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Preliminary studies on nitrous oxide emissions from the ornithogenic soils on Xi-sha atoll, South China Sea

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Abstract: The preliminary measurements of nitrous oxide fluxes from the ornithogenic soils on tropical Xi-sha atoll were made using a closed chamber technique for the first time. N_2O fluxes from the ornithogenic soils ranged from 1.8 to 40.3 μg/(m^2·h) on Dong Island and 3.2 to 20.4 μg/(m^2·h) on Yongxing Island and their flux averaged 11.0 μg/(m^2·h) and 8.3 μg/(m^2·h), respectively. N_2O fluxes from two wetland sites in salt marsh of Dong Island were approximately one order of magnitude lower than those from the ornithogenic soils and averaged 2.1 μg/(m^2·h) and 2.4 μg/(m^2·h). The diurnal variation cycle in the fluxes was obtained at the observation sites; the N_2O flux increased with the increase in soil temperature. The sudden increase in soil moisture greatly stimulated N_2O emission from the ornithogenic soils on Dong Island due to the heavy rainfall. The undisturbed soils showed the lower N_2O fluxes and the average was 4.8 μg/(m^2·h) and the soils via the reclamation showed the higher N_2O fluxes and the average was 16.6 μg/(m^2·h) on Yongxing Island, suggesting that the changes of land use have an important effect on N_2O fluxes from the ornithogenic soils. In addition, the N_2O fluxes at the different sites showed high spatial variations. The fluxes were positively correlated with the concentrations of NO_3^-, PO_4^{3-} and Mn in the soils. The negative correlation between the fluxes and total S concentration in the ornithogenic soils was also found for the first time. Coastal soils or sediments constitute an important source of global atmospheric N_2O and the increases in nitrogen loading from seabird guanos will lead to significant increases in the flux of this atmospherically active gas.

Keywords: South China Sea; ornithogenic soil; nitrous oxide; Xi-sha atoll; emission

Introduction

Nitrous oxide (N_2O) is one of the most important greenhouse gases, and it plays an important role in the depletion of stratospheric ozone (Crutzen, 1977; Zhu, 2004). The atmospheric N_2O concentrations have been increasing at the rate of 0.2%—0.3% per year and the global average N_2O concentration has reached 314 nl/L (Flückiger, 1999; Sun, 2000; Zhu, 2003a; 2003b). Such a concentration increase is generally ascribed to direct emissions of N_2O to the atmosphere through fertilizer production, crop production, fossil fuel combustion, biomass burning, nitric acid production and sewage, as well as to the enhancement of biogenic N_2O emissions by additional fixation of nitrogen by cultivation of leguminous plants and deposition of anthropogenic nitrogen compounds through the atmosphere (Mosier, 1991; 1997; Xing, 1998; Corre, 1999). Although the marine environment is also recognized as a net source of N_2O to the atmosphere (Cohen, 1979; Butler, 1989; Zhu, 2003a; 2003b), current global budgets appear to underestimate the marine source of this radiatively active gas (Nevisson, 1995; Corredor, 1999). The increasing atmospheric N_2O concentration and imbalance in its global budget have triggered interest in quantifying N_2O fluxes from various ecosystems (Chen, 1997; Li, 1997; Xing, 1997; Zheng, 1997; Corre, 1999; Dobbie, 1999; Dong, 2000; Sun, 2002).

Coastal areas have been recognized as major marine contributors to atmospheric N_2O (Bang, 1996; Corredor, 1999). N_2O fluxes from mangrove sediments have been measured in the tropical coastline. Results showed that N_2O yield from these sediments varies greatly in response to both the magnitude and chemical nature of the sediment nitrogen source (Corredor, 1999; Munoz-Hincapie, 2002). Such contributions may be further enhanced by the releases of nitrogenous products of seabird guano and anthropogenic sources in coastal soils or marine sediments that enhance microbial nitrogen metabolism and the rates of N_2O flux. However, the N_2O emissions from island ecosystem have not been exclusively reported. During March 10 to April 11, 2003, we carried out an integrated environmental investigation in the Xi-sha atoll. These tropical islands are the important rookeries for all kinds of the seabirds. The accumulation of a lot of seabird guanos forms the phosphorite and the ornithogenic soils. In this paper, for the first time N_2O fluxes from the ornithogenic soils were observed at Dong Island and Yongxing Island. The relationships between N_2O fluxes and environmental variables were also analyzed and discussed preliminarily in this paper.

