B-16.6 Technological measures for controlling CH_4 and N_2O from domestic wastewater treatment by using a bioengineering and ecoengineering system

Contact Person Inamori Yuhei
Head, International Water Environment Renovation Research Team
Regional Environment Division
National Institute for Environmental Studies
Environment Agency
Onogawa 16-2, Tsukuba, Ibaraki, 305-0053 Japan
Tel: +81-298-50-2400 Fax: +81-298-50-2560
E-mail: inamori@nies.go.jp

Total Budget for FY 1995-FY1997 67,280,000 Yen (FY1997; 22,779,000Yen)

Abstract

The final goal of this project study is, to develop and establish the effective technology for controlling the greenhouse gases, CH_4 and N_2O, emitted through the wastewater and its treatment process. The DO controlled intermittent aeration activated sludge method with coagulant added can be used to prevent the discharge of N_2O at the same time as it effectively removes nitrogen and phosphorus to prevent eutrophication. It is also clear that they can respond to a certain degree of load fluctuation. The process is, therefore, considered to be an extremely effective way to prevent global warming and to improve water environments. The emission characteristics of CH_4 and N_2O varies in the difference in swine waste treatment system greatly. The accumulation of NO_2-N in nitrification process happens easily in swine waste treatment, and leads to increase N_2O emission. Therefore, to control emission of N_2O with proceeding with nitrogen removal, promotion of nitrification and denitrification is important by the combination of anoxic and aerobic treatment process under the proper condition. CH_4 emission from the wetland varied under different vegetation, and that showed daily, and seasonal change. Five strains of CH_4 oxidizing bacteria were isolated from the wetland sediment and soil

Key Words Greenhouse Effect Gas Reduction, Methane (CH_4), Nitrous oxide (N_2O), Bioengineering, Ecoengineering

1. INTRODUCTION

It has been pointed out that, of the various kinds of environmental problems which have appeared as a consequence of the rapid growth of the world's population and industrial development, global warming is a problem with great potential to create ecosystems in poorer condition than any we have previously seen and to even threaten the survival of the human race. Global warming is caused by the excessive discharge of greenhouse effect gasses such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) into the atmosphere as a result of human activities. Of these greenhouse effect gasses, It is considered that the greenhouse effect per one molecule of CH_4 is 20-30 times, and that of

N_2O is 200-300 times, as large as that of CO_2. And because it is also known that wastewater treatment processes discharge N_2O, there is an urgent need to both clarify the mechanism of N_2O generation in wastewater treatment processes and to develop technologies to prevent this phenomenon. N_2O is also produced by microbiological metabolism of nitrogen compounds in the natural environment. CH_4 is well known as a typical gas produced in bottom sediment of a swamp or a lake in which organic compounds are accumulated. Therefore, a wetland which is artificially polluted by wastewater or other organic pollution load is suspected as an important source of atmospheric CH_4. For these reasons, it is of much importance to develop the technologies that can control CH_4 and N_2O emission from wastewater and its treatment process.

2. RESEARCH OBJECTIVE

Following studies were made through the project term, FY 1995- FY1997.

(1) Development of the advanced wastewater treatment system that can achieve both control of N_2O emission and eutrophication

(2) Advanced swine waste treatment process that can suppress N_2O emission

(3) Investigation of CH_4 emission in polluted wetland and the technology for its preventive countermeasure

3. RESULTS and DISCUSSION

3.1 Examination of the development of the advanced wastewater treatment system that can achieve both control of N_2O emission and eutrophication

This study was undertaken to develop a highly efficient nitrification and denitrification and phosphorus removal reactor that can simultaneously reduce the generation of N_2O and improve the nitrogen and phosphorus removal functions during domestic wastewater treatment processes in order to prevent eutrophication.

The first step was to compare the N_2O discharge situation and nitrogen removal capabilities of continuous aeration and intermittent aeration at a domestic wastewater treatment plant practicing the activated sludge method, and perform analysis to evaluate the effects of introducing anoxic conditions to a bioremediation system. The following five paragraphs summarize the results of this work.

1) The intermittent aeration method reduced both the quantity of N_2O generated and its conversion ratio to a level far below the levels observed using the continuous aeration method.

2) Most of the N_2O generated was discharged into the atmosphere as part of the aeration process with very little N_2O discharged in dissolved state.

