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Greenhouse gas emissions from a constructed wetland for municipal sewage treatment

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Abstract: The fluxes of greenhouse gases (methane and nitrous oxide) emission from a constructed wetland in the Eastern China as municipal sewage treatment were measured from June 1999 to August 2000 by the closed chamber method. The constructed wetland for municipal sewage treatment is a significant source of methane, up to 976.6×10^6 g CH_4/a, which was emitted from the constructed wetland with the area of 495000 m^2 and wastewater loading rate of 12000 m^3/d. Its daily mean methane flux reached 5.22 g $CH_4/(m^2 \cdot d)$, 250 times as much as that in natural wetland in the same latitude region. 227.8 mg CH_4 was produced from the treatment of 1 liter wastewater, up to 700—1000 times as much as that in the secondary treatment. The emission of nitrous oxide from the constructed wetland is not higher than that from secondary treatment of wastewater, only 0.07 mgN_2O/L.

Keywords: greenhouse gas emission; constructed wetland; municipal sewage; methane; nitrous oxide

Introduction

CH_4 and N_2O produced from municipal sewage system, as important part of anthropogenic emission of greenhouse gases into atmosphere, have attracted worldwide attention in recent years because of their association with global warming. As a result of this, numerous studies have been conducted on the emissions and controlling factors of various greenhouse gases from various ecosystems in the past decade. However, it should be pointed out that most of these studies were carried out in agricultural system, which is commonly regarded as the largest anthropogenic contributor to global greenhouse gas budget. Though emission from waste management systems was also documented lately, the studies mainly focus on greenhouse gases emitted from conventional sewage treatment facilities, like activated sludge system (Hanaki, 1992; He, 1999). Few literature reported greenhouse gas emission from natural sewage treatment system such as land application and stabilization pond systems (Tai, 2000).

Nowadays wetland and other land application systems are widely applied in China and other developing countries as innovative/alternative sewage treatment processes because of their economic, social and environmental benefits. To carry out a research on greenhouse gas emission from such a natural sewage treatment system is undoubtedly of significance to this research field.

Wetland is an important source of greenhouse gases (Prieme, 1994), and contribute 109×10^{12} g/a of methane to atmosphere, accounting for 21% of the total methane emission from the surface of the earth (Karen, 1993). Constructed wetlands, as sewage treatment facilities, is supposed to produce greenhouse gases, such as CH_4, N_2O, CO, and CO_2 in a larger amount than natural wetland because the contents of organic carbon and nitrogen in constructed wetland may be 10 times as much as those in natural wetlands. From this standing point, this research intend to investigate the CH_4 and N_2O emissions from constructed wetland for municipal wastewater treatment, the results will be a great help to estimate the general greenhouse gas emissions from land application system.

1 Description of research site

The wetland system for municipal wastewater treatment was established in Jiaonan County, which located in the southwest coast of Shandong Peninsula(120°00′E, 35°55′N) of China. The population in the urban area is about 100000. Besides on traditional agriculture, most residents live on travel service, food

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processing and paper-making. The major pollutants in municipal sewage come from the paper-making and food processing.

Jiaonan Town is situated in the hilly land near the Yellow Sea. The climate is warm with an annual average temperature of 12.2℃. The lowest temperature is −1.9℃ monitored in January, and the highest is 26.0℃, observed in August. The precipitation is about 775.6 mm mainly occurring in June, July and August. The humidity is high, around 73% in average per year.

The wetland application system was established in August, 1998. To save farmland, a dam was built in a tidal zone to form a freshwater wetland. The key parameters of design and operation of the project were shown as follows: (1) Total area of wetland: 495000 m^2; (2) Wastewater loading rate: 12000 m^3/d; (3) Quality of influent: COD < 1200 mg/L; BOD < 500 mg/L; SS < 1300 mg/L; (4) Quality of effluent: COD <400 mg/L; BOD < 180 mg/L; (5) Water depth: 15—25 cm, velocity of flow: 2 cm/min; (6) Vegetation: reed (phragmites autrulis), bulrush (Acorus calamus), duckweed (lemna minor); (7) Biomass above the earth's surface: 6000 kg/(hm^2·a) (contain nitrogen 1.19%); (8) Animals: frogs, ingdove and bittern and so on.

