B-2.7 Study Amounts of Methane and Nitrous Oxide Release from Sewage Treatment Plant

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Abstrust A study of amounts methane and nitrous oxide release from wastewater treatment processes was performed. The amount released from a household nightsoil treatment tank (summer and fall) was studied and the result was combined with those obtained in winter to estimate the total amount released In Japan. The behavior of the release of N_2O from fluidized bed furnace which incinerates the sludge dewatered coagurating by the polymer was studied accompanying fluctuations in the operating conditions. The actual state of N_2O release from a sludge melting furnace was also studied, but in comparison with that of other incineration processes, its emission factor could be ignored. The results already obtaind were summed up to estimate of the amounts CH_4 and N_2O released from sewage treatment plants both in Japan and throughout the world.

Key Words CH_4, N_2O,Sewage treatment, Sewage sludge,Household nightsoil treatment tank

1. Introduction

Some estimations about release sources and the amounts of CH_4 and N_2O have been made, but in fact, their range has been inflated to the point that it is impossible to estimate accurately. In order to determine the amounts of these gasses released, it will be necessary to carry out repeated abstractions of anticipated release and absorption sources and measure the released amounts, and it will also be important to clarify the release mechanisms and establish release control measures.

At sewage treatment plants, CH_4 is generated under anaerobic conditions and N_2O is produced by the nitrification and denitrification in the water treatment process and by the incineration in the sludge treatment process. But there are few examples of measurements of the release of these greenhouse effect gasses from sewage treatment plants, so it can be assumed that this is one of the missing sources.

2. Purpose of the Research

This research project was implemented to find the emission factor for CH_4 and N_2O by measuring the amount of these two substances released during each treatment process performed at sewage treatment plants and at the same time to estimate the total amounts released during sewage treatment processes within Japan and around the world.

This amounts of CH_4 and N_2O released from a household nigfhtsoil treatment tank were studied during the summer and fall and the amount of N_2O released from a fluidized bed furnace which incinerates the polymer sludge and a sludge melting furnace, then summarized the results obtained to estimate the total amounts of CH_4 and N_2O released from sewage treatment plants and from household nightsoil treatment tanks.

3. Results of the Research

(1) Amounts of methane and nitrous oxide released from household nightsoil treatment tanks

[1] Study method

At the end of 1991, 33.89 million persons, or 27.3% of the total population of Japan, lived in households with household wastewater treatment tanks and approximately 7 million of these tanks were installed. The vast majority of these, or 6.72 million, were used solely to process the nightsoil. A survey CH_4 and N_2O released from household nightsoil treatment tanks was carried out during the summer and fall.

The amounts of CH_4, N_2O, and CO_2 released from household nightsoil treatment

Table 1. Treatment condition in household nightsoil treatment tanks

Tank No.	No. 1	No. 2	No. 3
Type of tank	6-person tank	7-person tank	7-person tank
Tank capacity (m^3)	1.28	1.41	1.41
Number of users (persons)	3	4	3
Aeration (L/min) (summer)	16	24.5	28
Aeration (L/min) (autumn)	24	23.5	28

tanks were estimated by measuring the amount and the concentration of gas released from the contact aeration chambers and changes in the concentration of gas in the solid separation chambers in three household nightsoil treatment tanks consisting of a solid separtion chamber, a contact aeration chamber, and a sedimentation chamber. The densities of the DOC, IC, NH_4-N, and NOx-N in the treated wastewater were also measured. The summertime survey was done throughout the day at two hour intervals. The results of the summer survey were refered to select the times when the average data for each day was obtained. The survey in fall was done at these selected times.

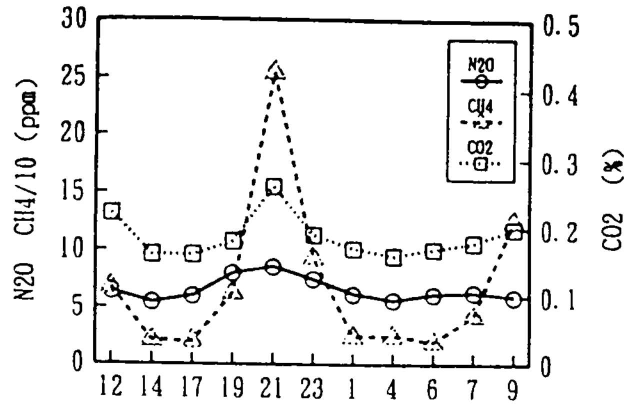

Figure 1. Change in the concentration of gas released from a contact aeration chamber (Tank No.3)

The treatment conditions of the household nightsoil treatment tanks studied are shown on Table 1. Since the winter survey, the sludge within the tanks has not been removed nor have the tanks been cleaned. The water temperature was 21℃ on the day of the summer survey and it was 17℃ on the day of the winter survey.