1 Materials and method

1.1 Study area

Xi-sha atoll (15°47′—17°08′N, 111°10′—112°55′E) locates in the center of South China Sea and belongs to Hainan Province, China. It is about 300 km southeast of Sanya, Hainan Island. It extends from northeast to southwest with a length of 250 km and a width of 150 km, enclosing over 50000 km^2 sea area. This largely enclosed sea is probably one of the most biologically diverse bodies of water on the planet and has thousands of reefs, atolls, submerged reefs and banks like Dongsha area (provided by Oceanic Office of Hainan Province, 1999). However, this region is very mysterious for people due to rare reports. Many islands are covered with flourishing tropical thicket, which is very suitable for the propagation of all kinds of seabirds. The accumulation of seabird guano forms phosphorite and the ornithogenic soils, which provides the development of the vegetation with abundant nutrients (Gong, 1997). N_2O flux

Foundation item: The Key Project of National Natural Science Foundation of China (No. 40231002); the National Natural Science Foundation of China (No. 40406001) and the Specialized Research Fund for the Doctoral Program of Higher Education (No. 20030358002); * Corresponding author observation sites were set in the areas with vegetation cover on Dong Island and Yongxing Island. Dong Island (16°39′N—16°41′N, 112°43′E—112°45′E) is located in the east of Xi-sha atoll, with the area of 1.55 km^2. Annual average air temperature is 26—27℃, annual precipitation is about 1500 mm, and dry and wet seasons are very evident. Rainfall is concentrated during the period from June to November due to the effects of tropical cyclone, occupying 87% of all precipitation, and southwestern monsoon prevails on this island. The rest months show dry seasons due to the effects of northeastern monsoon. The annual evaporation is about 2400 mm, well above annual precipitation; annual average relative humidity is 78%—84%. Shrub, such as *Scaevola sericea*, *Guettarda speciosa*, *Aporosa villosa*, distributes on the periphery and continuous *Pisonia grandis* grows in the middle part of this island. This island is also the rookery of thousands of the rare *Sula sula*. The N_2O flux observation sites D1 and D2 are located in the area of the shrubs on Dong Island. The ground surface is 20—30 cm black organic layer, under which is about 30 cm brown soil layer amended by seabird guanos until gray coral floor. Two observation sites LT1 and LT2 on Dong Island were set in the salt marsh named Liu Tang, which is only fresh water reservoir with deep sediments and abundant organic matter, and it is 100 m far away from the coastline. The *Sesuvium portulacastrum* and some herbaceous vegetation flourished in the salt marsh. Yongxing Island is the largest one on Xi-sha atoll with 1.85 km^2. Thousands of the trees *Cocos nucifera* grow in the middle part of this island and the shrubs are mainly *Guettarda speciosa* and *Aporosa villosa*. Two observation sites YX1 and YX2 were set on Yongxing Island. The sites YX1 and YX2 were plotted into two subsites with and without natural regetation, respectively.

1.2 N_2O flux measurement

The net N_2O fluxes were determined by a closed chamber technique (Mosier, 1997; Xing, 1998; Sun, 2002). At the measurement sites, open-bottomed acrylic resin chambers (50 cm × 50 cm × 50 cm) were placed on stainless-steel collars installed at the measurement sites for the entire study period. The collars enclosed an area of 0.25 m^2 and were inserted into the ground to a depth of about 10 cm in the soils. The use of flux collars allows the same spot to be measured repetitively, minimizes site disturbance, and ensures that flux chambers are well sealed in the uneven ground surface since the chambers fit into a water-filled notch in the collars. Head-space samples were removed from the chamber every 20 min (including zero time) over an hour period and stored in vacuum vials (17.5 ml) made in the Institute of Japanese Agricultural Environment, which had been vacuumized to -1.013×10^5 Pa in advance. The vials was sealed with a butyl rubber septum and then covered by a plastic cap. High vacuum of -1.0×10^5 Pa inside the vial can maintain for a year at least (Xing, 1998; Sun, 2002). In addition, gas standards for N_2O stored in the vials showed no significant changes in concentration during one month of storage in the laboratory and during the transport from the field site to the laboratory, suggesting that the quality of sample air in the vials did not change during the sampling and transport period as well. Net N_2O fluxes from the shrub soils on Dong Island were measured on March 20, 22 and 24. Net N_2O fluxes from the salt marsh were measured on March 29. The fluxes from the soils on Yongxing Island were measured on April 8 and 9. The N_2O fluxes from all the observation sites were observed at four time intervals of 0:00—1:00, 6:00—7:00, 12:00—13:00 and 18:00—19:00 within one day. A total of four samples were taken during a flux measurement. Air temperature inside the chamber was simultaneously measured.

