SCIENCE OF THE TOTAL ENVIRONMENT 407 (2009) 3330–3337

available at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/scitotenv

The spatial distribution and emission of nitrous oxide (N_2O) in a large eutrophic lake in eastern China: Anthropogenic effects

Shilu Wang[a,*], Congqiang Liu[a], Kevin M. Yeager[b], Guojiang Wan[a], Jun Li[a], Faxiang Tao[a], Yingchun Lǚ[a], Fang Liu[a], Chengxin Fan[c]

[a]The State Key Laboratory of Environmental Geochemistry, the Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550002, P.R. China
[b]Department of Marine Science, University of Southern Mississippi, 1020 Balch Blvd., Stennis Space Center, MS, 39529, USA
[c]The Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing, 210008, P.R. China

ARTICLE DATA

Article history:
Received 18 August 2007
Received in revised form
13 October 2008
Accepted 13 October 2008
Available online 20 February 2009

Keywords:
Nitrous oxide
Spatial variation
Anthropogenic pollution
Eutrophied lake
Eastern China

ABSTRACT

The emission of N_2O from China is globally significant, but relatively few direct observations have been made in many of the fresh water environments most likely to be important sites of N_2O production. In this paper, N_2O saturations were examined in the ecologically heterogeneous, eutrophied, Lake Taihu, as well as in surrounding rivers in eastern China. The emissions of N_2O were estimated and compared with those from other landscapes within the Lake Taihu drainage basin. We found that anthropogenically-enhanced inorganic N inputs act as a limited primary control on the spatial distribution of N_2O saturations in heavily eutrophied parts of the lake only and that overall, lake N_2O production and emission are not raised as significantly as expected due to high N inputs. In comparison, the heavily eutrophied river network is an important fraction of the local N_2O budget, and when considered together with emissions of N_2O from the lake, constitute a major (10–50% depending on season) fraction of total N_2O emissions from the Lake Taihu drainage basin.

1. Introduction

Based on long-term records from ice cores, it is known that global concentrations of atmospheric N_2O began to rapidly increase during the last century, and that this trend continues, with anthropogenic sources estimated to contribute approximately 1/3 of the total (Khalil and Rasmussen, 1992; Seitzinger et al., 2000). While it is postulated that a global increase in the frequency of occurrence and extents of hypoxic zones in continental shelf and estuarine settings may seriously impact the global N_2O budget (de Wilde and de Bie, 2000; Naqvi et al., 2000), the potential effects of anthropogenic nutrient inputs on N_2O production in lakes and rivers is poorly understood.

Aquatic N_2O production is complex and sensitive to a variety of processes and variables, as evidenced by the following examples. Increased inorganic nitrogen (NH_4^++NO_3^-) concentrations can promote nitrification in oxic environments and denitrification in anoxic environments to enhance N_2O production (Seitzinger, 1985; McMahon and Dennehy, 1999; Cole and Caraco, 2001). Proliferation of various genera of green algae (*Chlorella*, *Scenedesmus*, *Coelastrum*, and *Chlorococcum*) in response to eutrophication, can significantly enhance N_2O production (Weathers, 1984). By increasing the supply of organic matter, eutrophication can drive hypoxia or anoxia in bottom waters, and stimulate denitrification (Naqvi et al., 2000; Liikanen and Martikainen, 2003). Enhanced denitrification and the addition of anthropogenic NO_3^- lead to significant accumulations of N_2O (Herbert, 1999; Cole and Caraco, 2001).

The importance of lacustrine systems in emitting or absorbing N_2O is poorly constrained. Generally, lakes are not

* *Corresponding author*. The Institute of Geochemistry, Chinese Academy of Sciences, 73 Guanshuilu Road, Guiyang, Guizhou, 550002, P.R. China. Tel.: +86 851 5891507; fax: +86 851 5891609.

0048-9697/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2008.10.037

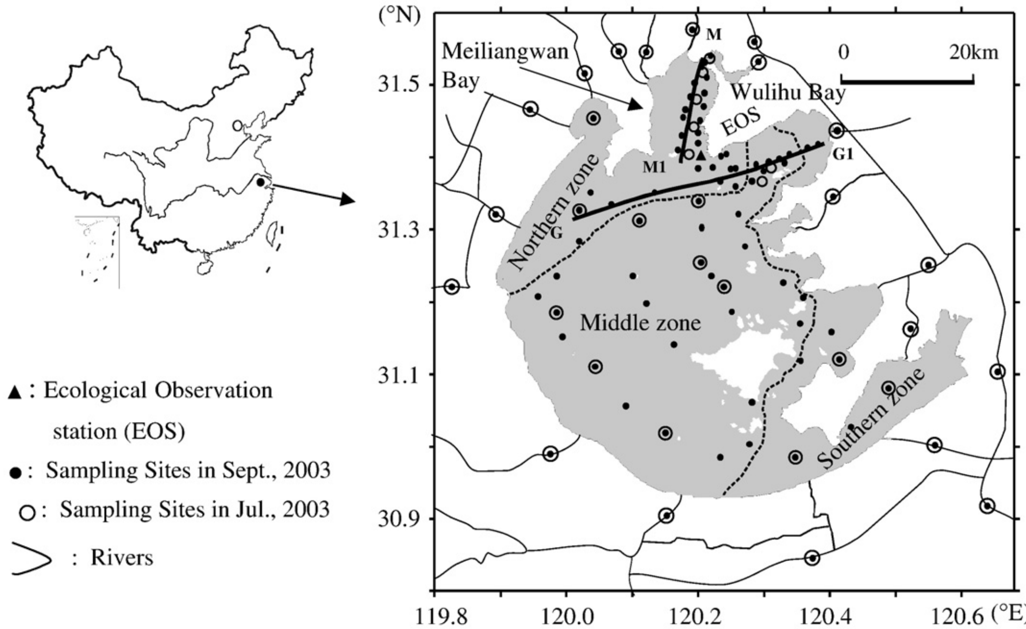

Fig. 1 – Map of Lake Taihu, China. The dashed lines delineate the boundaries of the three ecological zones, and the open and solid circles delineate the locations of sample sites in July and September, 2003, respectively. The sampling transects M–M_1 and G–G_1 are also shown.

