

## Nitrous oxide emission from the littoral zones of the Miyun Reservoir near Beijing, China

Hongli Li, Meng Yang, Ting Lei, Mingxiang Zhang, Peter Bridgewater, Cai Lu, Xuemeng Geng and GuangChun Lei

### ABSTRACT

Large dams may be substantial contributors to greenhouse gas emissions. Nitrous oxide (N<sub>2</sub>O) is the third most important greenhouse gas but studies on N<sub>2</sub>O emission from reservoirs are limited. We measured N<sub>2</sub>O emissions and environmental factors including atmospheric pressure, wind speed, air and soil/sediment temperature, biomass, soil water content and organic matter, total nitrogen, NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N of soil, from the littoral zones of the Miyun Reservoir, near Beijing, China, in January, May, June, August, and October during 2009 and 2010. Using the static chamber method we investigated the seasonal and spatial variation, relating it to environmental factors. Spatial and temporal variations in N<sub>2</sub>O flux appeared to be influenced by several environmental factors, working singly or in conjunction, including soil water depth, soil nutrition, biomass, and wind speed. In winter and spring, high N<sub>2</sub>O emissions (up to  $1.9 \pm 0.6$  mg N<sub>2</sub>O m<sup>-2</sup> h<sup>-1</sup>) were recorded at both eulittoral and infralittoral zones, while the flux from the supralittoral zone was low during all the seasons (from -0.04 to 0.01 mg N<sub>2</sub>O m<sup>-2</sup> h<sup>-1</sup>). This study suggests that the littoral zone is a substantial source of N<sub>2</sub>O. However, its spatiotemporal variation and environmental drivers are still not clear.

**Key words** | biomass, littoral zone, N<sub>2</sub>O flux, soil nutrients, soil water content, wind

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### INTRODUCTION

Nitrous oxide (N<sub>2</sub>O) is considered to be the third most important greenhouse gas (Hernandez & Mitsch 2006; Ravishankara *et al.* 2009) with the current atmospheric concentration of 324 ppb and a warming potential of approximately 298 times that of CO<sub>2</sub> over a 100-year time span (IPCC 2013). Over a decadal time scale, the atmospheric concentration of N<sub>2</sub>O has been increasing by 0.2–0.3% each year, mainly due to human perturbation of the global nitrogen cycle (e.g., Chen *et al.* 2011). It is believed that global warming and the deposition of nitrogen are increasing the flux of N<sub>2</sub>O from boreal lakes, and that such processes accelerate the release of N<sub>2</sub>O through positive feedback mechanisms (Liikanen *et al.* 2002; Cantarel *et al.* 2011; McCrackin & Elser 2011).

Wetland ecosystems have been suggested as important sources of N<sub>2</sub>O emission; in particular, many authors have focused on rice paddy wetlands which are a major N<sub>2</sub>O

source (Kreye *et al.* 2007; Li *et al.* 2009). Limited studies on N<sub>2</sub>O emission of natural wetlands show big spatial variations. Nitrous oxide emission from rivers appears to be much lower than estuaries, contributing for example only 0.5–12.5% of the total N<sub>2</sub>O emission from some English and Welsh rivers and estuaries (Dong *et al.* 2004). Open water areas of natural lakes do not contribute significantly to N<sub>2</sub>O emissions either (Mengis *et al.* 1997), but N<sub>2</sub>O emissions from their littoral areas maintain relatively high levels (Senga *et al.* 2001; Huttunen *et al.* 2003; Wang *et al.* 2006).

Reservoirs are an important type of artificial wetland. The global area of reservoirs was estimated as 251,000 km<sup>2</sup> (Lehner & Döll 2004) but may be much higher now. In 2000, 46% of all large dams in the world were found in China (Dams 2000). Under global climate change mitigation strategies, hydropower has often been considered as a clean energy source compared to fossil

fuel; however, greenhouse gas emissions from reservoirs may have been underestimated (Fearnside 2002).

Reservoirs are associated exclusively with human development (Stadmark & Leonardson 2005) and are intimately linked to water dynamics, which in turn influences the nitrogen cycle, as a key controlling factor in N<sub>2</sub>O emissions (Hefting et al. 2004; Teiter & Mander 2005; Hernandez & Mitsch 2006). There have been an increasing number of studies on greenhouse gases released from reservoirs (e.g., Barros et al. 2011; Wehrli 2011). Most of these studies, however, have quantified emissions of CO<sub>2</sub> and CH<sub>4</sub>, with little focus on N<sub>2</sub>O.