[2] Research Results

Figure 1 presents the changes over time in the concentration of the CH_4, N_2O, and CO_2 in the gas released from the contact aeration chamber of household nightsoil treatment tank No.3 during the summertime survey. In contrast to the stability of the CO_2 and N_2O concentration, the CH_4 concentration tended to increase just before the household members went to bed and in the morning, two periods of the day when the amount of water flowing into the system rises. It is assumed that this fluctuation in the CH_4 concentration is a result of the fact that unlike the CO_2 and the N_2O, most of which seem to be produced in the contact aeration chamber, the CH_4 is generated in the soild seperation chamber, pushed out into the contact aeration chamber by water flowing in from the toilets, and released during the gaseous phase by the aeration.

Figures 2 to 5 show the amounts of carbon and nitrogen released by the household nightsoil treatment tanks for each form of release. The amounts of substances which are dissolved in the treated wastewater and discharged with it were computed using the using water volume ratio: 50 L/capita/day.

Turning to carbon, the results showed that the amount of CO_2 discharged in summer was greater than in fall and it was also larger tan the discharge ratio of 21 gC/capita/day. It is

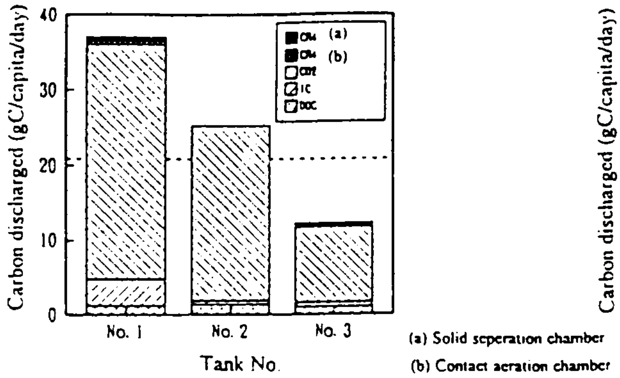

Figure 2. Amount of carbon discharged

(summer)

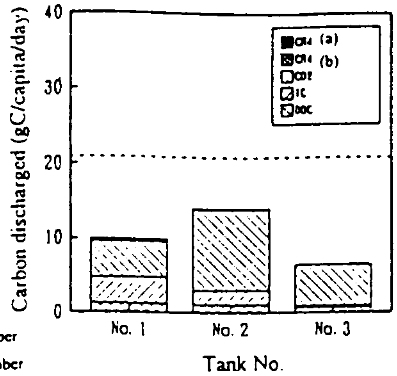

Figure 3. Amount of carbon discharged

(autumn)

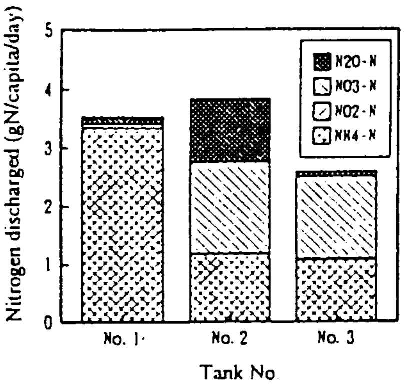

Figure 4. Amount of nitrogen discharged

(summer)

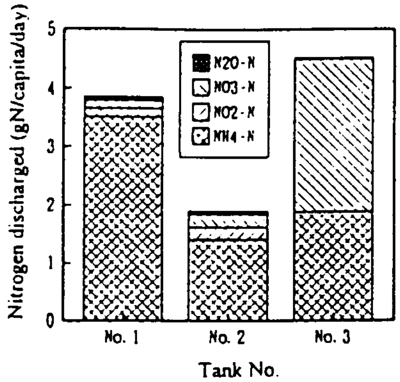

Figure 5. Amount of nitrogen discharged

(Autumn)

Table 2. Amount of CH_4 and N_2O discharged and the conversion rate for the inflow load