The constructed wetland consists of 1200 plots, which is 150 meters long and 30 meters wide. The municipal sewage was pumped into the wastewater treatment plant for pretreatment, and then drained into the constructed wetland plots through water-distribution system. The effluent was discharged into a storage pond temporarily, finally discharged into the Yellow Sea through a floodgate during ebb-tide period. The procedure is illustrated in Fig.1.

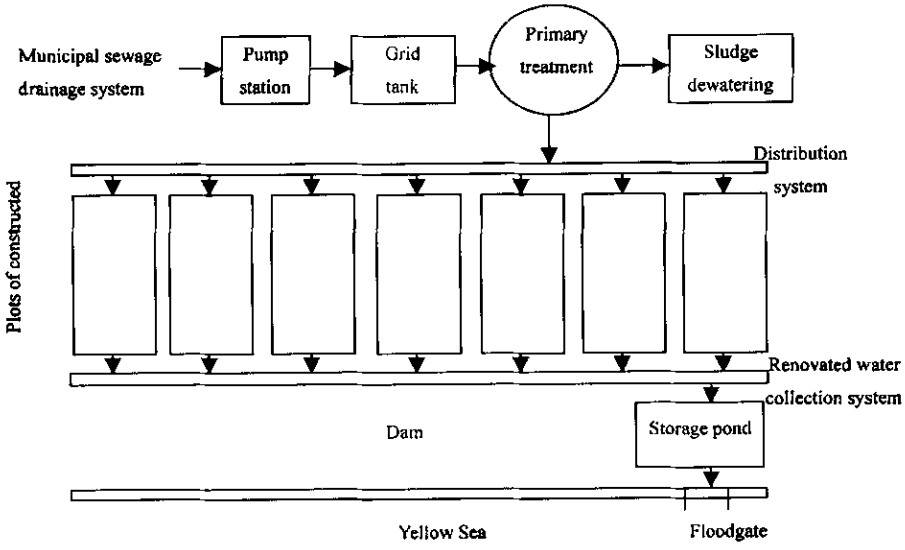

Fig.1 The schematic map of constructed wetland for municipal sewage treatment in Jiaonan Town

2 Materials and methods

2.1 Experimental design and sampling

A typical plot in the constructed wetland system was used as experimental site. Along the direction of water flow, five sampling sites (A, B, C, D and E in Fig. 2) were selected.

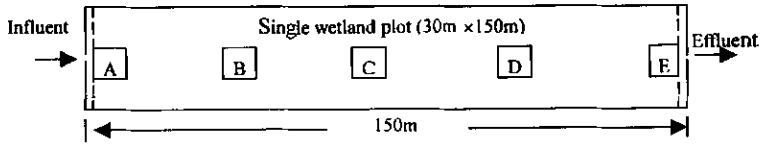

Fig.2 Distribution of sample sites in experimental plot

A closed chamber shown in Fig. 3 was used to collect air samples over the surface of wetland system (Czepiel, 1993). A thermometer was inserted into the chamber to measure the air temperature at time of sampling. To guarantee the uniformity of air quality inside the chamber, a battery-operated fan was installed on the roof of chamber to circulate air within the chamber. To overcome the vacuum effect produced when inhaling air, an aluminum bag was used to adjust the pressure. The edges of the sampling chambers were placed in troughs of a movable brackets under the water surface. The bottom of chamber was immersed into water by 50 mm. The air samples were pumped into sampling bags through the sampling port. The interval time of each sample is 4 hours during the sampling day. The closed chambers will be moved away from the measurement site, to recharge the fresh air in the chamber.