The collected gas samples were analyzed in the Laboratory of Material Cycling in Pedosphere, Institute of Soil Science, Chinese Academy of Sciences, for the N_2O concentrations. N_2O mixing ratios were determined by HP5890 GC with detector-ECD (Xing, 1998; Sun, 2002; Zhu, 2004). GC-ECD was equipped with back flush system with 10-port valves. A pre-column (2 m) and a main-column (Porapak Q, 100 mesh) were used with an argon-methane (95%:5%) mixture as the carrier gas at a flow rate of 30 ml/min. The detector and column temperatures were 330℃ and 85℃, respectively. The back flush time was 2.8 min. The injecting gas volume was 3.0 ml by using an adjustable pressure syringe injector. Compressed air was used as standard gas with the value of 303 nl/L demarcated by the National Institute of Japanese Agricultural Environment. The variance coefficient for standard samples was within 0.1%—0.4% in ten hours. N_2O fluxes were calculated by linear regression of the concentration changes in the four samples and the average chamber temperature (measured each time a sample was taken).

1.3 Environmental variables

The air temperatures in the chambers were taken by the thermometer inserted into it. Ambient air temperatures, surface ground temperatures and the precipitation were collected at the weather station on Yongxing Island. Determinations of pH in the soils were made on fresh soils mixed with distilled water (soil : water, 1 : 2.5) and the pH measured in the supernatant. The analyses of chemical and physical properties were carried out in the laboratory of metallurgical and geological center of Eastern China.

2 Results and discussion

2.1 N_2O flux and environmental variables

The relationships between N_2O fluxes and environmental variables were illustrated in Fig.1. The average N_2O fluxes ranged from 1.8 $\mu g/(m^2 \cdot h)$ to 40.3 $\mu g/(m^2 \cdot h)$ with the average of 11.0 $\mu g/(m^2 \cdot h)$ at the observation sites D1 and D2 on Dong Island. The diurnal variation cycle in the fluxes was obtained at the observation sites. The N_2O fluxes showed the highest values at midday, corresponding to the local air or soil temperatures. N_2O is by-product of nitrification and denitrification (Ryden, 1981). A soil temperature of more than 5℃ is usually required for a significant denitrification rate (Aulakh, 1983; Watanabe, 1997). Continuous short-term measurements indicated that N_2O fluxes followed the same diurnal variation as soil temperature at 10 cm depth, with N_2O production peaking in late afternoon (Williams, 1999). At the seasonal scale highest emissions were often observed in summer (Brumme, 1995; Martikainen, 1993). Therefore soil temperature is one of the most important factors affecting N_2O emissions from the ornithogenic soils on Dong Island.

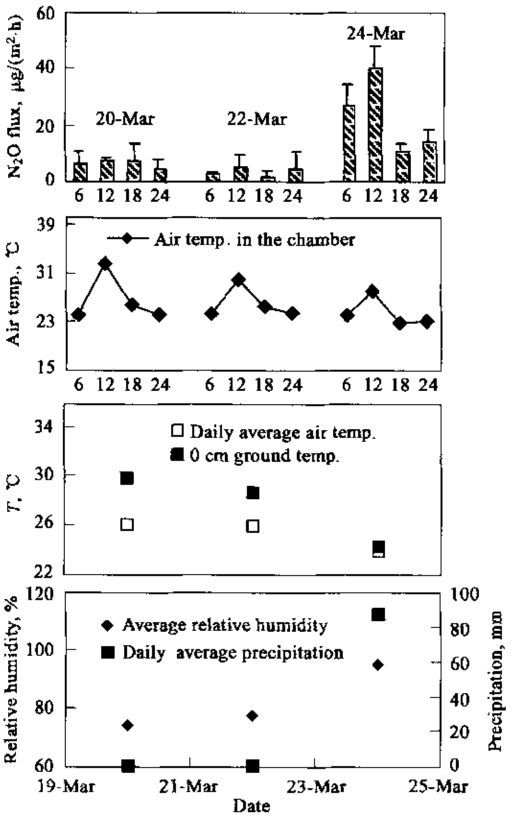

Fig.1 The relationships between the average N_2O fluxes and environmental variables at the observation sites (D1 and D2) on Dong Island. N_2O fluxes at different time were given in the form of the average value ± standard deviation