Greenhouse gas emissions from reservoirs in the tropics have attracted much attention (Galy-Lacaux et al. 1999; Rosa et al. 2004; Guérin et al. 2008) and reservoirs in other climatic regions have also been studied (Diem et al. 2008, 2012; Deemer et al. 2011). Lowland reservoirs were found to be major sources of N<sub>2</sub>O, while sub-alpine and alpine reservoirs appeared to be in equilibrium with atmospheric concentrations (Diem et al. 2012). Rather little is known about N<sub>2</sub>O emission from reservoirs in temperate regions, with data especially lacking on seasonal and diurnal patterns. Nitrous oxide production is influenced by a number of factors, such as nitrate concentration, organic matter availability, and pH (Hefting et al. 2003; Wu et al. 2009; Baulch et al. 2011). Reservoirs can include a wide range of primary production and redox conditions, so that N<sub>2</sub>O flux studies in reservoirs, in relation to environmental conditions, may contribute to a better overall understanding of the N<sub>2</sub>O flux in aquatic systems as well as contributing to our knowledge of the total greenhouse gas emissions associated with construction and use of reservoirs.

The objectives of the present study are: (1) to understand how N<sub>2</sub>O emitted from the Miyun Reservoir in China varies spatially and temporally; and (2) to assess key environmental factors contributing to any spatiotemporal variation in emission patterns.

## METHODS

### Study site

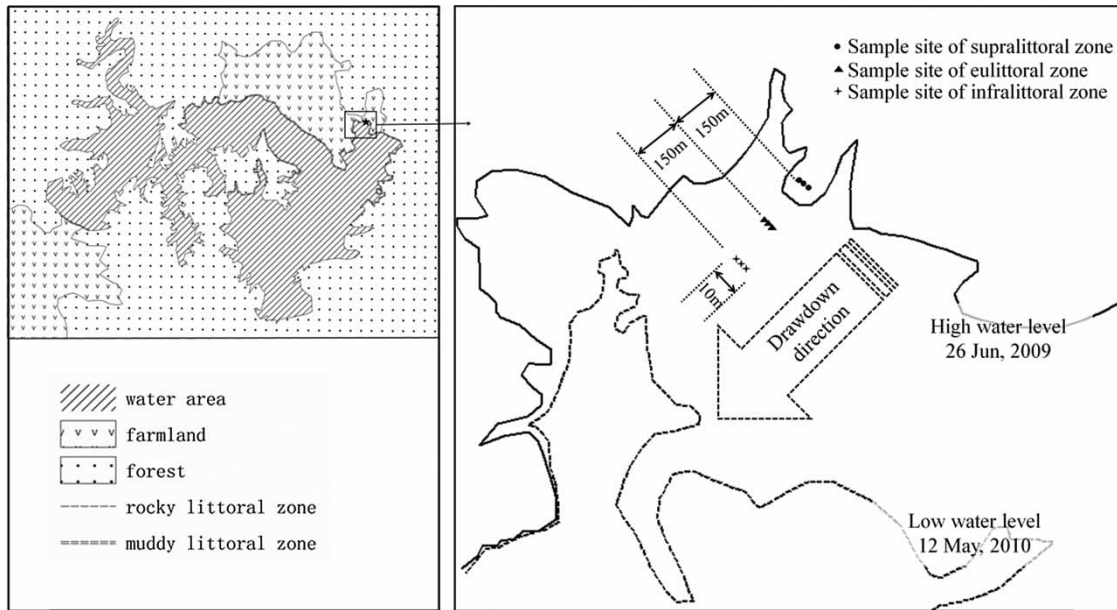
The research was conducted at Miyun Reservoir (40°29'N, 116°50'E) near Beijing, China during 2009–2010. The

reservoir is a 188 km<sup>2</sup> water body and is up to 60 m deep. It has a capacity of 4.4 billion m<sup>3</sup> and is the most important water resource for Beijing. The reservoir was constructed in 1960 by clearing vegetation and building a 66 m dam. Inflows to the reservoir are mainly from the Chao and Bai Rivers. The river catchments are characterized by warm-temperate semi-humid monsoonal climatic conditions with an average annual precipitation of 669 mm, with 80% of precipitation falling between July and September. The annual average regional air temperature, lowest air temperature, and highest air temperature is, respectively, 10.6 °C, –18 °C and 38 °C. There are, on average, 176 days annually without frost (Xie & Wang 2006). The reservoir is covered by ice approximately from mid-November to the end of March (Gao 1989). The littoral area includes rocky parts without vegetation and other parts which are muddy and vegetated. Eighty percent of the littoral area is muddy (Guan et al. 2007). Most of the muddy littoral area is distributed at the northern part of the reservoir (Figure 1), and we chose to study this part of the reservoir because it had the more typical habitat.

