 1 degree grid scale in China. The maximum is 6.8 Mt in the 40 degree north and in the 113 degree east, at Datong mining area in Shanxi province. If the emission coefficient 33 m^3/ ton is used to estimate the methane emission, it amounts 25.6 Tg/year for China. This is larger than former estimation, for example, 9.3 Tg/year by Kirchgessner et al., 1993. The reasons are; (1) the surface coal mining is common in China and the emission factor for the surface mining is small, about 1/10 of the underground mining, and (2) some mining sites are provided the degasification system. This estimation is maximum methane emission because we did not consider these factors. It is necessary to cooperate with Chinese for further investigation.

Methan emission is monitored at 657 coal mining site of 88 Tongpei Mining Bureau in China. Total methan emission was 4,480 Mm^3. The emission factor is 11 m^3 per ton of coal product. Figure 3 shows the distribution of methane emission from major coal mine in China, 1993, in 1 degree each of longitude and latitude. The maximun emission comes from 37 north and 113 east, central part of Shanxi province. It is 0.25 Tg per year. Total coal product in China is 1,138 Mton in 1993 and 8.8 Tg of methan is estimated to discharged into atmosphere due to the coal mining. This is smaller than the forgoing research, ex. 16 Tg by Boyer et al. 1990 or 9.3 Tg by Kirchgessner et al[10]. 1993. This figure seems reasonable because this is getting smaller as the investigation proceeds.

3. Nitrous oxide emission from industrial facilities

3.1 N_2O emission from Japan[13]

Industrial sources in the anthropogenic source are power plants, nylon manufacture, sewage disposal and automobiles. To reduce their N_2O emission, the mechanism of N_2O formation and destruction must be understood.

Power plants

Austrian data[19] shows the detailed source distribution among different type of combustors. The

annual N_2O emissions of stationary combustion sources are estimated to be 715 t ± 20% based on the measurements at 45 combustion sources. 40% comes from conventional combustion systems and 12% from non-conventional fuels. Fluidized bed combustion (FBC) produces 41% and DeNOx 7% of the pyrogenic emissions. Field assessments[16] of N_2O emissions by joint research in European Communities show the low emission characteristics of stoker-fired plants, 2-3 mg/m3 and of several pulverized fuel fired plant, less than 5 mg/m3. Measurements from FBC have confirmed generally higher emissions, 40-140 mg/m3, in particular 165 mg/m3 for circulating fluidized bed combustors (CFBC). In Japan, the average concentration of the stack flue gases from 43 conventional power plants (coal, oil and LNG fired) is 0.3 ppm[18]. The other combustion studies for the individual type of combustor show the same trend to above measurements[12].

Waste management and Incinerator

In the measurements, in Japan, for waste management system to animal, sewage and municipal waste, N_2O emissions from swine waste is higher than those of other animals. N_2O is not detected in anaerobic digestion gas but in aerobic digestion gas. N_2O concentrations from stationary sources such as gas boiler, municipal refuse incinerator and sewage sludge incinerator are respectively a few ppm, 1-20 ppm and 10-150 ppm. The concentration due to municipal waste incineration depends on the type of the incinerator, 0.8-24 ppm for the step grate type, 4.0-78 ppm for the fluidized bed type and 0.7-1.4 ppm for the fixed grate type. Especially for sludge incineration using the fluidized bed type, the concentration is significantly high, 270-600 ppm. The operating temperature is between 770 and 812 deg C. In the municipal waste incinerator of batch combustion type under rich air condition, relatively high concentration of N_2O is observed after ignition of the furnace. Consequently, the emission factors of N_2O is between 25 to 293 g/ ton, depending on the size and type of incinerator, or the composition of municipal wastes and the combustion conditions. N_2O emissions in 1988 from waste management systems in Japan are estimated to be 1.7-5.9 Gg N_2O for municipal waste incineration, 1.1-2.8 Gg N2O for sewage sludge incineration, and 0.1-2.0 Gg N_2O for animal wastes, totally 2.9-10.7 Gg N_2O per year. Waste management system is an important source of N_2O emissions. In the other estimation, N2O emissions from the stationary combustion, automobiles and fertilizer is respectively 5.7-20, 18, and 0.6-3.3 Gg N_2O per year.

Flue gas treatment

Efficiency of N_2O removal by NOx removal treatment is not so high, 8-9% for selective catalytic reduction (SCR) to pulverized coal fired boiler and diesel engine. SO2 removal treatment by the wet scrubber, for instance, equipped to cupola and municipal solid waste incinerator results in slight increase of N_2O concentration. The wet scrubber (NaOH) to sludge incinerator is increased as twice as the inlet concentration. In addition, dust removal treatment by wet electrostatic precipitator and bag filter is also ineffective on N_2O removal, the N_2O concentration after dry electrostatic precipitator is increased up to 44%. It was also reported that wet deSOx and dry deSOx with char cannot remove the N_2O in flue gas. This means that the ordinary flue gas treatment is not available to N_2O removal treatment, or rather some treatment increases N_2O emission. Although there is an exception of NOx reduction treatment using three-way catalyst to gas engine, which can remove N_2O emission up to 93%, the removal efficiency is considered to depend strongly on the operating condition as seen in three-way catalyst with automobiles.

Chemical industry.

