# 16.6.1 Development of CH<sub>4</sub> and N<sub>2</sub>O Emission Control Using Eco-engineering Systems Such as Wetland, Soil Trench and Oxidation Pond (Final Report)

#### **Contact Persons**

Yuhei INAMORI

Head, International Water Environment Renovation Research

Team

Division of Regional Environment

National Institute for Environmental Studies,

16-2 Onogawa, Tsukuba, Ibaraki, 305-0053, Japan

Tel: +81-298-50-2400

Fax: +81-298-50-2570 E-mail: inamori@nies.go.ip

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#### Abstract

Constructed wetland is an important eco-engineering countermeasure against water pollution for China because of its low cost, energy and maintenance requirement. But it has been increasingly recognized as a source of  $CH_4$  and  $N_2O$  emissions.

In this study, the possible resource and reason of CH<sub>4</sub> and N<sub>2</sub>O emissions was analysis on the basis of literature review and a comprehensive research framework to improve existing wetlands or develop new technique was proposed. As the key step for research implement, the design of wetland is also discussed here.

At the same time, a field experiment was carried out and the seasonal change of CH<sub>4</sub> and N<sub>2</sub>O emissions was also investigated in this study.

Results from field experiment showed the removal efficiency of pollutants and  $CH_4$  and  $N_2O$  emissions had closed relationship with season change. The removed  $BOD_5$  for a certain wetland has some definite relationship with  $CH_4$  and  $N_2O$  emissions in constructed wetland and it could be used to forecast  $CH_4$  and  $N_2O$  emissions in some occasions.

Keywords Constructed wetland, CH<sub>4</sub> and N<sub>2</sub>O emissions, Design, Greenhouse gases

### 1. Introduction

According to the report of Intergovernmental Panel on Climate Change(IPCC) in England<sup>[1]</sup>, greenhouse gases will result in quick temperature raise of 0.03°C per year in next century. As a result, the atmosphere temperature will increase 3°C till the end of next century. This will give adverse effect to people's life even result in the collapse of the ecosystem. So it has become very urgent to control the emission of greenhouse gases.

Among the greenhouse gases, the single molecular absorption of infrared radiation by  $N_2O$  and  $CH_4$  are 200-250 and 21times as much as that of  $CO_2^{[2]}$ . In less than 10 years after 1980, the contribution of  $N_2O$  and  $CH_4$  increase rapidly from 18% for nearly one hundred years before 1980 to 24%. It is deduced that  $CH_4$  and  $N_2O$  will play more and more important role in the future and it will be more effectively to

control CH<sub>4</sub> and N<sub>2</sub>O for their higher relative greenhouse effect potentiality.

As far as the contribution of greenhouse gas emission is concerned, wastewater treatment process was estimated to account for 7% of total CH<sub>4</sub> emission and 19% of total N<sub>2</sub>O emission [3]. As the largest developing country in the world, China has been responsible for 10.2% of total greenhouse gas emission in the world [4], at the same time it was faced serious problem of water pollution. With the popularization of wastewater treatment, the control of CH<sub>4</sub> and N<sub>2</sub>O emission will become another environment crisis in China. This aspect of work should begin as soon as possible by selecting the promising wastewater treatment technology in China.

China has more than 479 cities and over 2,000 county towns <sup>[5]</sup>. Most municipal sewage from these cities and towns has not been effectively treated due to the tight budget. Constructed wetland seems to be a very promising eco-engineering technology for China. Relatively low cost, energy and maintenance requirements make it preferable alternatives for the treatment of numerous, distributed and relatively small-scale waste streams that occur in spacious rural areas which are very typical in China. Furthermore, it could be a supplement measure for natural wetland restoration to supply the living space for various kinds of animals and plants.

The first wetland treatment system in China was built in 1982 in the southwestern suburb of Shenyang, Liaoning Province [6]. During the period of the seventh five-year plan, several pilot-scale land treatment systems were set up and sponsored by the Chinese Environmental Protection Agency as state research projects. At present, various type of wetland or hybrid system of wetland and other kind of land treatment system are being built or in operation in different climate regions of China. Table 1 gave the general review of wetland system in China. Those projects showed that wetland had rather high buffer capability and good performance in rather wide variation of hydraulic and pollutant loading and will be very prospective in future.