In our study, maximal N_2O fluxes were up to about 22 times greater than minimal fluxes during the three-day observation period when diurnal fluctuations were large, which is comparable with the 23-fold difference between daily peak and minimum N_2O flux observed by Christensen (Christensen, 1983); The diurnal range in temperature was not greater than 8℃ even when diurnal variations in N_2O fluxes were maximal, suggesting that the diurnal variation in N_2O fluxes was too large to be accounted for by temperature effect alone and, in the absence of significant rainfall, it is likely that variations in solar radiation contributed to the diurnal fluctuations in N_2O flux (Christensen, 1983). Therefore such a great difference may be attributed not only to diurnal variations in soil temperature, but also to high incoming solar irradiation. Our results indicate that there is a need for continuous flux measurements if allowance is to be made for these diurnal variations and, hence, the production of reliable budgets for N_2O emissions from these ornithogenic soils.

In addition, the rainfall appeared to have an important effect on the N_2O fluxes. The sudden increase in soil moisture greatly stimulated N_2O emission due to the heavy rainfall. Rainfall may cause lower oxygen concentrations in the soil pore space and thus the induction of enzymes for nitrate reduction in denitrifiers (Rudaz, 1991). On the other hand, the high temperature and moisture often occur at the same time in our study area. Therefore the hydrothermal interaction may contribute to the highest N_2O emission according to the studies by Kowalenko and Cameron (Kowalenko, 1976).

2.2 Effects of land use changes on N_2O flux

Under the same weather conditions, the N_2O fluxes were observed from the ornithogenic soils with the natural vegetation and without the vegetation via anthropogenic reclamation on Yongxing Island. Results are illustrated in Fig.2. The undisturbed soils with natural vegetation showed lower N_2O fluxes and the average was only 4.8 $\mu g/(m^2 \cdot h)$. However, the soils without vegetation via the reclamation showed the higher N_2O emissions and the average flux was 16.6 $\mu g/(m^2 \cdot h)$, about two and one half times higher than undisturbed soils. Therefore land use changes may have an important effect on the N_2O fluxes from these ornithogenic soils. Other studies also showed that conversion of tropical forest to crop production and pasture has a significant effect on the emissions of N_2O. Keller *et al*. (Keller, 1986) showed that emissions of N_2O increased by about a factor of two when a forest in central Brazil was clear cut, and Luizao *et al*. (Luizao, 1989) reported that pasture soils in the same area produced three times as much N_2O as adjacent forest soils. Our observations were almost in accordance with these literature above.

2.3 Spatial variation of N_2O fluxes

The spatial variations of N_2O fluxes from six sites were analyzed and compared in the Fig.3. The N_2O fluxes from the observation sites D1 and D2 were 12.7 $\mu g/(m^2 \cdot h)$ and 9.3 $\mu g/(m^2 \cdot h)$, respectively, on Dong Island. The fluxes from the sites LT1 and LT2 in salt marsh were 2.1 $\mu g/(m^2 \cdot h)$ and 2.4 $\mu g/(m^2 \cdot h)$, respectively, and the fluxes