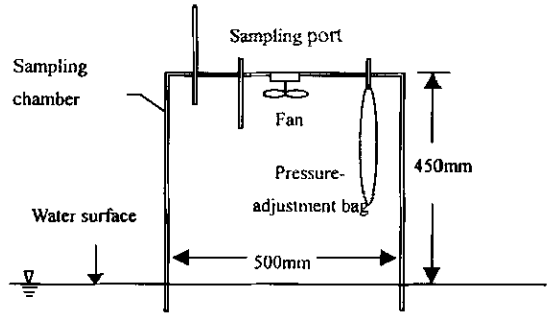

Fig. 3 Diagram of the sampling chamber (Size: 500 mm × 500 mm × 450 mm)

A head space technique was used in preparation of air samples. 50 ml wastewater was drawn into a 100 ml syringe, leaving a head space of 50 ml. Two milliliters of 1% $HgCl_2$ solution was then added for sterilization. After shaking thoroughly, the syringe was laid up for 1 hour at room temperature. The resulting gas phase in the syringe was collected as a gas sample (Kimochi, 1998a; 1998b).

During air sampling period, water samples were simultaneously collected in the corresponding sites where the chambers were placed. Air and water temperatures were recorded. BOD, COD, T-N, NO_3-N, NH_4-N, and T-P in water samples were determined.

2.2 Analysis of CH_4 and N_2O

Methane was analyzed on a gas chromatograph (GC-SP2305E, Shimadzu Co., Japan) implemented with an flame ionization detector and Poropak Q column, using nitrogen as carrier gas. Temperatures of oven and injector port were 140℃ and 180℃, respectively. Nitrous oxide was analyzed on a gas chromatograph equipped with an electron capture detector and Poropak N (1.0m) and Poropak Q columns (3.0m), using 30 ml/min argon containing 5% methane as carrier gas. Temperatures of the detector and oven were 300℃ and 60℃, respectively.

2.3 Calculation of greenhouse gas emission rate

The emission rates of the greenhouse gases from the surface of wetland were calculated from this following equations:

$$F = \frac{\Delta m \varepsilon_0}{A \times \Delta t} = \frac{\rho \times V \times (\Delta C - \Delta C_0)}{A \times \Delta t} = \rho \times h \times \frac{\Delta C - \Delta C_0}{\Delta t}.$$

Where ρ is gas density; Δm (or ΔC) is change of gas mass (or concentration) inside chamber during Δt; h, A and V are the height, bottom area and volume of the chamber respectively.

The concentration of methane in wastewater of wetland were calculated from this following equations:

$$C = \Delta C \cdot V_H / V_W.$$

Where ΔC is the methane concentration, V_H and V_W are the volume of the headspace and wastewater inside syringe.

3 Results and discussion

3.1 Diurnal change of methane emission in the constructed wetland

The samples were retrieved monthly from June 1999 to Aug. 2000. Each time, 6 samples with 4 hours intervals were collected during the sampling day. The diurnal variation of methane flux from the constructed wetland is irregular, differing from that in paddy rice field (Chen, 1995) or natural wetland (Karen, 1993). The maximum fluxes and second maximum fluxes occurred at 4:00, 8:00, 12:00 or

16:00 randomly (Table 1), having no correlation with the air temperature. It might be that the metabolism of methanogenic bacteria were affected not only by bio-rhythm itself, water-temperature or sunlight, but also by the irregular influence with variety of water quality. In particular, the wastewater with poisonous pollutants from paper mill or chemical industries could change the activities of microorganism remarkably.

Table 1 Diurnal change of CH_4 emission from the constructed wetland ($mgCH_4/(m^2 \cdot h)$)

Time	January	February	March	April	May	June	July	August	September	October	November	December
0:00	0	0	22.0	93.7	211.1	300.6	363.8	1005.0	255.4	128.9	25.9	11.7
4:00	0	0	27.9	98.2	156.6	279.0	539.6	1511.4	288.4	128.8	18.5	3.8
8:00	0	0	25.7	101.4	139.3	249.0	490.4	1403.4	315.4	110.2	19.6	12.0
12:00	0	0	16.3	125.3	252.7	341.6	465.2	1533.6	221.0	81.6	42.5	23.9
16:00	0	0	25.7	121.4	200.3	330.0	587.0	832.0	261.4	139.9	52.0	29.6
20:00	0	0	22.9	113.8	232.3	315.8	323.6	1113.0	259.2	114.9	45.3	9.7

* Greenhouse gas emission stop, because water in the wetland freeze in January and February in Jiaonan County.