As mentioned before, sustainable operation of wetland treatment systems depends on high rate conversion of organic and nitrogenous loading to gaseous end products including CH<sub>4</sub> and N<sub>2</sub>O. However, Up to now, there is no reference about the emission of CH<sub>4</sub> and N<sub>2</sub>O in constructed wetland, and no studies about the control of them in constructed wetland either. It has become very instant to make clear the emission of N<sub>2</sub>O and CH<sub>4</sub> in the constructed wetland and to develop the appropriate controlling technology.

Site	Feed	Capacity	Area	Comments	Ref.
		$(m^3 \tilde{U} d^{-1})$	(m <sup>2</sup> )	/Special Feature	
Tianjin City	Settled domestic wastewater	1,800	200,000	Consists of natural wetland, FWS Wetland, reed bed SF wetland and stabilization pond	5,7,8
Dagang Oil Field in Tianjin	Settled domestic wastewater	2,000	ı	Infiltration type wetland using salty sea sandy soil	Ċ
Qinghe District of Beijing	Effluent of stabilization pond (domestic wastewater)	120	1	Consists of reed bed SF wetland and fish pond	'n
Changping County of Beijing	Settled Domestic wastewater	2,000	10,000	Consists of reed bed SF wetland and fish pond	'n
Yanshan, Beijing	Effluent from oxidation ditch (Petrochemical Wastewater)	4,000	270,000	Consists of reed bed SF wetland and stabilization pond, using mountain land	ڼن
Weifang City; Shandong Province	Settled Domestic Wastewater	180,000	3,000,000	Using Coastal Saline-alkali Soil	<b>ب</b>
Zhuchen City; Shandong Province	Settled Domestic Wastewater	30,000	43,000	Consists of stabilization Pond and reed bed SF wetland	'n
Jiaonan City; Shandong Province	Settled Domestic Wastewater	000'09	495,000	Consists of 11 FWS wetlands and a sluice pond, using coastal tide zone	1
Jiaozhou City; Shandong Province	Mixed wastewater (1/3 domestic + 2/3 industrial)	30,000	1,000,000	Consist of stabilization pond and FWS wetland	10
Bannikeng of Shenzhen City, Guangdong province	Screened domestic wastewater	3,000	8,400	Consists of gravel bed wetland and fish pond	6
Leping of Shanshui City	Effluent from anaerobic hydrolysis pond (Pig raising farm wastewater)	80	500	Consists of gravel bed wetland and fish pond	6
Yantian Industry Area of Shenzhen City	Effluent from hydrolysis or aquatic pond (Industrial wastewater)	1,000	1,200	Consists of up-flow anaerobic hydrolysis pond, aquatic plant pond and gravel wetland	6
Conghua County of Guangzhou	Effluent form hydrolysis pond (Domestic wastewater)	500	200	Consists of upflow anaerobic hydrolysis pond and gravel wetland	6
Baiyan coal mine of Sichuan Province	Mixed wastewater of acidic pit and domestic wastewater	1,000		Consists of stabilization pond and gravel bed wetland	

#### 2. Research Framework

Two basic types of constructed wetlands according to their water level inside are now utilized commonly—free water surface (FWS) wetland and subsurface flow (SF) wetlands. It is the combination of saturated soil, plants, and microorganisms that provides both aerobic and anaerobic conditions for the removal of contaminants and encourages nutrient removal (Especially nitrogen) from the overlying wastewater. During the course of contaminant removal in wetlands, two processes will be involved in the production of N<sub>2</sub>O and CH<sub>4</sub>. The first is the stabilization of organic matters in the zone lack of oxygen. A large amount of CH<sub>4</sub> could be produced in this course [12]. The second is the removal of nitrogen. In soils, both nitrification [13,14]—oxidation of ammonia to nitrate and denitrification [13]—the anaerobic reduction of nitrate to gaseous forms of N, are responsible for the N<sub>2</sub>O production. Nitrification was though to produce more nitrous oxide than denitrification [15]. Moreover, under aerobic soil conditions that favor nitrification, soil pores are generally open and allow diffusion of gaseous products to the atmosphere, whereas in conditions that favor denitrification diffusion [16] is impeded and further transformations e.g. N<sub>2</sub>O to N<sub>2</sub> may occur.

Use the data from natural wetland and paddies for reference, soil water content <sup>[18]</sup>, soil porosity<sup>[16]</sup>, soil redox potentials<sup>[20]</sup>, water-soluble carbon source<sup>[19]</sup>, water soluble nitrogen source<sup>[17]</sup>, pH and soil temperature<sup>[18]</sup> are all indicated to be able to influence N<sub>2</sub>O and CH<sub>4</sub> emissions from soil. Among them, redox potential seemed to be most important. It was found that the redox potential range for CH<sub>4</sub> and N<sub>2</sub>O emissions was contradictive to each other and it result in the difficulty of realizing N<sub>2</sub>O controlling, CH<sub>4</sub> controlling and nitrogen removal at the same time.