The study area was divided into three zones (Figure 1) delimited by water level and vegetation. The three zones were termed supralittoral (rarely inundated), eulittoral (intermittently inundated), and infralittoral (emergent only at extremely low water level), respectively (Wang et al. 2006). During the sampling period, the water level declined around two meters continuously. Because the slope was gentle, the water level fluctuation affects a large area. Details of the water depths in each of these zones are shown in Figure 2. The vegetation of the supralittoral zone was dominated by *Artemisia capillaries* Thunb., *Eleusine indica* (L.) Gaertn, *Roegneria kamoji* Ohwi, the eulittoral zone by *Medicago sativa* L. and *Humulus scandens* (Lour.) Merr., and that of the infralittoral zone by *Typha orientalis* Presl., *Echinochloa caudata* Roshev, and *Echinochloa crusgalli* L. For more details on weather, biomass, soil water content (SWC), and soil C/N, see Figure 2 and Table 1.

### Flux measurements of N<sub>2</sub>O

Nitrous oxide samples for flux measurements were collected during the period from June 2009 to May 2010 (five collections over a 1-year period). For each of the three zones



**Figure 1** | Map of Miyun Reservoir and location of field plots.

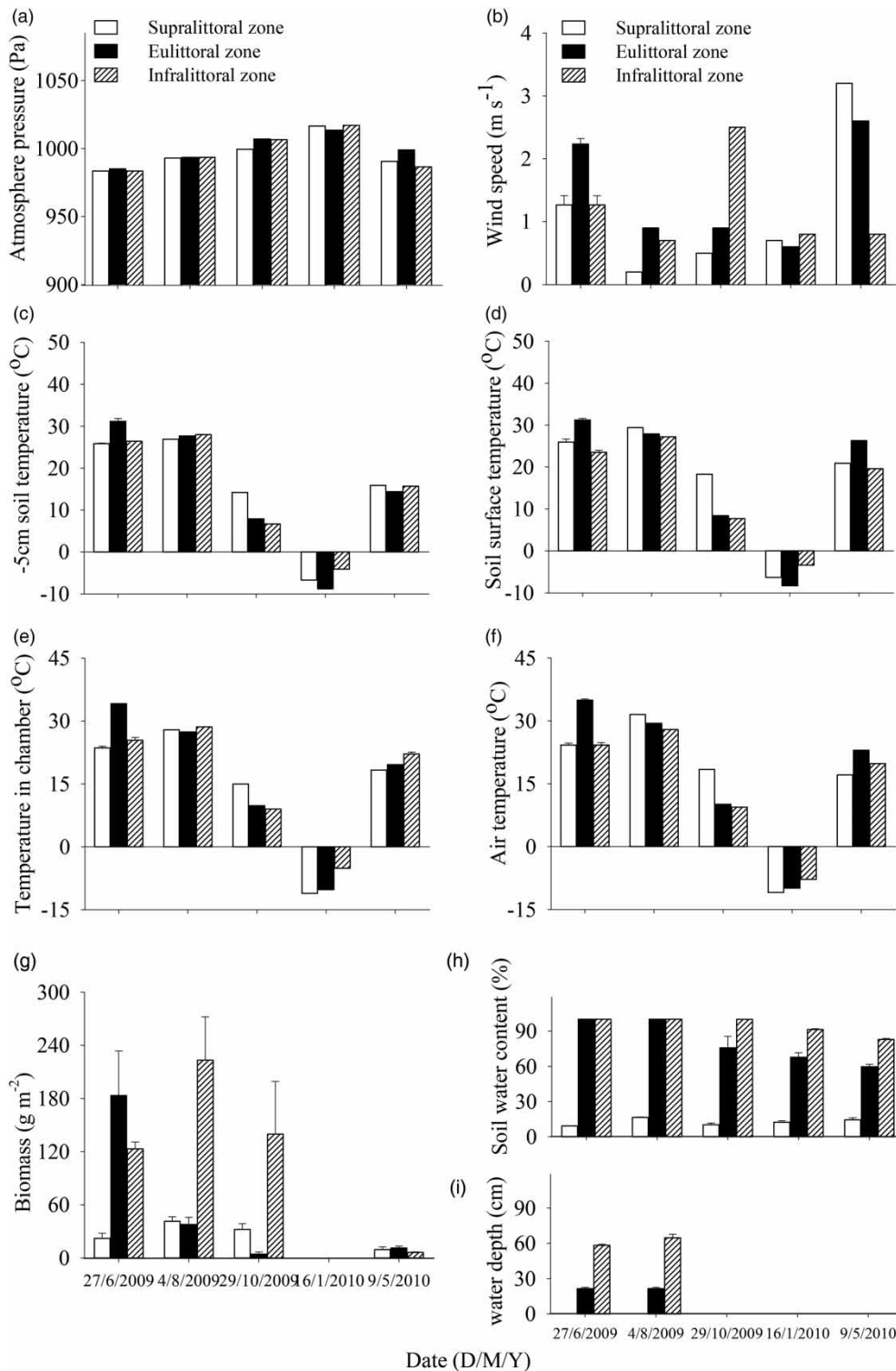
sampled, three replicate plots were established ( $n = 3$ ). To avoid disturbance to the soil, especially trampling pressure, and compaction of soil, a wooden platform was built. All samples were collected at the same time of day (09:00–09:30) in June, August, October 2009, January, and May 2010. To analyze diurnal variation, samples were collected at 3-hour intervals on 4 May, 11 August, and 25 October. Each sampling was completed in a 3-day period, with one zone sampled per day.