	C_4H				N_2O	
	Amount discharged (gCH_4/capita/day)			Conversion rate	Amount discharged	Conversion rate
	Solid separation chamber	Contact aeration chamber	Total	CH_4-C/TOC	(gN_2O/ capita/d)	N_2O-N/T-N
High water temperature period	0.37	0.60	0.97	3.5%	0.16	1.1%
Moderate water temperature period	0.13	0.30	0.43	1.5%	0.083	0.6%
Low water temperature period	—	—	0.46	1.6%	0.058	0.4%

Table 3. Amount of CH_4 and N_2O released from household nightsoil treatment tanks (Total in Japan)

Period	CH_4(Gg)	N_2O(Gg)
High water temperature period (June to Sept.)	4.0	0.7
Moderate water temperature period (April, May, Oct. & Nov.)	1.8	0.3
Low water temperature period (Dec. to March)	1.9	0.2
Annual total	7.7 Gg/y	1.2 Gg/y

oxygen concentration from 5% to 14% and the free board temperature at 730℃ and 850℃. The operating conditions were varied by fixing the amount of sludge inserted at a constant level and adjusting the fludzing air and supplementary fuel. Samples were extracted at the cyclone outlet port, and the N_2O, NOx, HCN, NH_3, O_2, CO_2, CO and the flow volume were measured. The sludge had a moisture content of 82%, was 83% volatile solids, and the nitrogen content of its solids was 5.5%.

A study of the relationship of the amount of N_2O released with the furnace interior oxygen concentration and the temperature in various locations of the furnace revealed substantial effects from the free board temperature in particular; as this temperature increased, the amount of released N_2O declined (Figure 6). This is assumed to occur because the higher its temperature, the more readily N_2O decomposes. This testing also confirms that results obtained at other fluidized bed furnaces for the polymer sludge conform closely to this figure. Here the unit for the amount of N_2O released is the conversion rate N (gN-N_2O/gN-sludge): one not susceptible to the effects of the amount of sludge inserted.

NOx on the other hand, was fully removed and was at or below the measurement limit

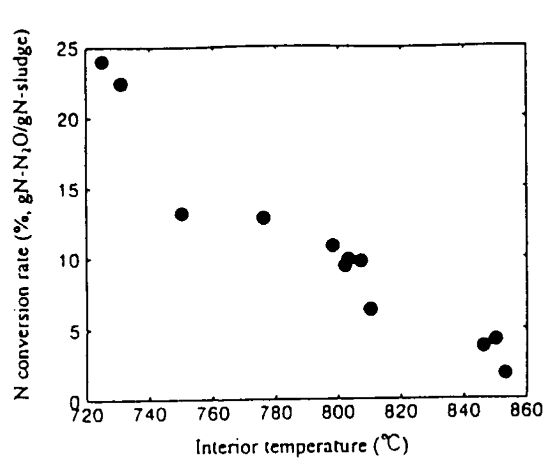

Figure 6. Interior temperature and amount of N_2O released

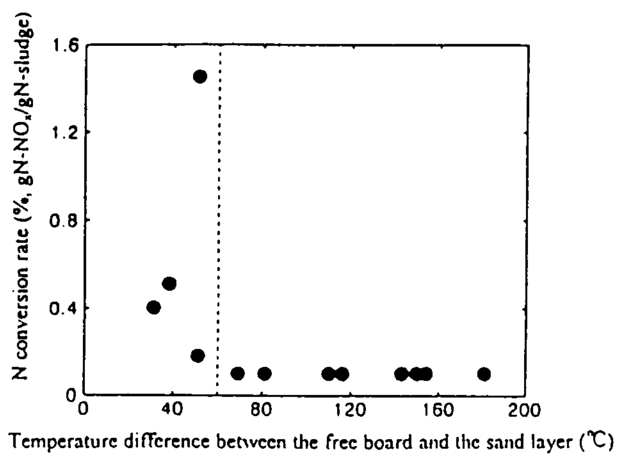

Figure 7. Temperature difference between the free board and the sand layer and the amount of NOx released

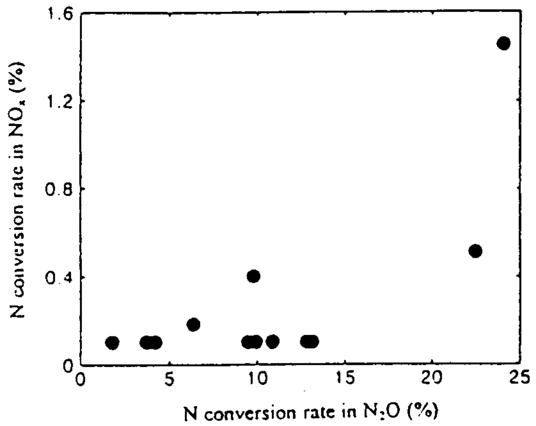

Figure 8. Relationship of the amount of NO_x released with the amount of N_2O released