3) The quantity of N_2O discharged by intermittent aeration was calculated as between 0.43 and 1.89 g-N_2O $person^{-1}$ $year^{-1}$.

4) It was possible to nitrify the incoming NH_3 completely to NO_3^- by keeping the DO concentration inside the aeration tank during the aerobic process to at least 0.5 mg l^{-1}. And an anoxic process of 60 minutes was sufficiently long for the denitrification reaction to be completed.

5) The results have suggested that it is extremely important to appropriately combine and control various aerobic and anoxic processes in order to improve nitrogen removal capabilities and to restrict the discharge of N_2O.

These facts suggest that it is essential to evaluate and analyze factors that contribute to the generation of N_2O in each reaction that occurs during nitrification and denitrification. And because a large quantity of N_2O was generated during aerobic processes, it is necessary to study the generation of N_2O during the nitrification reaction in particular. So with particular attention paid to the biological nitrification reaction, the effects of the DO concentration, the pH value, and the accumulation of NO_3^- in water on the N_2O generation reaction were studied and evaluated. The following is a summary of the results obtained (Figs. 1 and 2).

1) When HRT was maintained for 8 hours, it was possible to conduct the nitrification reaction sufficiently regardless of the DO value and the NO_3^- accumulation. And the rate of discharge of N_2O into the atmosphere in the low NO_3-N concentration range remained almost constant regardless of the DO value, but in all systems, the emission rate increased as NO_3^- accumulated, and the higher the DO value of the system, the higher the emission rate.
2) The nitrification reaction proceeded sufficiently regardless of the pH value of the water with a nitrification rate of 90% or greater obtained in all experimental systems, and even the speed of nitrification was not effected. And the rate of discharge of N_2O into the atmosphere under pH 7.0 and pH 6.0 conditions was constant regardless of the DO value, but if the pH value dropped to 5.0, the generation rate increased abruptly in all systems. This clearly indicated that a low pH has a big influence on the increase in the generation of N_2O. These results clearly demonstrate that pH is an important management factor and that preventing the fall of the pH value is an extremely important way to prevent the discharge of N_2O.

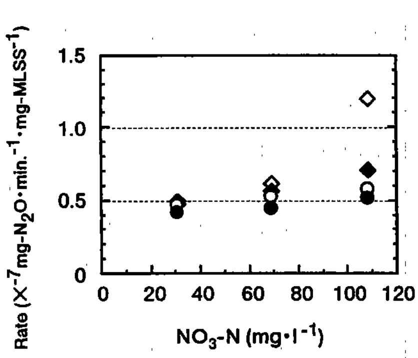

Fig. 1 Influence of DO and NO_3-N accumulation on N_2O emission rate

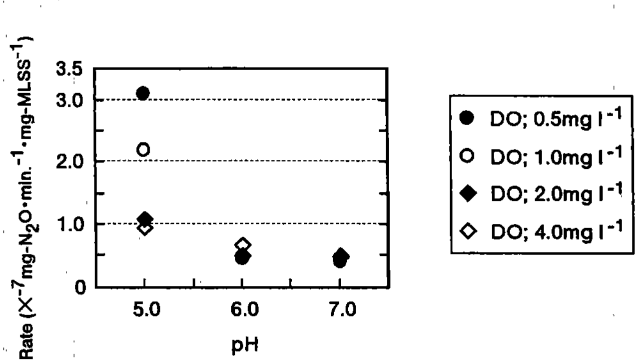

Fig. 2 Influence of DO and pH on N_2O emission rate

The next step was a basic study and evaluation based on laboratory experiments of the optimum operating conditions for the aerobic and anoxic processes in the intermittent aeration activated sludge method that is a typical aerobic/anoxic treatment process. A comparative study of the timer control method and DO control method of setting the timing of the aerobic/anoxic phases of intermittent aeration was performed in order to evaluate their nitrogen removal capabilities and N_2O generation prevention capabilities. With the timer control method, 60 minutes of aerobic processing and 60 minutes of anoxic processing is considered a single cycle that is repeated over and over again. The DO control method on the other hand involves monitoring the DO concentration inside the aeration tank and, based on the results, determining the time allotted to each aerobic and the anoxic process during each cycle. The following summarizes the results obtained from these studies (Fig. 3).