considered significant sources of N_2O to the atmosphere (Mengis et al., 1997; Seitzinger and Kroeze, 1998), and in some cases have been shown to serve as minor or seasonal sinks (Hendzel et al., 2005). However, higher average N_2O saturations have been observed in surface waters of eutrophic as compared to oligo- and meso-trophic lakes (Mengis et al., 1997; Huttunen et al., 2003). Lakes are important sinks for watershed-derived N (e.g., Gulati and van Donk, 2002; Wang et al., 2006; Bunting et al., 2007), the extent to which lacustrine N_2O saturation is enhanced by increasing N levels and expanding eutrophication is not well characterized.

In the aquatic environment, nitrous oxide is mainly produced by two biogenic mechanisms. The first is reductive, microbial production during denitrification (Knowles et al., 1981), and the second is oxidative, microbial production during nitrification (Yoh et al., 1988). As an intermediate in denitrification (NO_3^- to N_2), N_2O may accumulate when O_2 is present, NO_3^- concentrations are high and pH is low (Knowles, 1996). The rate of denitrification is influenced by temperature, abundance of organic carbon and supply of N (Knowles, 1996); all save N supply are interrelated with O_2 concentrations. As such, it is not surprising that incubation experiments show that N_2O dynamics in eutrophic lakes are regulated by the availability of O_2 (Liikanen and Martikainen, 2003). In nitrification (NH_4^+ to NO_2^-), N_2O can be produced as a by-product when NH_4^+ is oxidized by nitrifying bacteria and methanotrophs (Mengis et al., 1997). Methane oxidation can interfere with NH_4^+ oxidation due to competition for O_2 (Liikanen and Martikainen, 2003). Methane and N_2O concentrations have been compared to explore the processes governing N_2O production in fresh waters (Mengis et al., 1997).

Calculated N_2O emissions from China are globally significant, accounting for over 90% of those in the Pacific Basin (Seitzinger and Kroeze, 1998). Despite this, few direct observations of N_2O production and emission have been made in the many impacted watersheds, lakes and estuaries of China (Xing et al., 2001; Wang et al., 2006). This shortage of data in key areas is a major contributor to the uncertainty of modeling results (Seitzinger and Kroeze 1998; Seitzinger et al., 2000). In this study, we investigated the spatial patterns of N_2O saturation in a large, eutrophied lake and its surrounding rivers in eastern China. The objectives of this study were to address the following hypotheses: (H_1) variable nutrient inputs and degrees of eutrophication act as the primary control on the distribution of N_2O saturations and emissions throughout the lake, and (H_2) when considered together, lake and river N_2O emissions constitute a considerable fraction of the total N_2O emissions from the Lake Taihu drainage basin.

2. Study site

Lake Taihu is the third largest lake in China with a surface area of approximately 2338 km^2, an average water depth of 1.8 m and a volume of approximately 5.77×10^9 m^3. It is located in a heavily polluted region in eastern China (Fig. 1). A population of more than 37 million and a significant industrial complex are located within its drainage basin (~36,500 km^2). Cultivated lands comprise approximately 15,100 km^2 of the drainage area and are heavily fertilized annually (~34.5 g N m^{-2} a^{-1}) (Xing et al., 2001; Qin and Luo, 2004). The lake annually receives approximately 30,635,000 kg total nitrogen (TN), 1,751,000 kg total phosphorus (TP) and 131,223,000 kg chemical oxygen demand on chromium (COD_{Cr}) from a combination of municipal and industrial wastewaters and agricultural soil runoff (Qin and Luo, 2004). Consequently, the lake is eutrophied, particularly in the northern area. Lowlands comprise more

Table 1 – Summary total nitrogen, total phosphorus, dominant vegetation types, N_2O saturation, CH_4 saturation and the estimated N_2O exchange fluxes in Lake Taihu and adjacent rivers.

Area	Vegetation	TN [a] (mg L^{-1})	TP [a] (mg L^{-1})	CH_4 saturation [b] (%)		N_2O saturation [b] (%)		N_2O fluxes [b] (μmol m^{-2} d^{-1})	
				Jul.	Sept.	Jul.	Sept.	Jul.	Sept.
North L. Taihu	Algae	3.52	0.13	8746±9336 (98.0–757.5)	2616±2552 (39.9–332.7)	689±472 (21.9–98.8)	331±189 (11.5–46.3)	54.9±63.5	24.8±22.0
Middle L. Taihu	Algae/macrophyte	2.0	0.07	783±781 (1.5–64.7)	1330±1318 (3.5–115.3)	184±44 (6.6–16.8)	176±38 (9.4–21.1)	8.3±4.4	7.3±3.1
South L. Taihu	Macrophyte	0.77	0.03	3554±1731 (44.5–127.3)	1579±1350 (12.7–165.0)	80±10 (4.6–7.9)	144±12 (9.8–14.0)	−1.7±0.9	5.0±1.4
Lake averages [c]		2.08	0.07	2556	1581	246	195	14.0	9.7
Rivers		4.27	0.16	20,157±15,054 (17.1–1588)	3148±3048 (25.7–286.1)	1501±744 (10.5–169.2)	342±223 (9.8–62.2)	142.1±72.9	28.8±26.4