The closed and opaque chamber technique was applied (Chen *et al.* 2011), with three replicate chambers (not far from 10 m from each other) at each sampling site. The stainless steel chamber consisted of two parts, a pedestal and an upper chamber. The pedestal (length, width, and height: 50 cm × 50 cm × 20 cm) had a gutter around the outside in the upper rim that could be filled with water to make an airtight seal with the upper chamber. When there was standing water, four length adjustable legs were equipped onto the pedestal in order to avoid gutter submerging. An internal chamber (length, width, and height: 50 cm × 50 cm × 80 cm) could be added to extend the height if plants were tall. The upper chamber (length, width, and height: 50 cm × 50 cm × 50 cm) was equipped with two fans for air mixing. Pedestals were inserted into the soil/sediment in the sampling area the night before measurement to allow

settling. Four air samples (200 mL for each) from each chamber were collected with a 100 mL plastic syringe at 10 min intervals over a 30 min period after enclosure and stored in the four 500 mL bags separately made of plastic and aluminum membrane material which is impermeable to gas (Guangming Research and Design Institute of Chemical Industry, China). All samples were analyzed for  $N_2O$  concentration by gas chromatography (7890A, Agilent, USA) within 7 days. The instrument was equipped with an electron capture detector and gases were separated with a column (length: three meters; diameter: 3.2 mm) packed with Porpak Q (80/100 mesh). The temperatures of oven, injector, and detector were 70 °C, 20 °C, and 330 °C, respectively. The flow rate of carrier gas ( $N_2$ ) was 25 mL  $min^{-1}$ . Standard  $N_2O$  gas (303 ppb in air, China National Research Center for Certified Reference Materials, China) was used for precision verification between every eight samples for  $N_2O$  concentration. The coefficient of variation was below 1.5%. The flux of  $N_2O$  ( $F$ ) was calculated as (Chen *et al.* 2011)

$$F = \frac{M}{V_0} \times \frac{P}{P_0} \times \frac{T_0}{T} \times \frac{dC_t}{dt} \times H \quad (1)$$

where  $F$  is the flux of  $N_2O$  ( $mg\ m^{-2}\ h^{-1}$ );  $M$  is the molar mass of  $N_2O$  ( $g\ mol^{-1}$ );  $P$  (kPa) is the atmosphere pressure



**Figure 2** | The environmental parameters including atmosphere pressure, wind speed, soil temperature at 5 cm below the surface (–5 cm soil temperature), soil surface temperature, temperature in chamber, air temperature, biomass, SWC, and water level of different sample zones and sample months from 2009 to 2010 (mean  $\pm$  SE,  $n = 3$ ). Several SE bars are not visible.

**Table 1** | The soil parameters including SOM, TN, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> content of different sample sites are shown (mean ± SE)

Variable	Supralittoral zone	Eulittoral zone	Infralittoral zone
SOM (g kg <sup>-1</sup> )	18.4 ± 3.6	13.0 ± 3.0	35.8 ± 1.4
TN (g kg <sup>-1</sup> )	1.0 ± 0.2	0.8 ± 0.1	2.1 ± 0.1
NO <sub>3</sub> <sup>-</sup> (mg kg <sup>-1</sup> )	4.1 ± 1.9	2.2 ± 1.0	12.1 ± 5.9
NH <sub>4</sub> <sup>+</sup> (mg kg <sup>-1</sup> )	10.9 ± 1.5	7.9 ± 0.7	16.3 ± 0.3

of the sampling site;  $T$  (K) is the absolute temperature of the sampling time;  $V_0$  (22.4 L),  $P_0$  (101.325 kPa), and  $T_0$  (273.15 K) is the molar volume, atmosphere pressure, and absolute temperature, respectively, under standard conditions;  $dC_t/dt$  (ppm h<sup>-1</sup>) is the rate of concentration change; and  $H$  (m) is the chamber height over the water or soil surface.