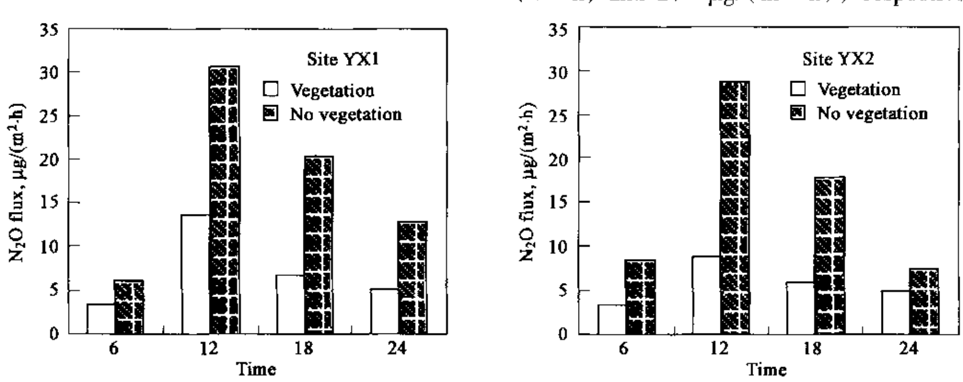

Fig.2 The effects of land use changes on N_2O fluxes in the ornithogenic soils on Yongxing Island

from the sites YXS1 and YXS2 in Jiangjun forest on Yongxing Island were 8.4 μg/(m^2 · h) and 8.2 μg/(m^2 · h), respectively. These observation sites showed a high spatial variation. The wetland of salt marsh generally showed lower N_2O fluxes while dry soils showed higher N_2O emissions. The chemical properties at the different observation sites were also analyzed and compared as listed in Table 1. The chemical properties of soils at the observation sites on Dong Island were very similar to those at Jiangjun forest on Yongxing Island. However, the soils at the salt marsh on Dong Island showed higher salinity, conductivity, TOC, TON, and NH_4^+-N. The positive correlations between N_2O fluxes and the concentrations of NO_3^--N and PO_4^{3-} were obtained in our study according to Fig.4. The content of NO_3^--N is one order magnitude higher than that of NH_4^+-N in the ornithogenic soils, indicating that N_2O may be mainly produced by the denitrification of nitrate. The nutrients in these ornithogenic soils are added usually in the form of nitrate and phosphates. The phosphate content indicated the levels for the nutrients, therefore N_2O emissions from the ornithogenic soils were positively correlated with the nitrate and phosphate concentrations(Fig.4a; Fig.4b).

In addition, the positive correlation between the N_2O flux and Mn concentration in the ornithogenic soils was found for the first time (Fig. 4c). According to the literature (Ivanova, 2004; Slavinskaya, 2004), complex manganese dioxide-based catalysts for oxidation of ammonia by oxygen are the most active and selective with respect to N_2O. Manganese ions with different oxidation degree (Mn^{3+}, Mn^{4+}, and $Mn^{\delta+}$) serve as active sites of the catalyst surface. According to the study of Vione *et al*. (Vione, 2004), the Mn (Ⅲ, Ⅳ) (hydr) oxides show some photocatalytic activity and they can act as thermal oxidants at

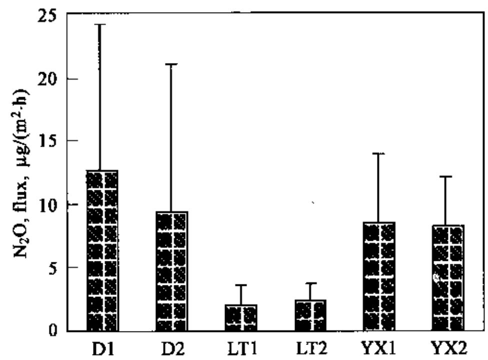

Fig.3 The spatial variations of average N_2O fluxes at different observation sites on Xi-sha atoll. N_2O fluxes from different sites were given in the form of the average value ± standard deviation

acidic pH. The photoinduced oxidation of nitrite and the thermal oxidation of nitrous acid by Mn(Ⅲ, Ⅳ)(hydr)oxides yield nitrogen dioxide and these processes can take place at the water-sediment interface. Therefore the manganese level in the ornithogenic soils had an important on the N_2O emission. However, the N_2O fluxes from the ornithogenic soils and total S concentration showed a negative correlation (Fig. 4d). According to the study by Hasegawa *et al*. (Hasegawa, 2000), in the case of adding elemental sulfur or iron sulfide into the soils, N_2O production was suppressed, indicating the possibility to decrease N_2O emission by adding sulfur into the soil and proceeding sulfur denitrification. Sulfur denitrification is based upon autotrophic denitrification by sulfur oxidizing bacteria such as *Thiobacillus denitrificans* and *Thiomicrospira denitrificans* (Hasegawa, 2000). Under aerobic conditions these bacteria use oxygen as an electron acceptor, but under anoxic conditions these oxidize reduced sulfur(S^{2-}, $S_2O_3^{2-}$, S^0) to sulfate while reducing nitrate