1) The results clearly demonstrate that the DO control method restricts the generation of N_2O and removes nitrogen far more effectively than the timer control method.
2) The results also confirm that the DO control method can efficiently perform the nitrification and denitrification reactions, preventing the generation of N_2O if the aerobic and anoxic conditions inside the aeration tank are appropriately controlled.

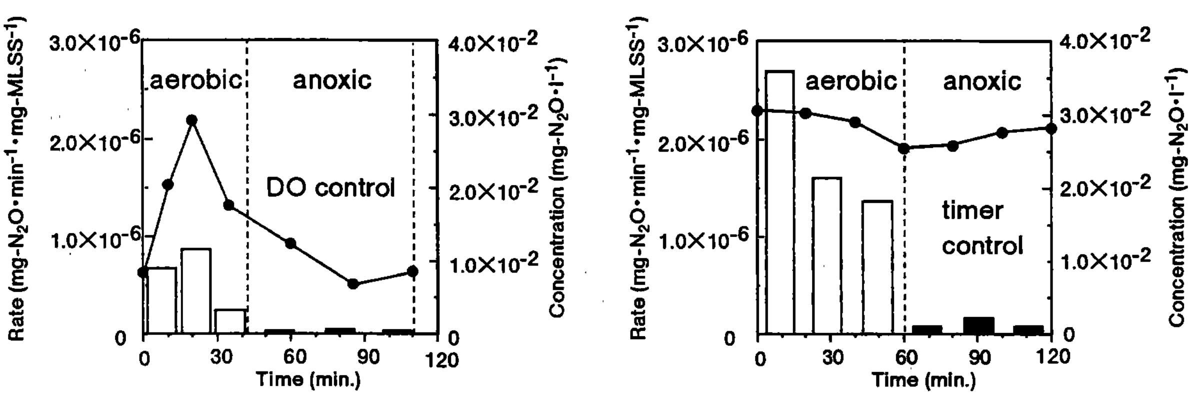

Fig. 3 Time course of the N_2O emission rate into the atmosphere and the dissolved N_2O concentration under each intermittent aeration

In conclusion, the application at an actual plant of the N_2O discharge prevention type DO controlled intermittent aeration activated sludge method (hereafter referred to as "the process") was studied. A full scale pilot plant using a DO controlled intermittent aeration activated sludge method with coagulant added was used for this experiment in order to achieve simultaneous N_2O generation prevention and nitrogen removal, and to link these with phosphorus removal: a process that limits eutrophication. And nitrogen removal capability, N_2O discharge properties and prevention capability including the nitrification and denitrification reactions during domestic wastewater treatment were studied along with treatment capabilities under incoming nitrogen overload conditions. The wastewater treatment facilities are shown in Fig. 4. The results were studied in order to establish optimum nitrogen removal and N_2O discharge prevention conditions for actual domestic wastewater treatment plants. The following is a summary of the results obtained (Table 1, Fig. 5).

1) Under correct nitrogen load conditions, the discharge of N_2O into the atmosphere by the process during the aerobic and anoxic phases is 244.2 and 60.9 mg day^{-1} respectively in all bioremediation reaction tanks, and the qu

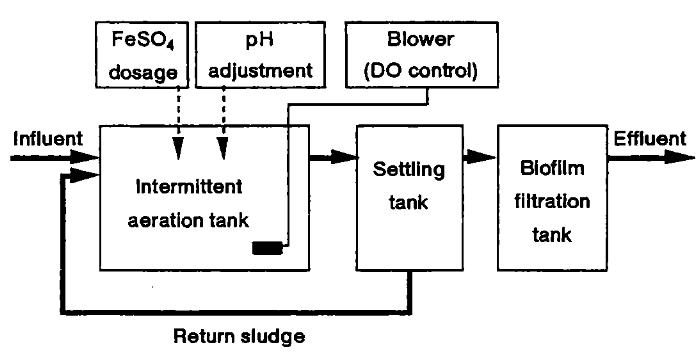

Fig. 4 Scheme of pilot plant for coagulant dosed and DO controlled intermittent aeration activated sludge process.

antity discharged outside the process in dissolved form was 92.1 mg day^{-1}.