[a] Data are from Zhai and Zhang (2006).
[b] Given are means±SE, concentration range (nmol L^{-1}) is shown in parentheses.
[c] Averages are weighted to surface areas.

than 80% of the drainage basin. A dense river network, with a total distributary length of approximately 120,000 km, covers 7% of the total drainage area. Water pollution is more severe in some of these rivers as compared to the lake, particularly those which flow into the lake, as evidenced by higher concentrations of TN and TP (Table 1) (Qin and Luo, 2004; Zhai and Zhang, 2006).

Spatial contrasts in ecology, vegetation and nutrient levels are evident, allowing Lake Taihu to be divided into three zones (Fig. 1): the hypereutrophic–eutrophic northern zone where pollution (TP, TN and COD) is most serious (Qin and Luo, 2004; Table 1), vegetation is dominated by large blue-green algae blooms (cyanobacteria) and sediments consist mainly of organic and nutrient-rich sludge; the middle zone where TN and TP concentrations and the degree of eutrophication are intermediate between the northern and southern zones (Table 1), vegetation consists of a mix of algae and macrophytes and sediments are sandy, with relatively little organic matter; and the mesotrophic southern zone where pollution is relatively light, concentrations of TN and TP are the lowest (Table 1), vegetation is dominated by submersed and emergent macrophytes and sediments are a thick, poorly consolidated and organic-rich slurry. These spatial heterogeneities provide an excellent setting to compare the importance of various factors on aquatic N_2O production and emissions.

3. Materials and methods

Wind-driven mixing provides for a well mixed water column in this very shallow lake (McCarthy et al., 2007), which is confirmed by a high observed vertical velocity range (1.5 to 2 cm s^{-1}) (Luo et al., 2004). Thus, concentrations of N_2O in surface waters are nearly equivalent to those in bottom waters (Wang et al., 2008). As a result, only intermediate-depth lake waters (0.7–0.9 m depth) were sampled, and their N_2O concentrations are considered representative of conditions at each site. Lake waters were sampled at 18 sites, encompassing all three lake zones, in July, 2003. Additional sampling sites were added in September, 2003 with the intention of profiling N_2O concentrations along environmental gradients from both east to west and north to south (Fig. 1, transects G–G_1 and M–M_1, respectively). Additionally, 19 major rivers flowing into the lake were sampled in both July and September, 2003 (Fig. 1).

Field measurements included total dissolved solids (TDS) and pH, which were determined using a PIONneer 65 multi-parameter instrument (Radiometer Co.); and dissolved oxygen (DO), which was determined using a portable DO analyzer (HANNA Co.). Small water samples were collected at each station using serum bottles for the laboratory determination of NO_3^- and NO_2^- using an HPLC1100 liquid chromatograph; and for NH_4^+, which was determined by a spectrophotometer (Unico 2000). Water samples for gas measurements were collected in serum bottles, followed by the addition of 10 M NaOH to raise the pH (10^+), after which the bottles were sealed with no air bubbles. At the laboratory, the headspace equilibrium technique was used to determine concentrations of dissolved gases (Mengis et al., 1997; Wang et al., 2008). Approximately 20 ml of ultra-pure N_2 was injected into the sample bottle and water was displaced. After equilibration at 25 °C by vigorously shaking for 30 min in a water bath, the N_2O concentration in the headspace was analyzed using an ECD-GC (HP6890) equipped with a 4.5 m×3 mm packed Porapak Q (80/100 mesh) column. The column and ECD detector were conditioned at 50 °C and 320 °C, respectively. A mixture of Ar/CH_4 (95/5 v/v) was used as a carrier gas at a flow rate of 20 ml min^{-1}. Headspace CH_4 concentrations were determined using an FID-GC (HP6890) equipped with a 2 m×3 mm carbon molecular sieve column (60/80 mesh). The column and FID detector were conditioned at 90 °C and 250 °C, respectively. Ultra-pure hydrogen gas was used as a carrier gas at a flow rate of 20 ml min^{-1}.

From headspace concentrations, the formula described by Butler and Elkins (1991) was used to calculate gas concentrations, with a mean error of ±4%. Gas concentrations are expressed by the saturation degree relative to air (Mengis et al., 1997):

$$N_2O\ \text{saturation} = (C_{N_2O}/C_{N_2O\text{atm.}}) \times 100$$

where C_{N2O} is the measured concentration of N_2O, and $C_{N2Oatm.}$ is the saturation concentration of atmospheric N_2O at the given