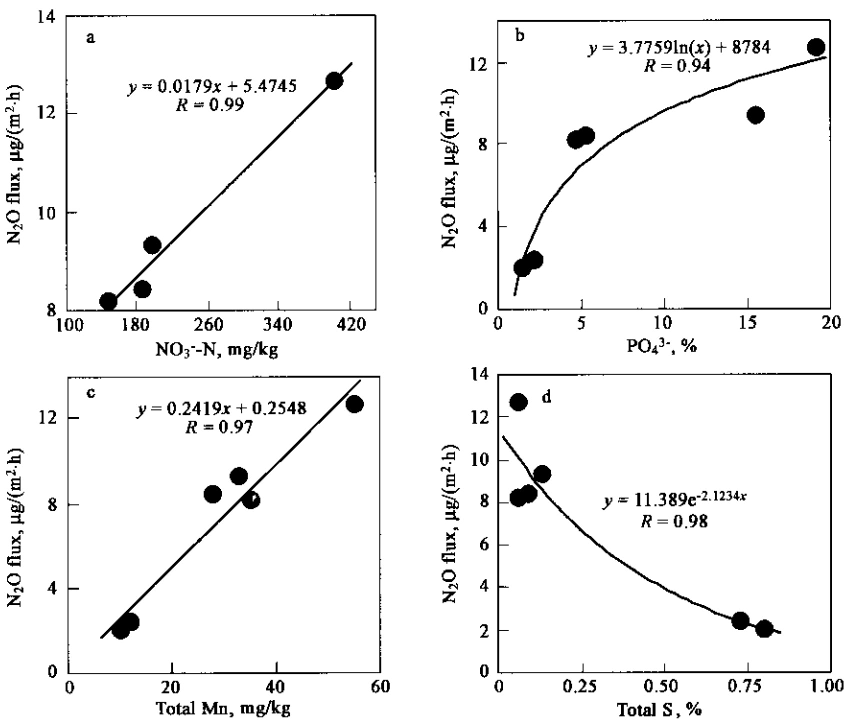

Fig.4 The relationships between N_2O fluxes and chemical properties for the ornithogenic soils at the different observation sites on Xi-sha atoll

finally to nitrogen gas. The equation of sulfur denitrification is shown as follows: $5S + 6NO_3^- + 2H_2O \rightarrow 5SO_4^{2-} + 4H^+ + 3N_2 \uparrow$. Our study indicated that the increase of total S concentration in the ornithogenic soils suppressed the N_2O emission, which is in accordance with the experimental effects in the laboratory.

Table 1 The chemical and physical properties for the soils at different sites on Xi-sha Islands

Sampling sites	The shrubbery on Dong Island		Salt marsh on Dong Island		Jiangjun forest on YX Island	
	Site D1	Site D2	LT1	LT2	YXS1	YXS2
Salinity, 10^{-3}	0.1	0.1	7.0	6.3	0.1	0.1
Conductivity, ms/cm	0.24	0.25	12.3	11.1	0.20	0.18
TDS, mg/L	117.6	124.3	6.14	5.56	99.8	136.0
pH	8.14	8.25	7.65	7.68	8.46	8.47
Redox, mV	186.6	220.1	178.3	152.6	175.0	222.0
TOC, %	6.27	7.30	9.37	9.27	2.95	2.35
Total S, %	0.06	0.13	0.80	0.73	0.09	0.06
Total N, %	1.12	1.30	2.31	2.44	0.54	0.58
NO_3^-, 10^{-6}	402.0	198.0	—	—	185.5	148.5
NH_4^+, 10^{-6}	41.1	81.0	306.0	—	37.1	32.9
PO_4^{3-}, %	19.2	15.6	1.44	—	5.26	4.65
Fe, 10^{-6}	347.5	232.5	295.0	—	552.5	662.5
Mn, 10^{-6}	55.0	32.5	10.0	—	27.5	35.0