### Monitoring of environmental parameters

While collecting N<sub>2</sub>O samples in the field, several environmental parameters were also measured. Atmospheric pressure was measured with a barometer (DYM3, Baoping, China), and wind speed with an anemometer (4101, Testo, Germany) at the top of the chamber during sampling. Soil or sediment temperature at 5 cm below the surface, soil or sediment surface temperature, and air temperature inside and outside the chamber were recorded using a portable digital thermometer (JM624, Jinming, China) during collection of the gas samples.

After gas sampling, above-ground biomass was determined from three replicate harvests of 50 cm × 50 cm in June, August, October, and May, after drying at 80 °C to constant weight.

Soil water content of plots without standing water was measured using a Soil Water Sensor (UNI1000, Shunlong, China). Water depth of each of the plots with standing water was measured with a meter ruler at the same time that the gas samples were collected.

Soil samples of each replicate of the three areas were collected at 0–10 cm depth in May 2010. Total nitrogen (TN) content and soil organic matter (SOM) were monitored by the semi-micro Kjeldahl procedure and the potassium-dichromate oxidation procedure (Semenov *et al.* 2010). NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N were monitored by the potassium

chloride-indophenol blue colorimetric method and the calcium sulfate disulfonic acid method (Lu 1999; Chen *et al.* 2010).

### Statistical analysis

Two-way analysis of variance (ANOVA) was used to analyze the effects of sampling month, sampling zone, and the interaction of these two factors on N<sub>2</sub>O emission and environmental factors, including SWC, biomass, wind speed, soil temperature at 5 cm below the surface, soil surface temperature, and temperatures inside and outside the chamber in each sample zone. A one-way ANOVA was used to compare the fluxes among different sites in different months, and a Duncan's test was conducted to examine the difference in N<sub>2</sub>O flux among the treatments. Spearman correlation analysis was applied to determine the effect of environmental parameters on the N<sub>2</sub>O emission rate.

The relationship between the SWC and the N<sub>2</sub>O flux of each zone was analyzed separately by linear regression analysis. The SWC was considered 100% when there was water above the soil surface.

The effect of any variable was considered statistically significant if  $P < 0.05$ . All the analyses above were performed using IBM SPSS Statistics (19.0, IBM, USA). Charts were made using SigmaPlot (11.0, SYSTAT, USA) and Microsoft Excel (2010, Microsoft, USA).

## RESULTS

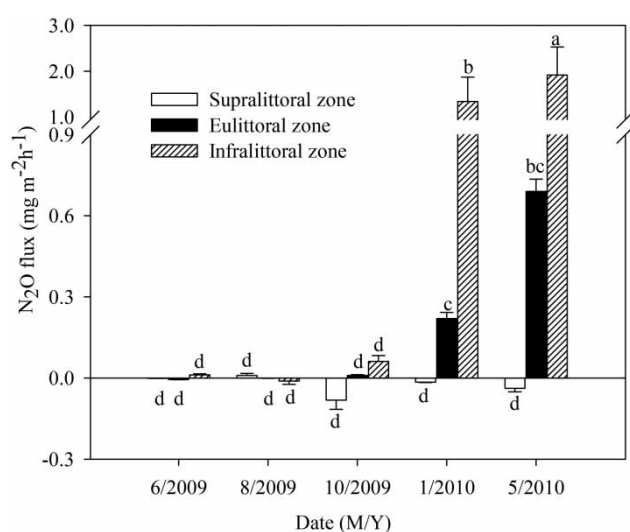
### Monthly and spatial variation in N<sub>2</sub>O flux

Two-way ANOVA analysis showed that month, sampling zone, and the interaction between these two factors all significantly affected N<sub>2</sub>O flux ( $P < 0.05$ ; Table 2). Nitrous oxide emissions from all sampling zones in June, August, and October 2009 were significantly lower than in January and May 2010 except in the supralittoral zone. There was no significant difference among the sample zones for the June to October period ( $P > 0.05$ ), although we noted that the flux was negative (i.e., N<sub>2</sub>O was absorbed) at the supralittoral zone in October (Figure 3). Nitrous oxide flux from the infralittoral zone in May was  $1.9 \pm 0.6$  mg m<sup>-2</sup> h<sup>-1</sup>,