Table 2 Change of methane emission from different sites in the constructed wetland ($gCH_4/(m^2 \cdot d)$)

Date	Air temperature, ℃	A	B	C	D	E
January*	-1.9	-	-	-	-	-
February*	0.0	-	-	-	-	-
15 - March	5.0	1.26	0.65	0.38	0.28	0.22
15 - April	11.4	6.95	2.66	1.22	1.21	1.00
15 - May	17.3	8.91	5.32	3.87	3.13	2.60
16 - June	21.6	17.69	8.40	4.56	2.78	2.89
16 - July	25.5	29.52	11.10	5.05	5.19	4.53
15 - August	26.0	57.12	30.67	24.01	19.36	16.81
14 - September	23.2	11.72	6.63	5.78	4.89	3.00
15 - October	15.2	7.46	3.13	1.92	1.03	0.54
15 - November	7.6	2.30	1.25	0.54	0.09	-0.10
14 - December	0.6	1.99	-0.15	0.10	-0.05	-0.07
Average	12.1	12.08	5.80	3.95	3.16	2.62

* Greenhouse gas emission stop, because water in the wetland freeze in January and February in Jiaonan County

3.2 Methane flux from different sites in the constructed wetland

Throughout the experimental period, methane flux of site A where is more closed to water entrance, is higher than other sites. And the fluxes of methane in different sampling sites decreased along the direction of flow (A-B-C-D-E) (Table 2). It indicated that emission of methane was closely related to BOD. In other words, BOD is a key factor controlling methane emission in the constructed wetland. Most of methane was produced in the upper-reach section from 0 to 60 meters (including sites A and B), near the entrance of sewage. Because of the higher loading rates of BOD and SS in this section, anoxic environment might be formed (Table 3). The anoxic/anaerobic condition is beneficial to the growth of methanogenic bacteria.

The concentration of methane in wastewater from different sites (include influent and effluent) was measured from August to December, the samples were taken at 14:00 of the sampling days. The result is shown in Table 4.

Methane concentration in raw wastewater and influent (after pretreatment) was very low (25 μgCH_4/L), but it increased rapidly when sewage was drained into wetland, and peaked at site C or D. It proved that substantial amount of methane was generated during wetland treatment processes. And in different sections from influent to effluent, methane generation varied significantly. Most of methane was produced in upper-reach sections of the wetland system (from site A to site B), this experimental result match with the measuring flux of methane from the wetland. Low methane concentration in effluent was resulted from release of methane during the running of the wetland system. While water flow through a waterfall, between the water level of the wetland and collection system dyke, it was aerated again. Aside from this release effect, inhibited activities of methanogenic bacteria caused by re-aeration might also contribute to the reduction of methane concentration in water.

Table 3 Water temperature and BOD in different sites in the constructed wetland

Date	Parameter	A	B	C	D	E
June 16	BOD, mg/L	496				113
	Water temp., ℃	24.0	24.0	23.5	23.0	22.5
July 16	BOD, mg/L	471	422	256	184	95
	Water temp., ℃	26.5	26.5	26.0	26.5	25.5
Aug. 15	BOD, mg/L	532	377	191	134	76
	Water temp., ℃	28.5	28.5	27.5	26.5	26.5
Sep. 14	BOD, mg/L	435		215		92
	Water temp., ℃	26.0	26.0	25.5	26.0	25.5
Oct. 16	BOD, mg/L	419	324			89
	Water temp., ℃	22.0	21.5	20.5	20	19.5
Nov. 15	BOD, mg/L	421				134
	Water temp., ℃	18.5	16.5	15.0	14.0	13.0
Dec. 14	BOD, mg/L	442	368	295	236	173
	Water temp., ℃	11.5	6.5	5.5	5.0	5.0

Table 4 Methane concentration in the sewage from different sites ($\mu gCH_4/L$)

Date	Influent	A	B	C	D	E	Effluent
Aug. 15	25.1	237.4	553.2	685.4	721.1	624.4	315.6
Sep. 14	16.8	173.0	646.0	1190.1	1040.8	850.3	365.9
Oct. 16	9.8	138.2	1043.8	1131.3	1262.5	1193.8	495.6
Nov. 15	4.2	85.7	247.2	350.3	286.7	339.4	199.9
Dec. 15	14.4	33.4	141.1	175.5	155.4	176.9	55.9

Seasonal diversity of methane concentration in sewage increased from August to October, it was caused by increasing solubility of methane in water with the drop of temperature. In winter, the decrease of methane concentration in water (November, December) was caused by the low methane production under low temperature.