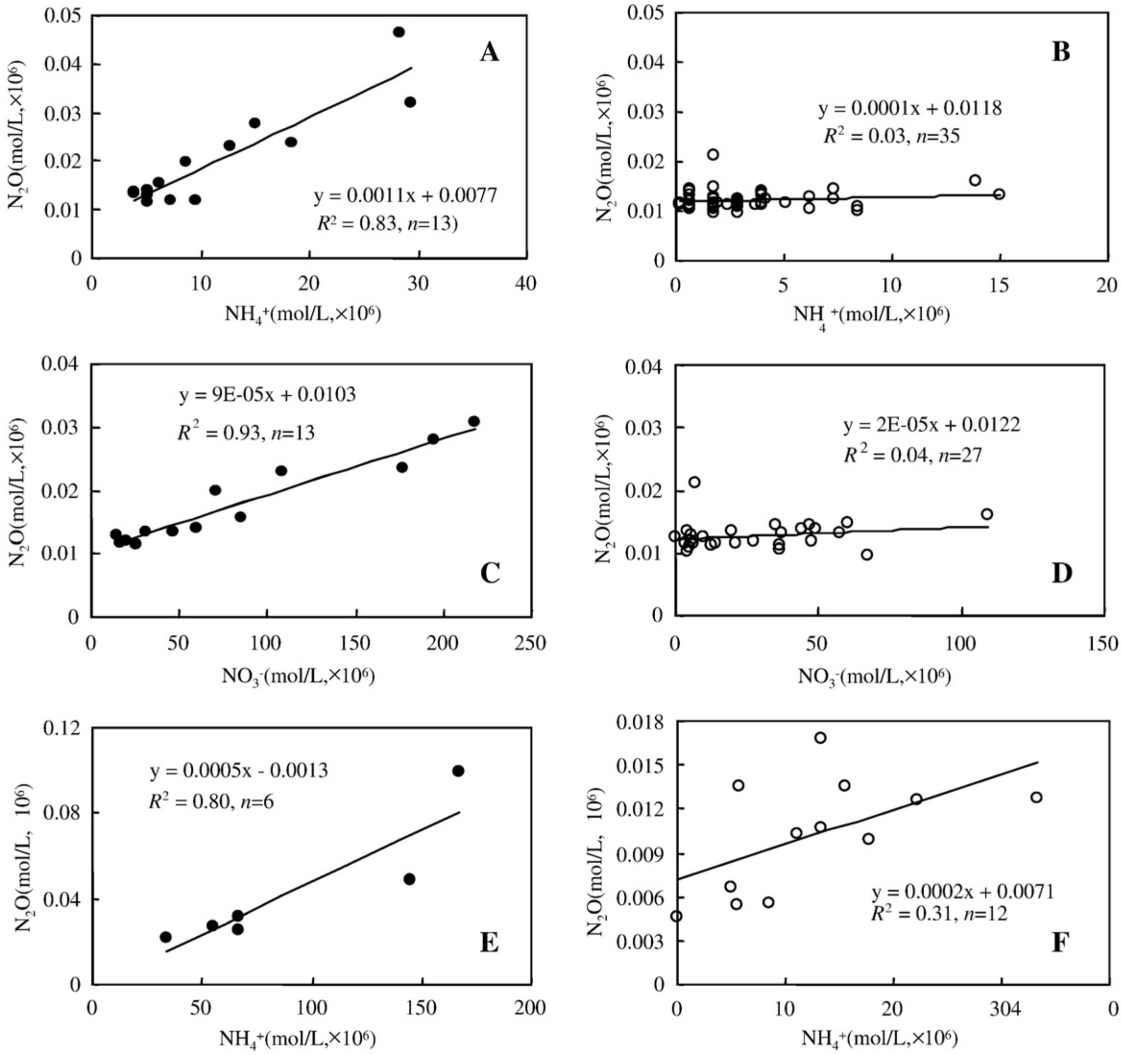

Fig. 2 – N_2O versus NH_4^+ (A, B) and NO_3^- (C, D) in Lake Taihu (September, 2003), and N_2O versus NH_4^+ (E, F) (July, 2003). The solid circles denote samples from the eutrophied northern zone and the open circles from the less eutrophied middle and southern zones.

water temperature. Expression of the CH_4 saturation degree is analogous to that of N_2O. The exchange flux of N_2O at the water–air interface is calculated using:

$$F = K\Delta C$$

where F is the gas exchange flux, ΔC is the difference between N_2O concentrations in the air and water, and K is the gas transfer velocity (calculated from the wind speed and Schmidt number (Wanninkhof, 1992; de Wilde and de Bie, 2000)).

4. Results and discussion

4.1. N_2O production

The rate of increase of N_2O relative to NH_4^+ that can be estimated as N_2O saturations was found to correlate linearly with NH_4^+ concentrations. Regressing the September data for the entire lake yielded a slope of 0.08×10^{-2}; in comparison, the slope was 0.11×10^{-2} using only those data collected in the eutrophied northern zone (Fig. 2A). These relationships suggest that approximately 1 mol of N_2O is formed during nitrification for every 900–1250 mol of NH_4^+ in a given volume during September in Lake Taihu. As a proportion of the NH_4^+ concentration, these data are within the range of N_2O yields reported from the Scheldt Estuary by de Wilde and de Bie (2000) (0.04% to 0.42% (mol N_2O per mol NH_4^+)), and are slightly lower than the average N_2O yield of 0.14% (1 mol N_2O for 700 mol NH_4^+) during nitrification in both fresh and salt water areas (de Wilde and de Bie, 2000). The same regression using the July data, for the entire lake resulted in a slope of 0.05×10^{-2} (Fig. 2E), which is presumably a result of the origin of N_2O from denitrification. Despite strong positive relationships between N_2O and both NH_4^+ and NO_3^- in the lakes eutrophied northern zone (Fig. 2A, C, E), large increases in NH_4^+ and NO_3^- resulted in only modest increases in N_2O, particularly for the September data set.