**Table 2** | Two-way ANOVA of N<sub>2</sub>O flux and environmental factors from different months and sampling zones

	Month (M)		Sampling zones (S)		M × S	
	F <sub>4,30</sub>	P	F <sub>2,30</sub>	P	F <sub>8,30</sub>	P
N <sub>2</sub> O flux (mg m <sup>-2</sup> h <sup>-1</sup> )	6.8	0.001	9	0.001	3.2	0.001
Soil water content (%)	25.5	<0.001	1178	<0.001	12.5	<0.001
Biomass (g m <sup>-2</sup> )	6.4	0.01	6.6	0.04	3.2	0.1
Soil temperature at 5 cm below the surface (°C)	19801.2	<0.001	28.2	<0.001	247.2	<0.001
Soil surface temperature (°C)	10649.9	<0.001	183.3	<0.001	263.9	<0.001
Temperature inside chamber (°C)	13880.2	<0.001	60	<0.001	267.4	<0.001
Temperature outside chamber (°C)	16109.9	<0.001	210.1	<0.001	363.9	<0.001
Wind speed (m s <sup>-1</sup> )	391.7	<0.001	32.7	<0.001	226.7	<0.001

Note: See Figures 2 and 3 for data.



**Figure 3** | The N<sub>2</sub>O fluxes of different sample zones and sample months from 2009 to 2010 (mean ± SE). Several SE bars are not visible. Bars with different letters a, b, c, and d indicate significant difference in rates of N<sub>2</sub>O (Duncan's test,  $P < 0.05$ ,  $n = 3$ ).

which was significantly higher ( $P < 0.05$ ) than that from the eulittoral zone ( $0.7 \pm 0.05 \text{ mg m}^{-2} \text{ h}^{-1}$ ), and the supralittoral zone, which was lower than zero ( $-3.8 \pm 1.2 \times 10^{-2} \text{ mg m}^{-2} \text{ h}^{-1}$ ). The same pattern was found in January for the different sampling zones (Figure 3).

### Diurnal variation in N<sub>2</sub>O flux

Daily variation in N<sub>2</sub>O flux varied by both zone and time of year (Figure 4). For the supralittoral zone, in August the flux peaked at 09:00, then declined and was maintained at a

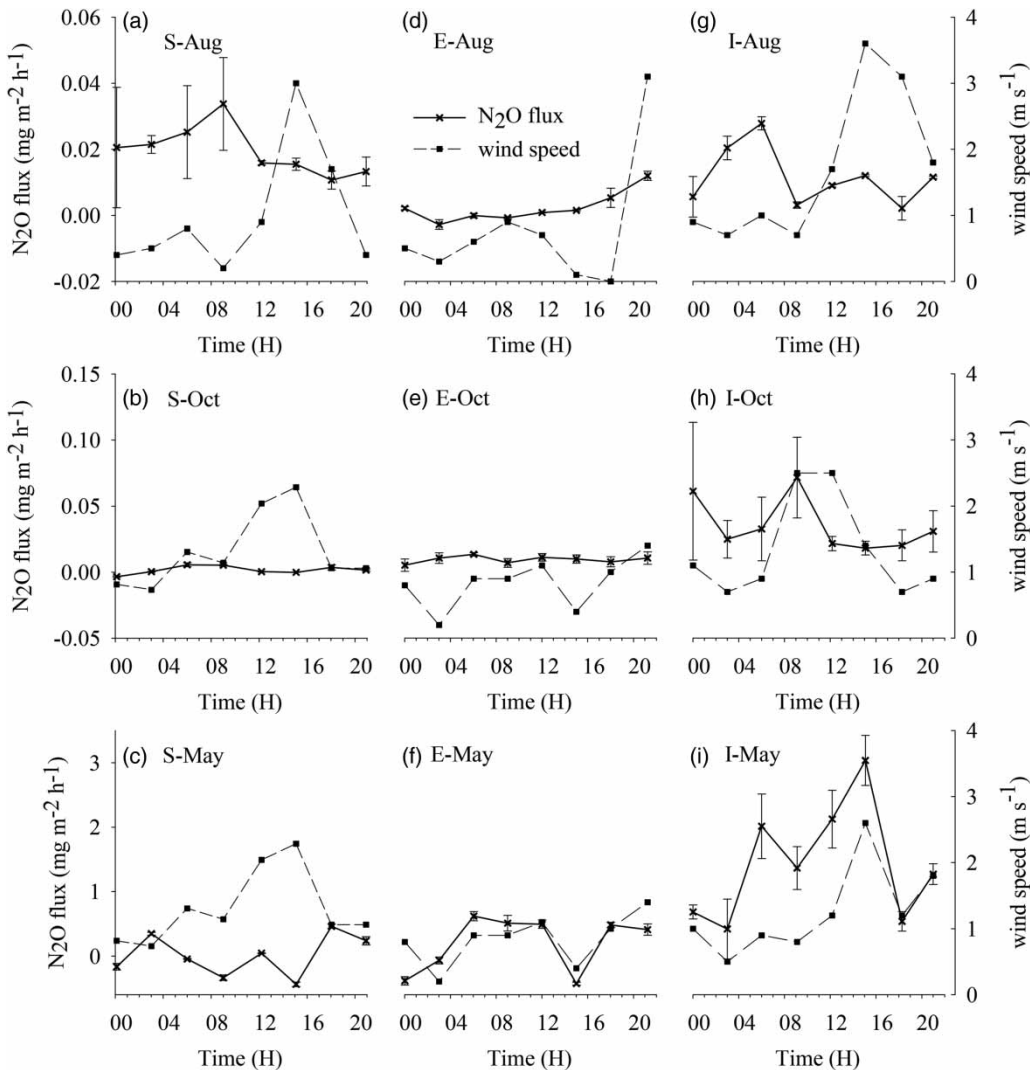
slightly lower level until the next morning (Figure 4(a)). In October, the N<sub>2</sub>O flux remained near zero at the same zone (Figure 4(b)). However, this pattern changed greatly in May. Emissions first increased and peaked at 03:00, followed by a decline until 15:00, increasing again to reach its peak at 18:00 (Figure 4(c)).