3.3 Seasonal changes of methane flux from constructed wetland

The methane flux from constructed wetland was easily affected by water temperature, so it is not surprisingly that, in our result, methane production have positive relationship with average air and water temperatures. Methane flux in summer is higher than that in autumn and winter, up to 29.58 $g/(m^2 \cdot d)$ at the hottest month (average temperature 26.0℃), and decrease gradually when water temperature dropped. In December, when the average temperature was at 0.6℃, water temperature was about 5—6℃, methane emission from wetland weakened sharply and even stopped, except site A near the entrance of influent, where the water temperature was 11.5℃, methane flux was still at a high level (Table 2,3; Fig.4).

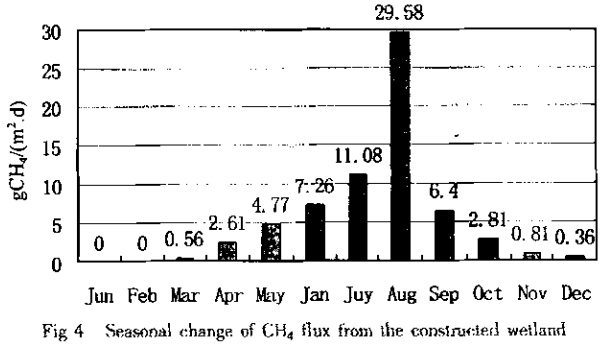

Fig 4 Seasonal change of CH_4 flux from the constructed wetland

3.4 Amount of methane emission from the constructed wetland

The constructed wetland is an significant source of methane, up to 976.6×10^6 gCH_4/a, which was emitted from the constructed wetland with the area of 495000 m^2. Its daily mean methane flux reached 5.22 $gCH_4/(m^2 \cdot d)$, around year, far higher than that from natural wetland located in the same latitude region (20 – 30 $mgCH_4/(m^2 \cdot d)$, even than tropical wetland (148—233 $mgCH_4/(m^2 \cdot d)$; Karen, 1993).

Evidently, the constructed wetland used for municipal sewage treatment is an important source of methane. At present, the sewage loading rate of the constructed wetland was 12000 m^3/d, then emission rate of methane of sewage was 227.8 $mgCH_4/L$ sewage, 700—1000 times as much as methane emission rate observed in the secondary treatment (approximately 300 $mgCH_4/m^3$ sewage; Czepiel, 1993; Mizuochi, 1998).

High methane emission from the constructed wetland was due to large amount of organic pollutant and decayed litter of hydrophilic plants which were not removed in time, supplied more organic carbon for methane generation; the existence of anoxic/anaerobic environment that is conducive to the methanogenic bacteria, discharge of sewage into wetland systems, providing a spacio-temporal environment (8.25 days) for methane production.

Therefore, methane emission can be reduced through improving aeration conditions of wetland; and removing sludge and biomass in time.

3.5 N_2O emission from the constructed wetland

Diurnal change of N_2O flux from the constructed wetland was as irregular as methane emission (Table 5). But its seasonal changes are correlated with air temperature closely, and reverse correspondent to methane emission. During the period with large methane emission (from June to September), N_2O flux was very low (-0.10—0.21 mg $N_2O/(m^2 \cdot d)$), even less than 0 and make the system become a sink. Along with decrease of methane flux due to the low temperature, N_2O flux increase gradually, reached to 2.50—4.65 $mgN_2O/(m^2 \cdot d)$ (Fig.4 and Table 6). Emission of N_2O from the constructed wetland is strongly influenced by DO. In summer, DO in water body was low because of the consumption of strong microbial activity under high temperature. The anoxic condition improve methane production and inhabit N_2O generation. On the contrary, in the winter, DO will be increased under low temperature, which can inhibit methane emission, while increasing N_2O emission.