2) It was confirmed that the process can effectively reduce the quantity of N_2O discharged and the N_2O conversion ratio during continuous aeration activated sludge treatment by performing optimum control of the aeration agitation based on DO monitoring.

3) Using the process, under a nitrogen load of 0.021 kg-T-N kg-MLSS^{-1} day^{-1} or less, the N_2O conversion ratioes per incoming and removed nitrogen were found to be approximately 0.05% and 0.06% respectively, but when the load was boosted to 0.035 kg-T-N kg-MLSS^{-1} day^{-1}, the values increased sharply to 0.66% and 1.13%.

4) The results have shown that when the incoming nitrogen load had been increased, under load conditions higher than the level where the nitrogen removal efficiency and nitrification efficiency decline, the quantity of N_2O discharged increases.

5) It was clearly shown that as long as treatment is performed under appropriate load conditions permitting the achievement of advanced nitrogen removal, it is possible to simultaneously prevent the discharge of N_2O.

Table 1 Comparison of the treatment efficiency among several domestic wastewater treatment process.

Treatment process	N_2O emission ($mg\text{-}N_2O\ m^{-3}$)	Nitrogen removal efficiency (%)	Conversion ratio to N_2O * (%)	Conversion ratio to N_2O ** (%)
(a) Coagulant dosed and DO controlled intermittent aeration	24.8	84.4	0.05	0.06
(b) Timer controlled intermittent aeration (aerobic/anoxic =30min./60min.)	20.4	75.6	0.05	0.06
(c) Continuous aeration	535.8	5.1	1.23	24.3

Notes:
* Ratio from the nitrogen influent to N_2O-N.
** Ratio from the removed nitrogen to N_2O-N.

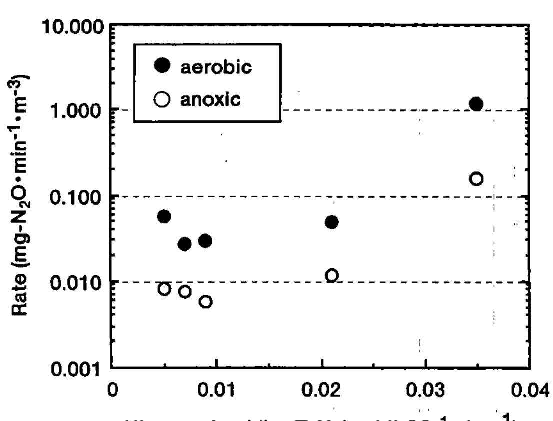

Fig. 5 Influence of nitrogen load on N_2O emission into the atmosphere

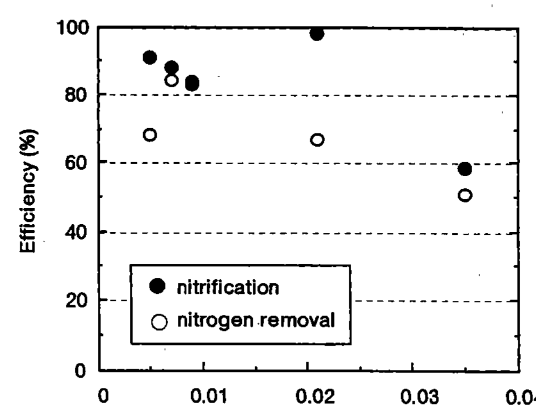

Fig. 6 Influence of nitrogen load on nitrogen treatment efficiency

In sum, the DO controlled intermittent aeration activated sludge method with coagulant added can be used to prevent the discharge of the greenhouse effect gas N_2O at the same time as it effectively removes nitrogen and phosphorus to prevent eutrophication. It is also clear that they can respond to a certain degree of load fluctuation. The process is, therefore, considered to be an extremely effective way to prevent global warming and to improve water environments.

3.2 Advanced swine wastewater treatment process that can suppress N_2O emission

The purpose of this study is to establish the countermeasure technology for the CH_4 and N_2O suppression which occurs in the treatment process of the waste from livestock breeding. We investigated the amount of emission of CH_4 and N_2O in the swine waste treatment process, and studied about N_2O suppression technology.