For the eulittoral zone, nitrous oxide flux increased consistently from 03:00 peaking at 21:00 in August, compared to a relatively stable high emission level in October (Figures 4(d) and 4(e)). However, in May, N<sub>2</sub>O flux increased from 00:00 to 06:00, then increased and remained stable at a high level until 12:00; then, suddenly dropping to the lowest level at 15:00 (Figure 4(f)).

For the infralittoral zone, nitrous oxide flux first reached a peak at 06:00, and decreased to a relatively stable condition in August, but in October it peaked later, at 09:00 before decreasing to a stable level (Figures 4(g) and 4(h)). In May the flux was significantly higher and peaked at 15:00 (Figure 4(i)).

### Key factors affecting N<sub>2</sub>O emission

The sampling month, sampling zone, and interaction of both factors significantly affected N<sub>2</sub>O emission (Table 2). We found environmental factors, including SWC, biomass, soil temperature at 5 cm below soil surface, soil surface temperature, air temperature within and outside the chamber, and wind speed were significantly different among seasons and among littoral zones (Table 2). Especially, the diurnal



**Figure 4** | Diurnal variation in  $\text{N}_2\text{O}$  flux and wind speed in different sample zones in May, August, and October (mean  $\pm$  SE). Several SE bars are not visible. (a), (b), and (c) show the supralittoral zone; (d), (e), and (f) show the eulittoral zone; (g), (h), and (i) show the infralittoral zone ( $n=3$ ).

variation of wind speed presented the same pattern as the  $\text{N}_2\text{O}$  flux in May in the eulittoral and infralittoral zones (Figure 4). The environmental parameters are shown in Figure 2. The atmosphere pressure was nearly the same at all the plots and months (Figure 2(a)), and the seasonal wind speed was not regular (Figure 2(b)). The temperature was the same pattern in all cases (Figures 2(c)–2(f)). Biomass from the infralittoral zone in August was the highest among all sites and months (Figure 2(g)). The SWC and water level were different among different sites and months (Figures 2(h) and 2(i)). The soil parameters,

including SOM, TN,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  content were the highest in infralittoral zone (Table 1).

The Spearman correlation analysis indicated negative relationships between  $\text{N}_2\text{O}$  emission and temperature and biomass (Table 3). There was no significant correlation between SWC and flux in infralittoral zone ( $R^2=0.13$ ,  $P>0.05$ ,  $df=4$ ; Figure 5) when SWC was lower than 20%. Negative correlations appeared when SWC was higher than 60% both in the eulittoral zone ( $R^2=0.62$ ,  $P>0.05$ ,  $df=4$ ) and the infralittoral zone ( $R^2=0.97$ ,  $P<0.01$ ,  $df=4$ ), but it was not significant in the eulittoral zone.