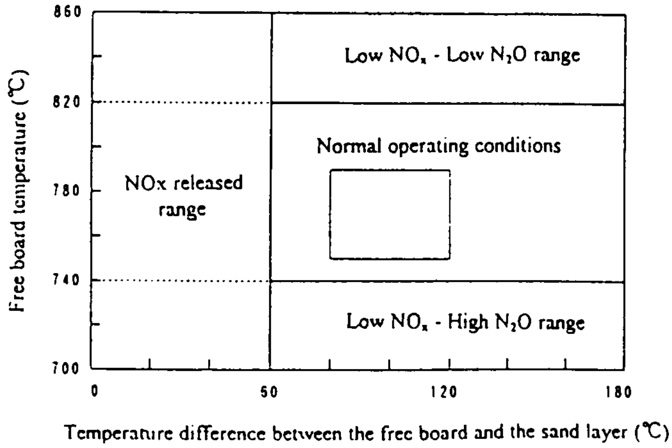

Figure 9. Release behavior of N_2O and NO_x

believed to be a result of the fact that the high water temperature causes anaerobic decomposition of the solid organic matter that has settled in the solid seperation chamber, making it soluble so that it flows into the contact aeration chamber. Because of sampling errors, part of the data concerning CH_4 in tanks 2 and 3 in the fall and for tank 2 in the summer is missing, but Table 2 shows the results of finding the amount released using measured data. More is released from the contact aeration chamber than from the soild separation chamber, and the amounts released decline as the water temperature falls between the summer and the autumn.

Before describing the nitrogen results, it should be noted that the treatment characteristics of the three tanks differ. In tank No.1 nitrification is not progressive, in tank No.2 it is progressive only in the summer, and tank No.3 it is a year-round progressive. This, it is believed, is a result of differences in the organic matter load on each household nightsoil treatment tank. Tank No.2, the one in which nitrification progresses rapidly only in the summer, releases an extremely high concentration of N_2O: approximately 100ppm. In tank 1, where nitrification does not progress and in tank 3 where nitrification is always occurring, such high N_2O concentrations were not measured, indicating the possibility that large amounts of N_2O are released when nitrification appears to be progressing rapidly. The data from the summer survey of tank No.2 was not used in later release amount estimations because it is believed to represent a special case. Table 2 shows the amount of N_2O discharged found by averaging the study data. But it is essential to point out that there is considerable uncertainty about the amount released during the period when the water temperature was high.

Table 3. shows the results of a estimation of the amount of CH_4 and N_2O released from household nighsoil treatment tanks throughout Japan based on the above data and data obtained in winter. The results of estimation of the amounts released in one year are 7.7Gg for CH_4 and 1.2Gg for N_2O.

(2) Amount of nitrous oxide released by sludge incineration furnaces

[1] Test of a working fluidized bed furnace for polymer sludge

The results of past studies have revealed that the N_2O emission factor for a fluidized bed furnace which incinerates the polymer sludge is larger than that of other types of sludge furnaces. The N_2O release behavior was investigated accompanying fluctuations in operating conditions of a working fluidized bed furnace (treatment capacity: 100tons/day), and the operating conditions that would reduce the amount of N_2O released were studied.

12 sets of furnace operating conditions were established by varying the interior

under many test conditions, but it was released when the oxygen temperature was approximately 12% or greater and the furnace interior temperature differential was at or below 60℃ (Figure 7). It is assumed that NOx produced inside the furnace is removed through its reaction with HCN, NH_3 or other reduction gasses, but it is possible to speculate that this reduction process does not function fully when there is lots of oxygen and conditions permit combustion all the way to the bottom of the furnace.

The above results show that the amounts of N_2O and NOx released are not in a negative relationship as it is usually claimed (Figure 8). Therefore, in a fluidized bed furnace for the polymer sludge, it is possible to reduce only N_2O without increasing NOx by varying the operating conditions. Specifically, it is appropriate to operate such a furnace so that only the freeboard temperature is increased while maintaining the furnace interior temperature differential at 60℃ or more (Figure 9). But raising the furnace temperature causes early deterioration of the furnace and an increase in the amount of CO_2 it releases, and it is necessary to do a reassessment from the point of view of its effect on overall global warming, costs, etc.