A source of N_2O was pointed out to be as a by-product in the manufacture of 6,6- and 6,12-nylon. Nylon polymers have typically been formed by condensation polymerization of a dicalboxylic acid and diamine. The most widely used diacid, adipic acid, is prepared primarily by air oxidation of cyclohexane to cyclohexanol-cyclohexane mixtures, followed by oxidation with N_2O to adipic

acid. When about 1 mol of N_2O per mole of products is produced, a 1989 worldwide production of 2.2 x 109 kg of adipic acid corresponds to about 1.5 x 1010 mole (660,000 tons) of N2O by-product. This N_2O production would then account for 10% of the observed annual increase of tropospheric N_2O. It is also reported that about 545,000 tons of N_2O were produced in 1990 via adipic acid manufacture, while about 371,000 tons were ultimately emitted. On the other hand, major adipic producers worldwide have agreed to substantially reduce industry emissions by 1996 through sharing of following technology options: (1) Conversion of N_2O to recoverable NO, (2) Catalytic dissociation to N_2 and $O_2$and (3) Improvement to the commercially demonstrated process for N_2O decomposition in specially designed boilers.

A recent study on the contribution of the combustion of fossil fuels has shown that the earlier results are incorrect because N_2O was being artificially produced in the flask. The latest estimate of the global flux of N_2O from combustion sources is between 0.1 and 0.3 Tg N per year. Updated estimation of annual nitrous oxide emissions from fossil fuel combustion in Japan was estimate as follow;

(1) Set the emission factor for each type of combustor
(2) Calculate the N_2O emission from above emission factor and annual fuel consumption which is listed in annual statistical data.

Calculation shows that the total N_2O emission from coal, oil and natural is 2.6 Gg N in conventional combustors and 2.3 Gg N in fluidized bed combustors. Thus, total annual N2O emission from fossil fuel combustion by stationary combustors is estimated as 5 Gg-N.

3.2 N_2O emission from coal combustion in China

Annual emission of N_2O from fossil fuel combustion in China was estimated. More than 70% of China's primary energy source depends on coal. China's annual coal consumption is 730 millions tons in 1988. In estimation of N_2O emission from coal combustion, share of fluidized bed combustion among the coal combustion is significantly important. If the share of fluidized bed combustion is assumed as 3%, N_2O emission from coal combustion in China was estimated as 12 Gg-N on the assumption that N_2O emission factors for coal combustors are as same as those in Japan.

3.3 Global N_2O emission

Annual N_2O emissions from global coal use may be estimated roughly. The global consumption of hard coal was about 3.6 Gt in 1991, of which 1Gt is used in steel manufacture and other industries. However, the temperature of coke oven is so high as 1000 deg C or more that N_2O emission may be negligible small. If 3% of the total coal use is assumed to be in fluidized bed combustion and the rest is in conventional boilers, global annual N_2O emission from coal combustion is estimate as 90-210 Gg-N. When the share of fluidized bed combustion increases in future, N_2O emission from coal combustion must increase. Fig. 4 shows a simulation of N_2O emission when fluidized bed combustion is widely used.

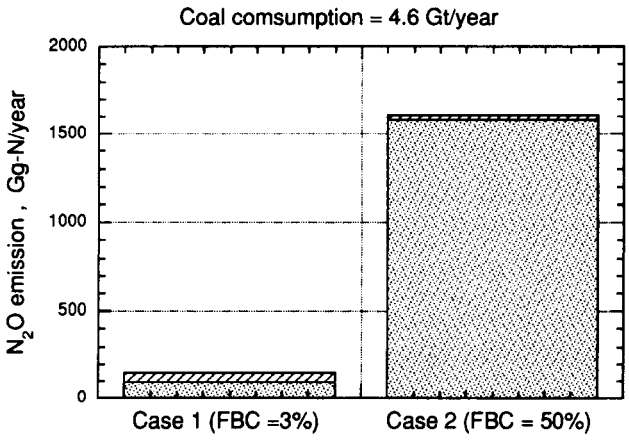

Fig. 4 Global N2O emission from coal use. Share of FBC is 3% and 50%.

4. Conclusion

The Source inventory of methane (CH_4) and nitrous oxide (N_2O) from industrial activities are studied. CH4 and N_2O emission were estimated as follow;

(1) The emission factors of CH_4 from coal mines in Japan are ranging between 12 to 116 m3 per ton of coal production. The average emission factor is 27 m3/ton. The annual total amount of CH_4 emission from coal mines in Japan is 0.22 Tg at 1989 fiscal year statistics and 0.15 Tg CH_4 was discharged into atmosphere.

(2) Total coal product in China is 1,138 Mton in 1993 and 8.8 Tg of methane is estimated to discharged into atmosphere due to the coal mining.

(3) Updated estimation of annual nitrous oxide emissions from fossil fuel combustion in Japan shows that the total N_2O emission from coal, oil and natural gas is 2.6 Gg-N in conventional combustors and 2.3 Gg-N in fluidized bed combustors. The annual N_2O emission from power plants in Japan was estimated as 1 Gg-N/year by latest data of power plants.

(4) Based on the assumption that 0.3 ppm of N_2O would be emitted from conventional coal combustion, oil and LNG combustion and 50 ppm of N_2O from fluidized bed combustion, the annual N_2O emission should be 13.7 Gg-N from whole stationary combustion sources.

(5) Total annual emission of N_2O from fossil fuel combustion in China was estimated as 12 Gg-N on the assumption that 3% of coal is burned by fluidized bed combustion. Global annual N_2O emission from coal combustion was estimate as 90-210 Gg-N from world's annual coal consumption.

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