If the redox potential was located in the range of higher redox potential for CH<sub>4</sub> control, the emissions of nitrous oxide will increase. If keeping on increasing the redox potential to a higher redox potential, the emissions of N<sub>2</sub>O could be reduced but the removal of nitrogen will be retarded for the lack of anoxic environment for denitrification. Several practical observations testify the existence of this contradiction [22,23].

Plant is another important factor that influences the emissions of CH<sub>4</sub> and N<sub>2</sub>O<sup>[21]</sup>. Aquatic plants transport oxygen to the soil, simultaneously provide a significant pathway for the gas emission. Some plants could also assimilate the nitrogen in the soil. So using different plants might result in very different gas emission. As the media for microorganisms' growth, the structure of root and its oxygen transportation will also affect the microbial activity greatly.

Accordingly, this research is carried out to make clear the mechanisms of greenhouse gases emission from two typical kinds of constructed wetlands and their contribution to the globe warming; And to find the optimal operation parameters or measures to reduce  $CH_4$  and  $N_2O$  emissions.

Two aspects of work will be included in this research. One is lab-scale experiment using small-scale constructed wetland. This part could be divided into 3 steps as shown in figure1. At first, The comparison of two kinds of wetlands will be accomplished in these lab scale instruments. The comprehensive assessment of these two wetlands will be made according to their ability of contaminant removal and potentiality of non-CO<sub>2</sub> greenhouse gases. Provided with the basic data, the most appropriate technologies under different conditions could be obtained.

Then the operational parameters of the constructed wetland will be changed to investigate

their influence on the performance of wetland. At the same time, different kinds of plants will be selected according to the structure of roots and leaves. Upon these results, the best structure and the appropriate operational condition for the constructed wetland will be got.

Finally, on the basis of the above results, a new technology with low even without non-CO<sub>2</sub> greenhouse gases emission wetland technology for domestic wastewater treatment will be developed and constructed in pilot scale.

Another aspect of experiment is carried out in field. This part of work is mainly to get some basic data about  $CH_4$  and  $N_2O$  emissions. A constructed wetland in Jiaonan City of Shandong province in China was selected as experiment site in this research.

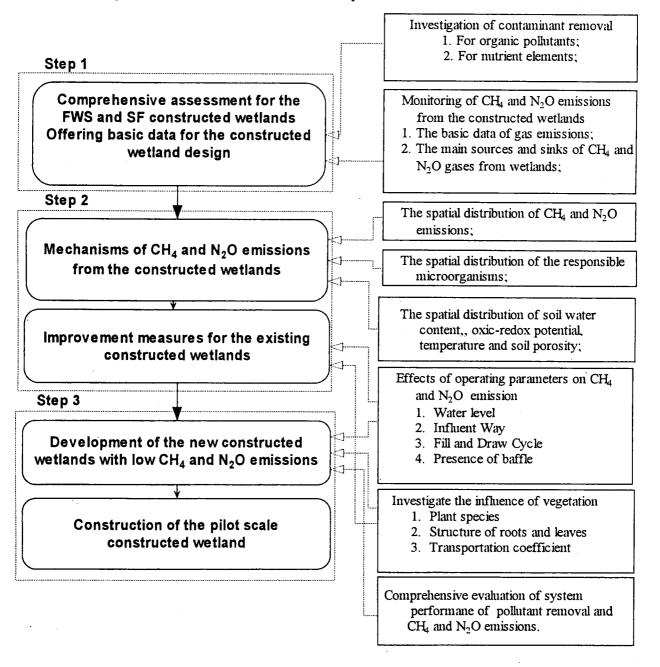
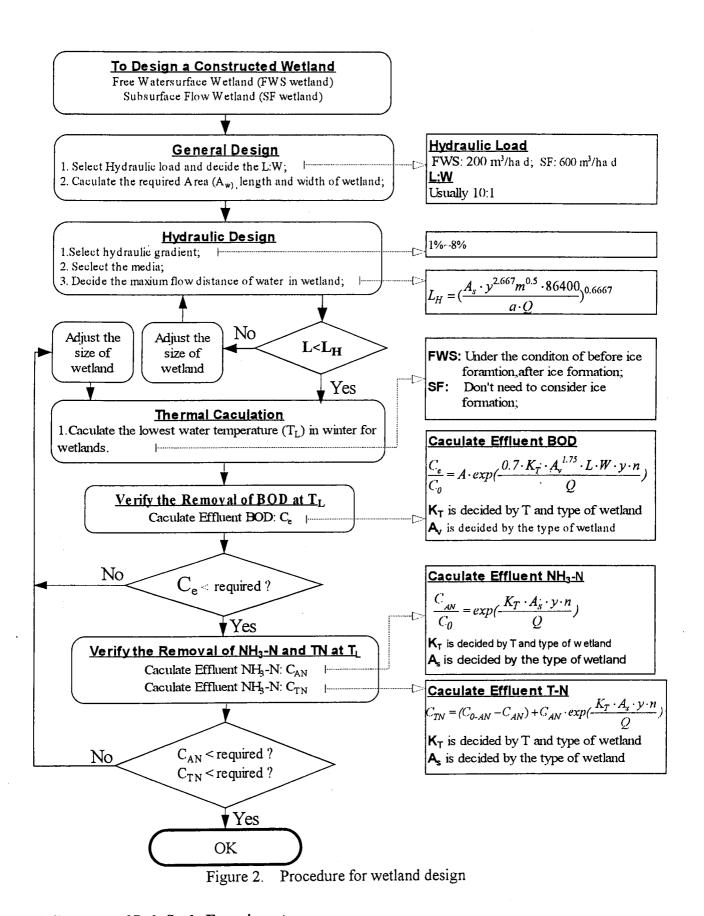