[2] Release from sludge melting furnaces

In 1992, 3.2% (in terms of solid base) of all sludge produced during the year was processed by sludge melting, and this proportion will continue to increase. For this study, the amount of N_2O released from 2 typical types of melting furnaces (cokes bed furnace and surface melting furnace)was tested.

The results are presented on Table 4. The emission factor for sludge melting furnaces is generally estimated to be $10gN_2O$/ton, but this is about 1/100 of that of fluidized bed furnace for the polymer sludge, a size which can be ignored when computing the total amount released. Reasons for the low value of the emission factor for sludge melting furnaces include the high interior temperature that decomposes N_2O and the fact that they process includes lime sludge (or a basicity adjustment agent).

Table 4. Amount of N_2O released from melting furnaces

Treatment plant	Type of furnace	Treatment capacity (t·DS/day)	Amount of sludge inserted (t·cake/hour)	Furnace interior tenperature (℃)	Properties of the gas emitted from furnace			Emission facter (gN_2O/t·cake)
					O_2(%)	NO_x(ppm)	N_2O(ppm)	
A	Cokes bed	10	polymer type ;1.5 lime type ;0.7	1,300	4.4	82	3.1	16.6
B	Surface melting	25	polymer * type ;4.0	1,300	10.0	94	1.3	7.3

* Lime mixed in as a basicity adjustment agent

4. Estimating the amount of methane and nitrous oxide released from sewage treatment plants

(1) Amount Released Within Japan

The total amount of these substances released during 1992 was estimated from the results already obtained. The emission factor for CH_4 was the average value from the winter and summer. The N_2O emission factor from wastewater treatment processes considered only the aeration tank. This value varied according to the degree of progress of the nitrification, but the average values were used in the same manner. The N_2O emission coefficient from the sludge treatment considered only the sludge incineration, and the emission factor was applied to four types categorized by furnace shape and by the coagulating agent used.

The results of the estimation are shown on Table 5. The following items must be kept firmly in mind.

Table 5. Estimation of amounts of methane and nitrus oxide released in Japan (1992)

Greenhouse effect gasses	Amount of treated wastewater or incinerated sludge	Emission facter	Range of emission facters	Amount released (t/y)
CH_4	11 X 10^9 m^3/y	730 $mgCH_4/m^3$	260～1,210	8,030
N_2O(Wastewater treatment)	11 X 10^9 m^3/y	10 mgN_2O/m^3	1～20	110
N_2O(Sludge treatment)	3.39 X 10^6 t/y	* gN_2O/t	330～1,200	2,830

* Fludized bed furnace for the polymer sludge:1,200, Multistage incinerator for the polymer sludge:750, Lime type:300, Others:750

1) The accuracy of the estimations of the N_2O released from the sewage treatment process and of the CH_4 is considered low because the emission factors are rough average values, and in the case of N_2O in particular, the emission factor fluctuates widely in response to slight differences in conditions during the sewage treatment process, so it is essential to carry out further study of this matter, including the clarification of the formation mechanism.

2) CH_4 produced during the anaerobic digestion process is ignored on the assumption that all of it is converted to CO_2.

But estimates of the amount released by the EPA and other organizations only look at CH_4 produced from the anaerobic digestion process, and they employ a

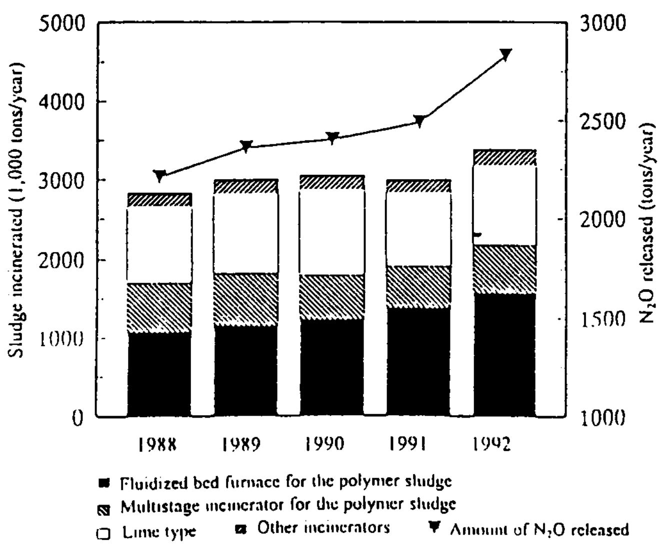

Figure 10 Amount of sludge incinerated and changes in the amount of N_2O released

substantially different estimation method. And two of the treatment plants studied practice anaerobic digestion.