3.2.1 The investigation of the amount of emission of CH_4 and N_2O in swine waste treatment process

The swine waste treatment system of the pig farm was shown in Fig. 7. Excreta was utilized as compost, and urine was treated by intermittent aeration lagoon system. The amount of wastewater from pigsty is about 100 m^3 per day. The amount of polluted water injected in one cycle becomes about 33 m^3, because 3 cycles were carried out in a day. BOD of raw water ranged from 2,000-4,000 mg l^{-1}. The concentration of T-N was 1,000-1,500 mg l^{-1}, and most of nitrogen was NH_4-N. As for the treated water, good condition was shown from April until October, and BOD was 100 mg l^{-1}, T-N was about 20 mg l^{-1}. It was found that not only organic substance but also nitrogen was removed highly by intermittent lagoon system. But treatment became worse from autumn in winter, and gas collection was carried out in this period. The concentration of N_2O in the gas was detected about 100 ppmv. That concentration was maintained during aeration process. The amount of aeration in lagoon was 30,000 m^3 day^{-1} as N_2O of 5.9 kg was emitted per a day. The discharge coefficient became 0.6 g-N_2O day^{-1} $head^{-1}$, and the value of 3.6 % could be obtained as a conversion ratio.

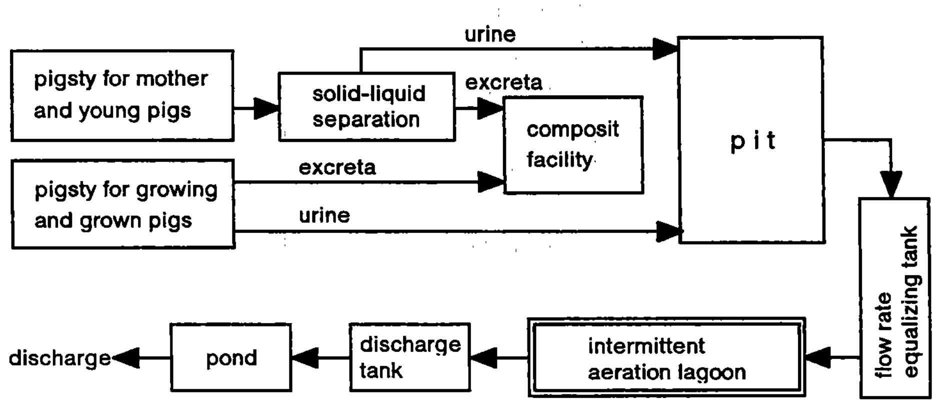

Fig. 7 Swine wastewater treatment system

3.2.2 N_2O suppression technology in swine wastewater treatment process

Sequencing batch nitrification and denitrification process for swine waste treatment was studied. Activated sludge was collected from the actual treatment facilities, and used after acclimation. The initial MLSS is 4700 mg l^{-1}, and a COD load is 1.0 kg m^{-3} day^{-1}. The reactor of the cylinder shape with a diameter of 10 cm has an effective capacity of about 5 l and an effective height of 55 cm. 4 experimental systems were carried out, control system (continuous aeration), 8-4 system (8-hours aeration and 4-hours non-aeration), 6-6 system (6-hours aeration and 6-hours non-aeration), 4-8 system (4-hours aeration and 8-hours non-aeration). Tested wastewater is actual pig urine.

The cycle of anoxic condition and aerobic condition was changed, and the emission tendency of N_2O was examined in this experiment. Experiment was done in 4 systems such as control system, 8-4 system, 6-6 system, and 4-8 system. Treatment characteristics were shown in Fig. 8. As aeration became long, NH_4-N showed a tendency of decreasing. This was because nitrification occurred, and the accumulation tendency of NO_2-N in continuous aeration was remarkable. The decline of pH occurred caused by nitrification. The accumulation of NO_2-N was not recognized by 6-6 system though the accumulation of NO_2-N was recognized as for 8-4 system. The emission of N_2O was remarkable in the control system with the progress of nitrification as shown in figure 7. Compared with the other 3 systems, about 1.3 times of the amount of N_2O was emitted in control system. The conversion rate ranged from 11.8 to 18.6 % in these systems. The reason why the emission of N_2O is big in swine wastewater treatment is that NO_2-N is easily accumulated in nitrification process. The promotion of nitrification and denitrification by the combination of the proper anoxic and aerobic condition was important to control the emission of N_2O with progress of the nitrogen removal. Intermittent aeration lagoon system is one of the proper technology to satisfy above condition. The characteristics of this system is that BOD and nitrogen load is lower than that of usual activated sludge process and so on. We think that to develop suppression technology more advanced control of operation is necessary with attempting technological introduction such as automatic control.