2.4 Comparisons with other studies

Many studies of N_2O emissions from temperate and tropical soils have been extensively studied. Recent reviews of the literature on emission rates show that N_2O fluxes from agricultural soils are generally greater and more variable than those from uncultivated land or natural ecosystems. N_2O fluxes from the agricultural soils for the different latitudes are listed in Table 2. Mosier *et al*.(Mosier, 1997) took earlier values from the studies of N_2O emissions from American grasslands (both with and without N-fertilizer application) and compared their flux magnitudes. Their data suggest a trend of decreasing fluxes with increasing latitude. Mean N_2O flux from the ornithogenic soils on Xi-sha atoll, in general, was higher than those from American temperate and tropical grasslands (1.5—6.5 $\mu g/(m^2 \cdot h)$), in spite of the differences of the environmental conditions and emission periods. The N_2O flux from the ornithogenic soils on Xi-sha atoll was one order higher than those from non-ornithogenic

Table 2 The comparisons of N_2O fluxes from the ornithogenic soils on Xi-sha atoll with other global measurements

Source/site	Type of fertilizer	Average flux, $\mu g/(m^2 \cdot h)$	Duration	References
Dong Island	Seabird guanos	12.7	Mar 2003	This study
Dong Island	Seabird guanos	9.3	Mar 2003	This study
Yongxing Island	Seabird guanos	8.4	Mar 2003	This study
Yongxing Island	Seabird guanos	8.2	Mar 2003	This study
Ornithogenic soils, Antarctica	Penguin guanos	89.7 ± 78.5	Jan—Mar 2000	Sun, 2002
Moss soils, Antarctica	No	1.4 ± 0.7	Jan—Mar 2000	Sun, 2002
Moss soils, Antarctica	No	0.9 ± 0.6	Jan—Mar 1999	Sun, 2002
Lichen soils, Antarctica	No	1.2 ± 0.5	Jan—Mar 1999	Sun, 2002
Cotton-winter wheat, Fengqiu	Urea	60.0	1993—1994	Xing, 1997
Paddy field, Guangzhou	Urea	26.0	1994	Xing, 1997
Paddy field, Nanjing	Urea	15.0	1994	Xing, 1997
Paddy field, Suzhou	Urea + pig droppings	107.3	1992	Xing, 1997
Soybean, Northeast China	+ N	42.5	1992	Chen, 1997
Spring wheat, Northeast China	+ N	14.3	1992	Chen, 1997
Flood paddy field, North China	+ Urea	5.22	1995	Li, 1997
Flood paddy field, North China	+ Manure	0.84	1995	Li, 1997
Grassland, Inner Mongolia	No N	10.0	1998	Dong, 2000
Grassland, Alaska	No N	2.3	Summer of 1992	Mosier, 1997
Flat	+ N	6.5		
Hill	No N	1.5	Summer of 1992	Mosier, 1997
	+ N	5.6		
Grassland, Colorado	No N	2.1	1990—1992	Mosier, 1997
	+ N	3.4		
Grassland, Mayaguez	No N	18.3	1992	Mosier, 1997
	+ N	35.4		
Grassland, Lajas	No N	15.8	1992	Mosier, 1997
	+ N	59.4		
Grassland, SE Scotland	+ N	427.5 ± 68.5	March—July 1996	Dobbie, 1999
Grassland, SW Scotland	+ N	650.0 ± 136.9	July 1997—July 1998	Dobbie, 1999

Antarctic soils. However, it is generally lower than those from some fertilized upland and paddy fields of China (15.0—107.3 $\mu g/(m^2 \cdot h)$) reported by some Chinese researchers (Xing, 1997) and those from the ornithogenic Antarctic soils (9.3—225.6 $\mu g/(m^2 \cdot h)$) reported by Sun *et al*. (Sun, 2002). Especially the N_2O flux from the ornithogenic soils on Xi-sha atoll is greatly lower than those from some intensive agricultural soils. For example, N_2O fluxes (136.9—804.4 $\mu g/(m^2 \cdot h)$) from the arable cropland and fertilized grassland of southern Scotland reported by Dobbie *et al*. (Dobbie, 1999).

In our study area dry and wet seasons are very evident. Rainfall is mainly concentrated during the period from June to November due to the effects of tropical cyclone, occupying 87% of all precipitation. Other months show dry seasons due to the effects of northeastern monsoon. In this study, N_2O fluxes from the ornithogenic soils were only conducted during the dry seasons on Xi-sha Islands due to the limited time and field inconvenience. Therefore this study is only given a preliminary study and continuous and long N_2O flux observations are still needed.

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