3) Most of the N_2O released is produced by sludge incineration and the annual rate of increase in the amount of N_2O released through incineration is calculated at roughly 5%. This is a result of an increase in both the total amount incinerated and in the proportion of fluidized bed furnaces for the polymer sludge (Figure 10).

(2) Amount released world wide

The estimation was done ignoring the following items.

Table 6. Basic statistical data used to estimate the amounts released around the world

Countries studied	Population (10,000 person)	Sewer coverage rate (%)	Amount of sludge produced (10^3tDS/y)	Percent incinerated (%)
Canada	2,652	70	–	–
USA	24,998	74	5,570	14
N. Zealand	339	42	–	–
Austria	771	72	250	37
Belgium	985	23	–	–
Denmark	514	98	150	28
Finland	499	76	–	–
France	5,644	68	900	20
Germany	7,948	86	2,750	10
Greece	1,005	10	–	–
Iceland	26	6	–	–
Ireland	350	11	–	–
Italy	5,766	61	800	11
Luxembourg	37	90	–	–
Netherlands	1,494	93	280	10
Norway	424	57	–	–
Portugal	1,053	21	–	–
Spain	3,896	53	–	–
Sweden	856	95	–	–
Switzerland	671	90	250	20
Turkey	5,869	1	–	–
UK	5,724	87	1,500	5
Total	71,521	–	–	–
Entire world Percentage	529,200 13.5%			

Population: United Nations, population in 1990 based on the Population Statistics Annual.
Sewer coverage rate: OECD Environmental Data 1993.
Sludge incineration: Only U.S.: UNEP, Environmental Data Report 1993 to 1994, Other countries: Korrespondentz Abwasser 1993. 1, 40. Jahrgang.

1) Wastewater discharged into the environment without processing.

2) That wastewater processed in sewage treatment plants, but included in statistical data than can not be used.

3) The effect on the environment of treated wastewater and disposed sludge

The countries included in the study include only those OECD countries whose statistical data can be utilized. Table 6 shows the populations, sewerage system coverage rates, amounts of sludge produced and the incineration rates for each country whose data is usable. It was assumed that the ratio of wastewater discharged was 620 L/capita/day and the water content of the dewatered sludge was assumed to be 75%. Table 7 presents the results of the estimation.

The emission factors for the N_2O from wastewater treatment processes and for CH_4 were the same as those used to compute the totals for Japan. The emission factor for N_2O from sludge treatment that was used is 750 gN_2O/ton, the average value for a multistage incinerator for the polymer sludge, but the rate in Japan is high, reflecting the hight incineration rate. An increase in the sludge incineration rate is forecast for many countries other than Japan as prohibition of the ocean dumping of sludge , beef up landfill regultions, and so on. This will be followed by a rise in the amount of N_2O released.

Table 7. Estimation of amounts of methane and nitrus oxide released worldwide (1990)

Greenhouse effect gasses	Amount of treated wastewater or incinerated sludge	Emission facter	Amount released (outside of Japan)	Amount released worldwide
CH_4	106 × 10^9 m^3/y	730 $mgCH_4/m^3$	77 Gg/y	85 Gg/y
N_2O(Wastewater treatment)	106 × 10^9 m^3/y	10 mgN_2O/m^3	1.1 Gg/y	1.2 Gg/y
N_2O(Sludge treatment)	6.44 × 10^6 t/y	750 gN_2O/t	4.8 Gg/y	7.6 Gg/y

5. Summary

1) The amounts of CH_4 and N_2O released from household nightsoil treatment tanks during the summer and autumn were studied, combined with results in winter, and a estimation was done of the amount of these substances released in Japan.

2) The release behavior of N_2O was studied as the operating conditions of a working fluidized bed furnace for the polymer sludge were varied to clarify the sand layer and free board temperatures that control the amounts of N_2O and NOx released.

3) The actual state of N_2O release from a sludge melting furnace was studied. The emission factor, which was about 10 gN_2O/ton, was small enough that unlike the emission factors for other types of incineration furnaces, it can be ignored.

4) All the results obtained were summarized and used to estimate amount of CH_4 and N_2O released from sewage treatment plants both in Japan and throughout the world.

Sources

1) U.S. EPA: Inventory of U.S. Greenhouse Gas Emissions and Sinks; 1990- 1995, EPA, 230-R-94-014, September 1994.