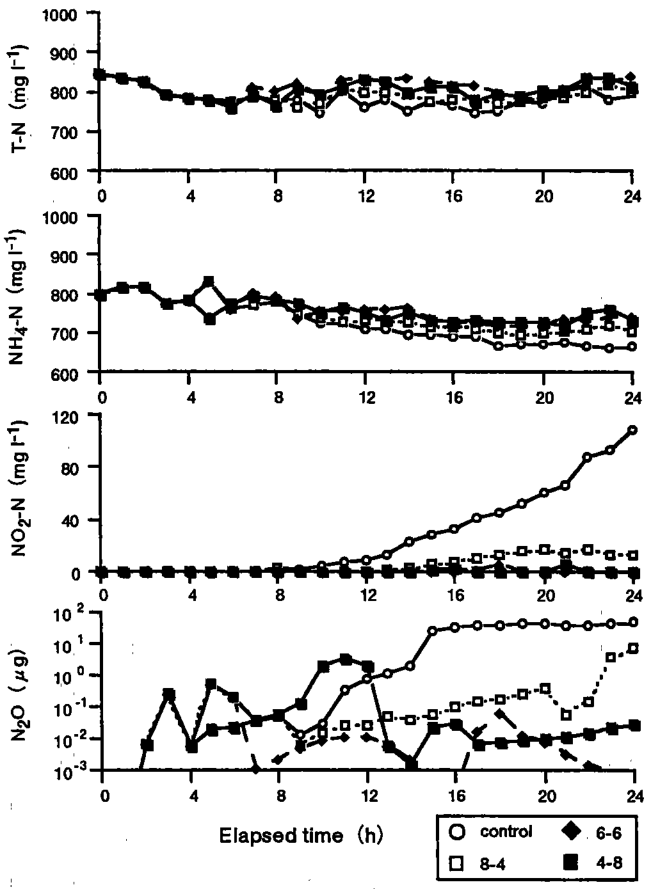

Fig. 8 Changes in water quality itens and N_2O gas

3.3 Investigation of CH_4 emission in polluted wetland and the technology for its preventive countermeasure

In this study, the methodology for investigation of CH_4 and N_2O emission was developed and the CH_4 emission was surveyed in a typical Japanese plain wetland. The mechanism of and affecting factors for the CH_4 emission were also researched and discussed. To establish a countermeasure to reduce CH_4 emission from a wetland, ecological engineering by using wild plants and utilization of microbial activity in the nature were also fundamentally investigated.

3.3.1 Developing the method for and surveying the emission of CH_4 and N_2O from a wetland

Main objective of this study was to establish effective method for measuring flux of methane and nitrous oxide from wetlands to the atmosphere, and to survey those emission quantitatively from an artificially polluted wetland. To discuss mechanism of the methane production and emission from a wetland, analysis the relationship between the methane flux, vegitation zone and characteristics of the soil or bottom sediments was performed.

First of all, we made the survey of CH_4 emission quantitatively from an artificially polluted wetland. The amount of CH_4 gas detained in the Izunuma Lake sediment within a day was different with vegetation zones (Table 2). At the area of sweet flag vegetation at which organic substance content of the bottom sediment was not high, the largest amount of CH_4 was detained, comparing with less amount of CH_4 at the area of lotus vegetation where the organic content of the sediment was the highest. The detained CH_4 gas showed seasonal change in each vegetation. It increased at autumn and decreased at winter.

Table 2 Relationship between the detained CH_4 and the vegetation

vegetation	CH_4 detained in the bottom sediment (mg-CH_4 sediment-m^{-2} day^{-1})			
	May 14-15	Aug. 7-8	Oct. 16-17	Dec. 18-19
sweet flag	18	112	129	0
lotus	22	26	69	9
none	9	15	41	0

The survey of CH_4 emission by using plastic chambers was done once a month through a year. In the reed zone, the most dominant vegetation of the outskirt wetted-land of Izumuna Lake, CH_4 emission was not high (Fig. 9). In the water oat which is secondary dominant vegetation, the largest emission of CH_4 was observed especially in autumn. The daily periodic change in CH_4 emission and absorption were also observed by this measurement. The maximum emission of CH_4 was 86mg m^{-2} day^{-1} at the water oat vegetation zone.