Table 5 Diurnal change of N_2O emission from the constructed wetland ($gN_2O/(m^2 \cdot d)$)

Time	January *	February *	March	April	May	June	July	August	September	October	November	December
0:00	0	0	182.3	133.4	80.7	3.6	-10.5	-7.7	4.0	63.4	167.6	200.3
4:00	0	0	224.6	98.5	55.8	3.3	-2.6	-2.4	23.9	102.6	206.5	134.3
8:00	0	0	143.4	137.5	79.4	8.1	15.0	-2.1	11.8	105.7	131.9	188.0
12:00	0	0	101.6	116.8	124.3	13.9	-3.6	-4.8	7.8	64.1	93.4	187.4
16:00	0	0	180.2	172.5	116.3	16.6	1.3	-1.4	-5.8	147.5	165.7	254.5
20:00	0	0	265.3	155.2	56.5	-0.4	-6.1	-7.7	11.8	142.6	243.9	199.3

* Greenhouse gas emission stop, because water in the wetland freeze in January and February in Jiaonan County

Table 6 Seasonal change of N_2O flux from the constructed wetland (mg $N_2O/(m^2 \cdot d)$)

Date	Air temperature, ℃	Site					Average
		A	B	C	D	E	
January *	-1.9	-	-	-	-	-	0
February *	0.0	-	-	-	-	-	0
15 - March	5.0	15.97	3.46	1.73	0.48	0.33	4.3
15 - April	11.4	10.59	2.31	2.06	0.92	0.44	3.26
15 - May	17.3	7.85	1.86	1.57	0.78	0.54	2.52
16 - June	21.6	0.10	0.31	0.38	0.04	0.05	0.18
16 - July	25.5	0.40	0.27	-0.24	0.26	-0.29	-0.03
15 - August	26.0	-0.23	-0.43	-0.13	0.16	0.11	-0.10
14 - September	23.2	0.44	0.26	0.82	0.34	-0.54	0.21
15 - October	15.2	8.43	1.18	1.85	0.71	0.34	2.50
15 - November	7.6	16.38	1.44	1.63	0.44	0.30	4.04
14 - December	0.6	17.17	2.85	1.58	0.86	0.82	4.65
Average	17.1	6.10	0.84	0.73	0.33	0.18	1.64

* Greenhouse gas emission stop, because water in the wetland freeze in January and February in Jiaonan County

Similarly to methane production, most of nitrous oxide was produced in the upper-reach section of the plot, a decreasing tendency from upper to down-reach was observed. However, N_2O flux was much lower (1.64 $mgN_2O/(m^2 \cdot d)$) compared with methane flux from the wetland (8.33 $gCH_4/(m^2 \cdot d)$).

The total of N_2O emission from the constructed wetland was 0.304×10^6g per year. On the base of wastewater amount, 0.07 mg of nitrous oxide was produced from each liter sewage applied. Compares with the N_2O emission data observed in the conventional sewage treatment plants (0.33 – 1.50 mgN_2O/L, Mizuochi, 1998; 0.05 mgN_2O/L, Czepiel, 1995), it was not high.

4 Conclusions

The constructed wetland for municipal sewage treatment is a significant source of methane emission. The mean methane flux reached 5.52 $gCH_4/(m^2 \cdot d)$, 250 times as much as that in natural wetland in the same latitude region. Total methane emission is about 976.6×10^6g. To treat one liter of wastewater, 227.8 mg CH_4 will be produced.

Methane emission mainly occurred in summer (from June to Sep.), production during this period account for is 90% of the annual total.

N_2O emission from the constructed wetland is little. The total emission amount to about 0.304×10^6g N_2O. Average N_2O flux is 1.73 $mgN_2O/(m^2 \cdot d)$, the contribution rate per liter of wastewater is 0.07 mg N_2O, our observation on N_2O is not higher than that in normal municipal sewage treatment plant.

N_2O emission mainly occurred in winter and spring, makes up more than 90% of the total emission, and fluctuates inversely with methane emission.

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