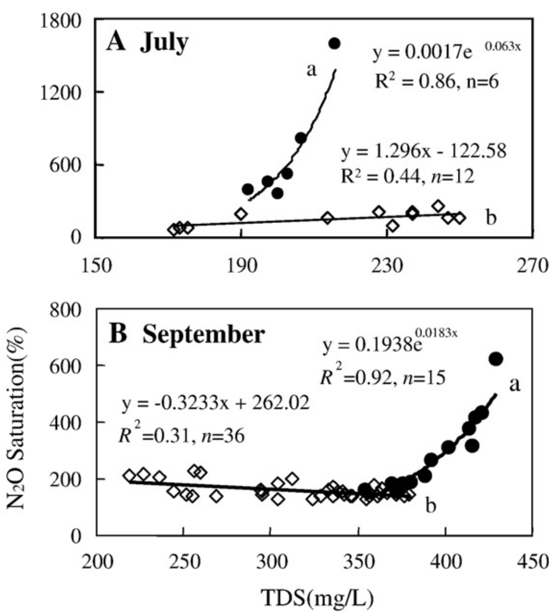

Fig. 3 – N_2O saturation relative to TDS concentration in the heavily eutrophied northern zone (solid circles) and less eutrophied middle and southern zones (open diamond) during July (A) and September (B), 2003.

4.2. *N_2O saturations and inorganic N*

Strong, positive correlations of N_2O saturations with NO_3^- ($r=0.85$, $p<0.01$) and NH_4^+ concentrations ($r=0.74$, $p<0.01$) were observed in the September data set (with the sole exception at the site in Wulihu Bay). In the July data set, N_2O saturations did not significantly correlate with NO_3^-, but did with NH_4^+ concentrations ($r=0.94$, $p<0.01$). Overall, N_2O saturations and inorganic nitrogen concentrations were most strongly associated in the heavily eutrophied northern zone of the lake (Fig. 2A, C and E), not in the less eutrophied middle and southern zones (Fig. 2B, D and F). Additionally, both the September and July data sets show that N_2O saturations are strongly related to TDS concentrations over a wide range (~190–220 mg l^{-1} in July; ~350–440 mg l^{-1} in September) in only the northern zone of the lake (Fig. 3). The exponential increase of N_2O saturations relative to TDS concentrations is consistent with enhanced N_2O production by increased N loading in the eutrophied northern zone of the lake.

4.3. *Distribution of N_2O and CH_4 saturations*

Significant spatial differences in N_2O saturation were observed in Lake Taihu's three zones (Fig. 4A, Table 1), ranging from 161% to 1579% in the northern zone, 140% to 267% in the middle zone and 70% to 164% in the southern zone. Maximum saturations of N_2O, 1597% (July) and 748% (September) were observed in Wulihu Bay (Figs. 4A and 5A) where nutrient pollution is most serious. In both the northern and middle zones, N_2O was supersaturated with respect to the atmosphere during both July and September, 2003. In the macrophyte-dominated southern zone, lake water was not saturated with respect to atmospheric N_2O during the growing season (July) and thus acts seasonally as a N_2O sink (Table 1). From north to south, N_2O saturations decrease along the M–M_1 transect (Wulihu to Meiliangwan Bays) (Fig. 5A) and are spatially consistent with concentrations of TN and TP, and the degrees of lake eutrophication during July and September (Table 1).

As previously noted, significant ecological contrasts were observed between the three lake zones, these are considered here as possible factors influencing the spatial distribution of N_2O. Overall, algal blooms dominate in the lake's northern zone, corresponding to high N_2O concentrations, while the abundant macrophytes of the southern zone correspond to low N_2O concentrations (Table 1). Generally, floating algae mats are not major sites for denitrification (Schaller et al., 2004), however, high N_2O concentrations and fluxes during algal blooms can result indirectly from the supply of dissolved and particulate organic carbon provided by the algae, which can stimulate denitrification (Harrison and Matson, 2003). In