Figure 1 Framework for the lab-scale research



3. Progress of Lab Scale Experiment

To offer the basic data for N<sub>2</sub>O and CH<sub>4</sub> emissions in constructed wetland, typical wetland that could represent the status of wetland application in China should be designed. According to Table 1, the constructed wetlands in China consist of two basic kinds of constructed

wetlands- SF wetland and FWS wetland, and most of the SF wetlands are reed bed SF wetlands. Using their typical parameters as reference, two lab scale of constructed wetlands, one in SF mode and the other in FWS mode were designed and constructed in this research.

During the course of design, no guidelines and principles for manipulating in practice could be got. After including existing references as far as possible, the common procedure was summarized in figure 3.

The key point for constructed wetland design could be summarized as worst condition control. At first, wetland should achieve planned goals of organic pollutants, nitrogen and phosphorous removal. All constructed wetland systems can be considered to be attached growth biological reactors, and their performance can be estimated with first –order plug flow kinetics for BOD and nitrogen removal. Since the biological reactions involved in treatment are temperature dependent, it is necessary to estimate the water temperature in the wetland. The final design should meet the requirement for the most critical pollutant removal even in the extreme case in winter. The major design variables, which include plant selection, hydraulic loading rates and hydraulic gradient was quoted from references [8,24,25]. Now the two wetlands have been finished at the site of Kokiru wastewater treatment plant.

## 4. Results of Field Experiment

#### 4.1 Material and Methods

A full-scale free water surface constructed wetland for municipal wastewater treatment in Jiaonan City of China was selected as experimental site. It was constructed in August 1998 using tided zone land. Effluent from the wetland was drained into Yellow Sea through floodgate during ebb-tide period. With 11 units of  $150 \times 30$  m<sup>2</sup>, it has capacity of 60,000 m<sup>3</sup>/d and HRT of 6 days. Macrophytes like reed, bulrush and duckweed were planted in the wetland. Animals like frogs, ringdoves and bitterns also roosted in it.

The schematic diagram of the wetland was shown in figure 4.

The influent of the wetland is the domestic wastewater from Jiaonan City. The wastewater quality was shown in Table 2.

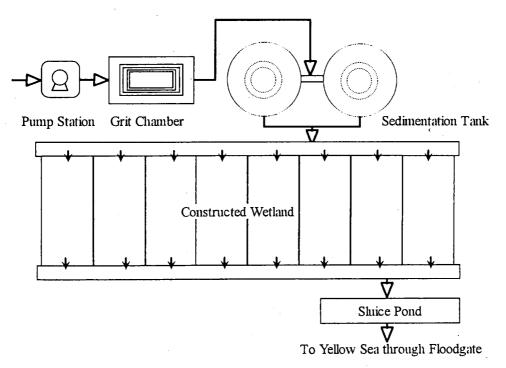


Figure 3 Schematic diagram of constructed wetland in Jiaonan City of China

Table 2 Quality of wastewater used in the experiment

		<u> </u>				
Items	T (°C)	BOD5 (mg·L <sup>-1</sup> )	COD (mg·L <sup>-1</sup> )	T-N (mg·L <sup>-1</sup> )	$NH_3-N$ $(mg\cdot L^{-1})$	T-P (mg·L <sup>-1</sup> )
Conc.	5~ 28.5	355~ 477	628~ 851	23.22~ 28.78	12.21~ 18.21	3.37~ 4.75