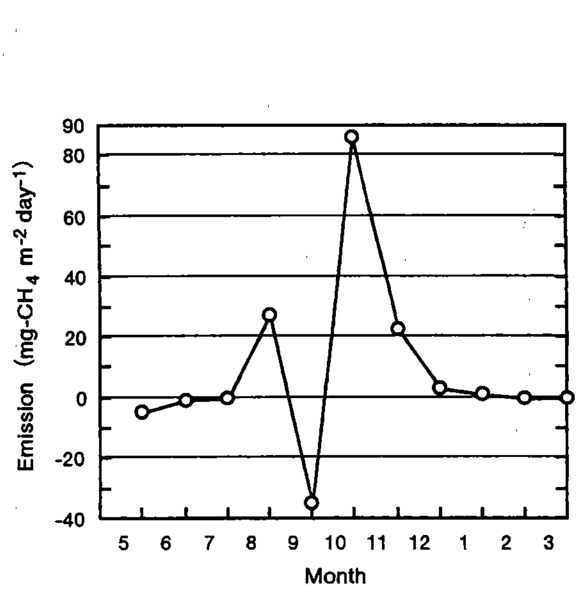

Fig. 9 CH_4 emission from the water oat grown outskirt wetted-land

The survey of CH_4 emission from aquatic surface of Izunuma Lake was done once a month through a year by using floating plastic chambers. The data showed that the larg est amount of CH_4 was at vegetation zone of lotus which was the most dominant veg etation in the lake was about one sixth of t hat at wetted-land of water oat zone (Fig. 10).

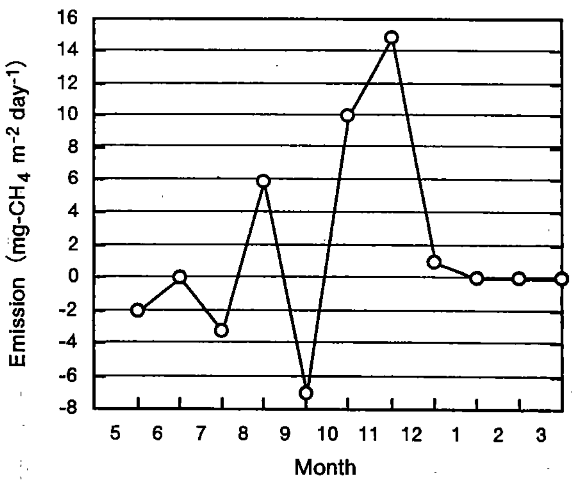

Fig. 10 CH_4 emission from the lotus grown aquatic surface

3.3.2 Analytical data of the characteris tics of soil and lake sediment and consideration about CH_4 emissio n from a wetland

Second objective of this study was to have an enrichment culture of indigenous methane oxidizing bacteria and to isolate t hem from a wetland. The content of organ ic substance in the wetted land soil or in t he lake sediment commonly increased in t he autumn, because almost all vegetation was withered and supplied into soil or sediment as organic substance in autumn. This is a reason why emission of CH_4 increase in autumn. The total amount of organic substance supplied to soil or sediment is determined by primary production of plants in the wetland. If the wetland polluted artificially and nutrient salts inflowed into the wetland, the primary production is enhanced. Therefore, the most fundamental way for the control of CH_4 emission from wetland polluted artificially is water pollution control especially of nutrients for vegetation.

3.3.3 Finding CH_4 oxidizing bacteria and obtaining the bacterial culture

If the preventive countermeasure for CH_4 emission is required, utilization of microbial CH_4 oxidizing activity in a wetland and plant ecological engineering are potentially adaptable. To realize these biotechnological countermeasure, five strains were isolated from the wetland sediment and soil. However, these CH_4 oxidizing bacteria were not easily grown in a pure culture. Mixed enriched culture with heterotrophic was much better to get active biomass for CH_4 oxidation. The enrichment culture should be applied in future research for development of ecological engineering for the control of CH_4 emission from wetlands.

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