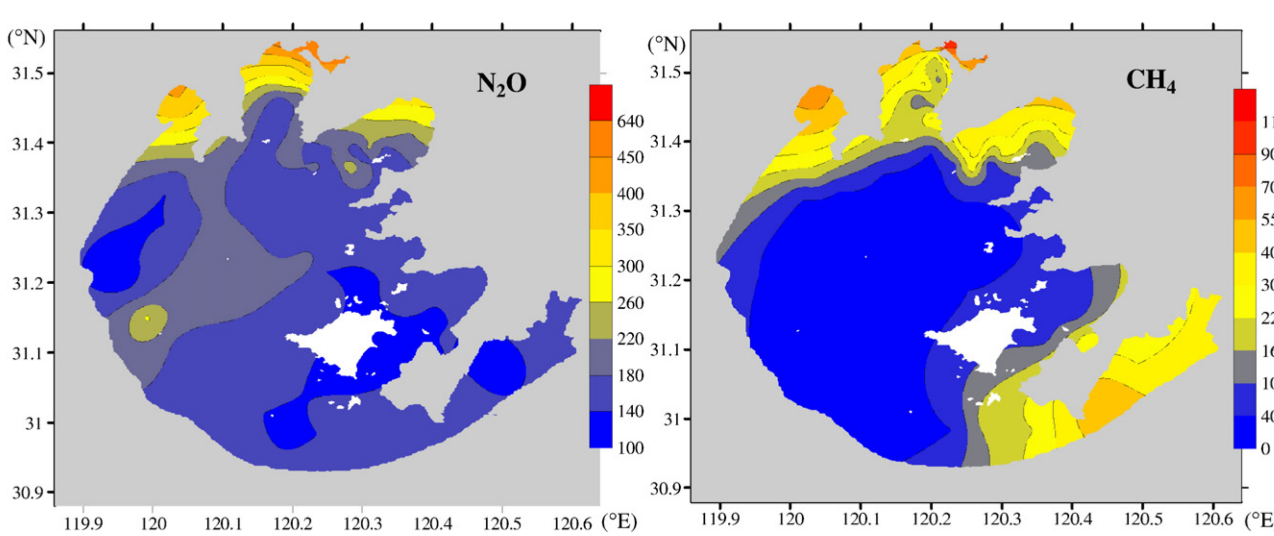

Fig. 4 – Variations in saturation degree (%) of N_2O (A) and CH_4 (B) (September 2003).

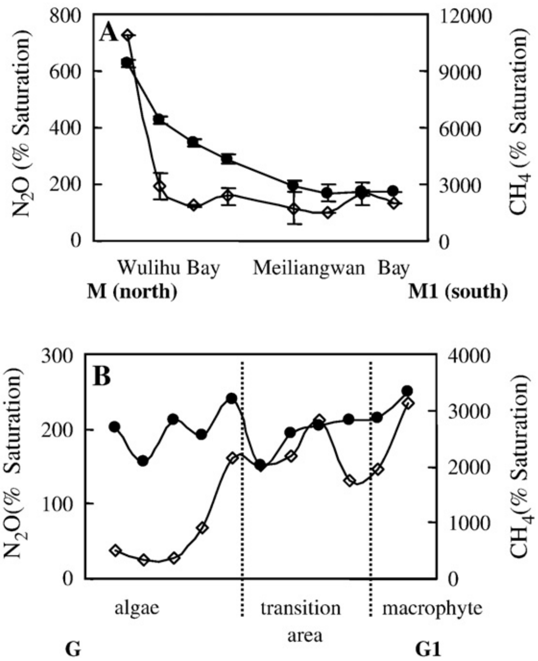

Fig. 5 – Variations in N_2O (solid circles) and CH_4 (open diamonds) saturations along transects M–M_1 (A) and G–G_1 (B).

the lake's northern zone, N_2O saturation does not show any obvious response to the gradient from algae to macrophyte dominance along the G–G_1 transect (Fig. 5B). In rivers, Chenier et al. (2006) showed a significant relationship between the denitrification rate and the biomass of algae and heterotrophic bacteria, but not with cyanobacteria. Cyanobacteria-dominated algal blooms in the lake's northern zone are likely not an important driver of high N_2O concentrations. The lack of N_2O saturation during the growing period (July) in the macrophyte-dominated southern zone of the lake (Table 1) is mainly attributed to consumption by denitrifiers, which can reduce N_2O to N_2 while NO_3^- concentrations remain low (Herbert, 1999; Harris, 1999). Denitrification can be stimulated by macrophytes, which can trap labile organic detritus in the water column or release it from roots (Herbert, 1999; Li et al., 2008). The observed unsaturated conditions lead to the hypothesis that macrophyte-dominated lakes with low nitrate concentrations may act seasonally as a sink for atmospheric N_2O. This requires further research and may play an important role in accurately estimating how aquatic ecosystems influence regional and global N_2O loadings. At Lake Taihu, vegetative variations alone are likely not a primary factor affecting the spatial distribution of N_2O. The indirect effects of vegetation variability on N_2O production, including regulating concentrations of various N species, the supply and nature of organic carbon, and the condition of sediment–water interface, are likely more significant.

Saturations of N_2O in the rivers around Lake Taihu were generally higher than those in the lake, coinciding with higher concentrations of TN and TP (Table 1). The maximum observed N_2O saturation (2708%) exceeded the maximum value (2500%) documented in the N-enriched South Platte River, U.S.A. (McMahon and Dennehy, 1999). Saturations of N_2O in the heavily eutrophied rivers in the northern and eastern areas ranged from 1655% to 2708%, and were generally much higher than those in the less eutrophied rivers of the western and southern areas (631% to 1077%).

Saturations of CH_4 were similar to those of N_2O in the heavily eutrophied northern zone of the lake during July ($r=0.97$, $p<0.01$) and September, 2003 ($r=0.75$, $p<0.01$) (Fig. 4A, B). As with N_2O, maximum CH_4 saturations were found in the heavily eutrophied Wulihu Bay; and also decreased from north to south along the M–M_1 transect (Wulihu to Meiliangwan Bays) (Fig. 5A). In the less eutrophied, middle zone of the lake, similarities in spatial distributions of CH_4 and N_2O were weak in September ($r=0.48$, $p=0.02$), and absent in July, 2003, and were absent entirely in the macrophyte-dominated southern zone of the lake. The overall north to south decreasing trend for N_2O saturations, as well as for the similarities in spatial distributions of N_2O and CH_4, likely reflects the weakening influences of human activities as evidenced by TN, TP and COD concentrations, in particular, the increasing distance from sewage outlets.