Emissions of N<sub>2</sub>O and CH<sub>4</sub> were sampled using closed chamber (0.5m×0.5m×0.45m). Concentrations of BOD, COD, NH3-N and T-N for influent and effluent were determined according to standard method every month.

#### 4.2Results

# 4.2.1 Seasonal change of N<sub>2</sub>O and CH<sub>4</sub> emissions

As shown in Fig.4, with the time entering autumn and winter, the water temperature decreased from 23.5°C to 0.6°C, and the removal efficiency of BOD, and NH3-N decreased correspondedly, especially that of NH<sub>3</sub>-N. These two processes have closed relationship with microorganisms, and their low activity at low temperature resulted in the drop of BOD<sub>5</sub> and NH<sub>3</sub>-N removal. The removal of T-N and T-P didn't fluctuate markedly. This implied that some other process like

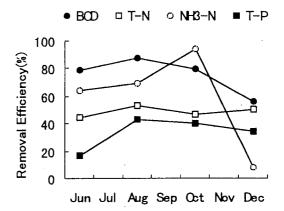


Fig.4 Seasonal change of pollutant removal adsorption also contributed to T-N and T-P removal.

## 4.2.2 Seasonal change of $N_2O$ and $CH_4$ emissions

As shown in Fig.5, CH<sub>4</sub> emission rate in summer was larger in summer than in autumn and winter, but N<sub>2</sub>O emission showed the contrary tendency and even was negative in summer. It indicated that the control of CH<sub>4</sub> emission and that of N<sub>2</sub>O emission in wetland contradictive to each other. This phenomenon is also reported by other researcher [26]. Both aerobic and anaerobic conditions exist in wetland. Anaerobic condition with soil redox value of <150mv

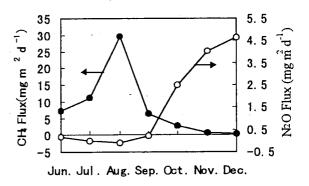


Fig. 5 Seasonal change of N<sub>2</sub>O and CH<sub>4</sub> emissions

was critical for CH<sub>4</sub> emission, but soil redox value of >400mv or between -200 and 300mv will also result in high  $N_2O$  emission [20]. It explains the contradiction in Fig. 1. At the same time, under anaerobic condition of <0mv, N<sub>2</sub>O emission will decrease.

# 4.2.3 Influence of removed BOD, on gas emissions

As shown in Fig. 6, with the increasing of removed BOD<sub>5</sub> amount, the CH<sub>4</sub> emission increased, originally slow and gradually fast and the  $N_2\text{O}$  emission decreased, originally fast and gradually slow. The reaeration through water surface or plants and the oxygen consumption through organic degradation determined the aerobic condition of soil. The point when CH<sub>4</sub> emission was first become zero could regarded as the balance point for oxygen reaeration and consumption. In Fig.2, it was around 220mg·L<sup>-1</sup>. After that, the soil transfer to anaerobic condition, CH<sub>4</sub> emission increased and N<sub>2</sub>O emission decreased. This implied that removed BOD<sub>5</sub> value could be helped to estimate gas emission in soil during practical operation.

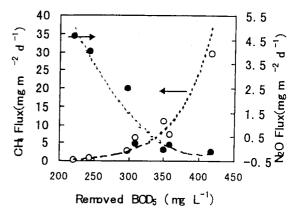


Fig 6 Influence of removed BOD<sub>5</sub> on N<sub>2</sub>O and CH<sub>4</sub> emissions

#### 5. Conclusions

For wetland design, a procedure according to the worst condition control was proposed in this study.

Results from field experiment showed the removal efficiency of pollutants and  $CH_4$  and  $N_2O$  emissions had closed relationship with season change. The removed  $BOD_5$  for a certain wetland has some definite relationship with  $CH_4$  and  $N_2O$  emissions in constructed wetland and it could be used to forecast  $CH_4$  and  $N_2O$  emissions in some occasions.

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