4.4. *Emission flux of N_2O*

Exchange fluxes of N_2O were estimated in the three zones of the lake in July and September (Table 1). The heavily eutrophied northern zone accounts for 40–60% of all N_2O emissions from the lake, despite occupying only 16% of the total surface area (Table 1). Surface area-weighted, average N_2O emission fluxes from Lake Taihu are 14.0 and 9.7 μmol m^{-2} d^{-1} in July, and September, 2003, respectively (Table 1), which equates to an annual emission flux of approximately 1.01×10^7 mol N_2O from Lake Taihu (at 11.8 μmol m^{-2} d^{-1}). The estimated emission rate (11.8 μmol m^{-2} d^{-1}) is slightly higher than the median value (10.4 μmol m^{-2} d^{-1}) of N_2O emission measured during 2003–2004 (Wang et al., 2006).

Approximately 4.46×10^7 kg TN was discharged into the lake in 2002, of which approximately 1.15×10^7 kg TN flowed out of the lake and 3.31×10^7 kg TN was lost in the lake (Zhai and Zhang, 2006), most of which was emitted into the atmosphere as N_2 and N_2O. Assuming the same lake N budget in 2003, ~0.63% of the total N discharged into the lake was released to the atmosphere as N_2O. While there are no available N_2O emission data from sewage-fed lakes for direct comparison, the ~0.63% value is less than the suggested default emission factor (N_2O:TN) from rivers (EF5-r, 0.75%, Houghton et al., 1996 (IPCC)). Estimated N_2O emission rates for the eutrophied rivers in the Lake Taihu drainage basin are 142.1 and 28.8 μmol m^{-2} d^{-1} in July and September, respectively, (Table 1). These rates are much higher than those from either the lake or the fertilized lands in the drainage basin (16.5 to 90.7 μmol m^{-2} d^{-1}) (Zheng et al., 1997).

If the mean emissions in Lake Taihu are extrapolated to the total surface area (~3160 km^2) of all lake water bodies in the Lake Taihu drainage basin (~36,500 km^2), approximately 0.49–0.73×10^6 kg N_2O are being released annually. In comparison, estimated, annual N_2O emissions from cultivated lands in the drainage basin are ~12.22×10^6 kg N_2O. No data are available on the N_2O emission flux from the forested lands in this area. It is clear from these data that eutrophic lakes are small contributors to regional N_2O emissions, despite enhancements resulting from increased nutrient loads. Applying the river N_2O emission rate to the entire drainage basin river network, gives approximately 1.19–5.89×10^6 kg N_2O emitted annually, which is 10–50% of the N_2O emissions estimated from all cultivated lands within the drainage basin. This estimate is in congruence with other findings which indicate that river systems with high nutrient loads are important sources of anthropogenic N_2O to the atmosphere (McMahon and Dennehy, 1999; Seitzinger et al., 2000; Garnier et al., 2006).

5. Conclusion

High levels of anthropogenic N inputs have a limited, positive effect on N_2O production and emission only in the eutrophied northern area of the lake and overall, N_2O production and emission are not raised as significantly as expected (H_1). In comparison, the heavily eutrophied river network is an important fraction of the local N_2O budget, and when considered together with emissions of N_2O from the lake, constitute a major (10–50% depending on season) fraction of total N_2O emissions from the Lake Taihu drainage basin (H_2). Despite the indication that plant type has little impact on N_2O production within the northern zone of the lake ($G–G_1$ transect, Fig. 5B), future research should focus on longer and higher resolution time series field measurements as well as mesocosm experiments which focus specifically on the role of various flora (algae, macrophytes) as direct or indirect modifiers of the production rate of N_2O in lacustrine systems.

Acknowledgements

We thank our colleagues Xiao H., Zhu Z., Zhou Z. and An N., as well as our friends from the Ecological Observation Station in Taihu Lake, the Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences for their assistance with sampling and analytical work. We also thank the anonymous reviewer for the helpful comments. This research was supported jointly by the CAS International Partnership Project, the National Natural Sciences Foundation of China (40873013, 90610037 and 40073032) and the National Key Project for Basic Research (2006CB403200).

REFERENCES

Bunting L, Leavitt PR, Gibson CE, McGee EJ, Hall VA. Degradation of water quality in Lough Neagh, Northern Ireland, by diffuse nitrogen flux from a phosphorus-rich catchment. Limnol Oceanogr 2007;52:354–69.

Butler JH, Elkins JW. An automated technique for the measurement of dissolved N_2O in natural Waters. Mar Chem 1991;34:47–61.

Chenier MR, Beaumier D, Fortin N, Roy R, Driscoll BT, Lawrence JR. Influence of nutrient inputs, hexadecane, and temporal variations on denitrification and community composition of river biofilms. Appl Environ Microbiol 2006;72:575–84.

Cole JJ, Caraco NF. Emissions of nitrous oxide (N_2O) from a tidal, freshwater river, the Hudson River, New York. Environ Sci Technol 2001;35:991–6.

de Wilde HPJ, de Bie MJM. Nitrous oxide in the Scheldt estuary: production by nitrification and emission to the atmosphere. Mar Chem 2000;69:203–6.

Garnier J, Cébron A, Tallec G, Billen G, Sebilo M, Martinez A. Nitrogen behaviour and nitrous oxide emission in the tidal Seine River estuary (France) as influenced by human activities in the upstream watershed. Biogeochemistry 2006;77:305–26.

Gulati RD, van Donk E. Lakes in the Netherlands, their origin, eutrophication and restoration: State-of-the-art review. Hydrobiologia 2002;478:73-106.

Harris GP. Comparison of the biogeochemistry of lakes and estuaries: ecosystems processes, functional groups, hysteresis effects and interactions between macro- and microbiology. Mar Fresh Res 1999;50:791–811.

Harrison J, Matson P. Patterns and controls of nitrous oxide emissions from waters draining a subtropical agricultural valley. Glob Biogeochem Cycles 2003;17(3):1080. doi:10.1029/2002GB001991.

Hendzel LL, Matthews CJD, Venkiteswaran JJ, St. Louis VL, Burton D, Joyce EM, et al. Nitrous oxide flux in three experimental boreal forest reservoirs. Environ Sci Technol 2005;39:4,353–60.

Herbert RA. Nitrogen cycling in coastal marine ecosystems. FEMS Microbiol Rev 1999;23:563–90.

Houghton JT, Meira Filho LG, Lim B, Treanton K, Mamaty I, Bonduki Y, Griggs DJ, Callender BA, editors. Greenhouse Gas Inventory Reference Manual. IPCC WGI Technical Support Unit, vol. 3. Cambridge, UK: Cambridge University Press; 1996.

Huttunen JT, Alm J, Liikanen A, Juutinen S, Larmola T, Hammar T, et al. Fluxes of methane, carbon dioxide and nitrous oxide in boreal lakes and potential anthropogenic effects on the aquatic greenhouse gas emissions. Chemosphere 2003;52:609–21.

Khalil MAK, Rasmussen RA. The global sources of nitrous oxide. J Geophys Res 1992;97:14,651–60.

Knowles R. Denitrification: microbiology and ecology. Life Support Biosph Sci 1996;3:31–4.

Knowles R, Lean DRS, Chan YK. Nitrous oxide concentrations in lakes: variations with depth and time. Limnol Oceanogr 1981;26:855–66.

Li L, Li Y, Biswas DK, Nian Y, Jiang G. Potential of constructed wetlands in treating the eutrophic water: evidence from Taihu Lake of China. Bioresour Technol 2008;99:1656–63.

Liikanen A, Martikainen P. Effect of ammonium and oxygen on the methane and nitrous oxide fluxes across sediment–water interface in a eutrophic lake. Chemosphere 2003;52:1,287–93.

Luo L, Qin B, Zhu G, Zhang Y, Ji J. Current circulation pattern in winter Meiliang Bay, Lake Taihu. J Lake Sci 2004;16:73–6.

McCarthy MJ, Lavrentyev PJ, Yang L, Zhang L, Chen Y, Qin B, et al. Nitrogen dynamics and microbial food web structure during a summer cyanobacterial bloom in a subtropical, shallow, well-mixed, eutrophic lake (Lake Taihu, China). Hydrobiologia 2007;581:195–207.

McMahon PB, Dennehy KF. N_2O emission from a nitrogen-enriched river. Environ Sci Technol 1999;33:21–5.

Mengis M, Gachter R, Wehrli B. Sources and sinks of nitrous oxide (N_2O) in deep lakes. Biogeochemistry 1997;38:281–301.

Naqvi SWA, Jayakumar DA, Narvekar PV, Nalk H, Sarma VVSS, D'Souza W, et al. Increased marine production of N_2O due to intensifying anoxia on the Indian continental shelf. Nature 2000;408:346–9.

Qin B, Luo L. Changes in eco-environment and causes for Lake Taihu, China. Quat Sci 2004;24:561–8.

Schaller JL, Royer TV, David MB, Tank JL. Denitrification associated with plants and sediments in an agricultural stream. J. North Am. Benthol. Soc. 2004;23:667–76.

Seitzinger SP. Eutrophication and the rate of denitrification and N_2O production in coastal marine sediments. Limnol Oceanogr 1985;30:1,332–9.

Seitzinger SP, Kroeze C. Global distribution of nitrous oxide production and N inputs in freshwater and coastal marine ecosystems. Glob Biogeochem Cycles 1998;12:93-113.

Seitzinger SP, Kroeze C, Styles RV. Global distribution of N_2O emissions from aquatic systems: natural emissions and anthropogenic effects. Chemosphere, Glob. Chang. Sci. 2000;2:267–79.

Wang H, Wang H, Yin C, Wang Y, Lu J. Littoral zones as the “hotspots” of nitrous oxide (N_2O) emission in a hyper-eutrophic lake in China. Atmos Environ 2006;40:5,522–7.

Wang, S., Yeager, K.M., Wan, G., Liu, C., Tao, F., Lǚ, Y., et al., 2008. Short term field observations of nitrous oxide saturations and fluctuations in a shallow eutrophic lake: Lake Taihu, China. Limnology. In review.

Wanninkhof R. Relationship between wind speed and gas exchange over the ocean. J Geophys Res 1992;97:7,373–82.

Weathers PJ. N_2O evolution by green algae. Appl Environ Microbiol 1984;48:1,251-1,253.

Xing G, Cao Y, Shi S, Sun G, Du L, Zhu J. N pollution sources and denitrification in waterbodies in Taihu Lake region. Science in China (series B) 2001;44:304–14.

Yoh M, Terai H, Saijo Y. A preliminary study on N_2O production through nitrification in Lake Kizaki. Japanese J Limnol 1988;49:43–6.

Zhai S, Zhang H. Water quantity and waste load variation of rivers around Lake Taihu from 2000 to 2002. J Lake Sci 2006;18:225–30.

Zheng X, Wang M, Wang Y, Shen R, Gong Y, Zhang W, et al. N_2O emission from rice–wheat ecosystem in Southeast China. Chin J Appl Ecology 1997;8:495–9.