**Table 3** | Spearman correlation analysis between N<sub>2</sub>O flux and environmental factors are shown

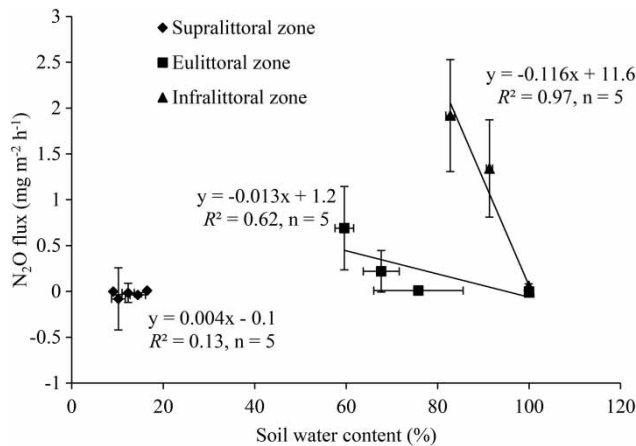
	N <sub>2</sub> O F	Atmos P	Wind S	– 5 cm T	Sur T	Cham T	Air T	Biomass	SWC	SOM	TN	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>
N <sub>2</sub> O F	1												
Atmos P	0.28	1											
Wind S	-0.03	-0.36*	1										
– 5 cm T	-0.41**	-0.73**	0.18	1									
Sur T	-0.32*	-0.71**	0.16	0.95**	1								
Cham T	-0.44**	-0.69**	0.15	0.95**	0.95**	1							
Air T	-0.32*	-0.75**	0.08	0.95**	0.98**	0.95**	1						
Biomass	-0.34*	-0.57**	0.13	0.74**	0.69**	0.80**	0.72**	1					
SWC	-0.01	-0.12	0.21	0.40**	0.30*	0.37*	0.34*	0.45**	1				
n	45	45	45	45	45	45	45	45	45				
SOM	0.47	-0.84**	-0.58	0.26	-0.84**	-0.84**	-0.26	-0.65	0.55	1			
TN	0.53	-0.79*	-0.63	0.16	-0.79*	-0.79*	-0.16	-0.6	0.57	0.98**	1		
NO <sub>3</sub> <sup>-</sup>	0.45	-0.79*	-0.47	0.32	-0.79*	-0.79*	-0.32	-0.65	0.36	0.87**	0.88**	1	
NH <sub>4</sub> <sup>+</sup>	0.52	-0.9**	-0.53	0.37	-0.9**	-0.9**	-0.37	-0.65	0.42	0.77*	0.78*	0.83**	1
n	9	9	9	9	9	9	9	9	9	9	9	9	9

Notes: \*\* Indicates significant correlation at  $P < 0.001$ ; \* indicates significant correlation at  $P < 0.05$ .

N<sub>2</sub>O F, N<sub>2</sub>O flux; Atmos P, atmosphere pressure; Wind S, wind speed; –5 cm T, soil temperature at 5 cm below the surface; Sur T, soil surface temperature; Cham T, temperature inside chamber; Air T, temperature outside chamber; SWC, soil water content; SOM, soil organic matter; TN, soil total nitrogen; NO<sub>3</sub><sup>-</sup>, soil NO<sub>3</sub><sup>-</sup>; NH<sub>4</sub><sup>+</sup>, soil NH<sub>4</sub><sup>+</sup>; n, sample size.

The horizontal line after the first n is a distinguishing mark for sampling sizes of different environmental factors.





**Figure 5** | The linear regression analysis between SWC and N<sub>2</sub>O flux of different sample zones. Several SE bars are not visible.

## DISCUSSION

The present study showed significant variation in the spatial and temporal N<sub>2</sub>O flux (Table 2). Most notably, the mean N<sub>2</sub>O emissions recorded in the eulittoral and infralittoral zones were very high in January and May 2010 (Figure 3) when the SWC was low and there was little biomass (Figure 2).

From other work, SWC caused by variation in the level of groundwater is considered to be the dominant environmental factor affecting N<sub>2</sub>O flux (Breuer *et al.* 2000; Hefting *et al.* 2004; Hernandez & Mitsch 2006). For example, N<sub>2</sub>O emissions slowly increased with the SWC and water depth of the littoral marshes at the Three Gorges Reservoir (Chen *et al.* 2010). In our work, the N<sub>2</sub>O emission pattern emerges when the data are presented on an axis of soil moisture (Figure 5). The different patterns of emission of the three littoral zones shown in this graph might be attributed to soil anoxic conditions, which is a strong function of soil water. Facultative anaerobic microbes will not begin denitrification until O<sub>2</sub> levels are insufficient (Batson *et al.* 2012). SWC normally affected both denitrification and N<sub>2</sub>O production (Senga *et al.* 2002; Dhondt *et al.* 2004). Both low levels of N<sub>2</sub>O flux and SWC suggested that the extreme low emission rates from the supralittoral zone might be caused by high available O<sub>2</sub> in the soil pore space. In our study, N<sub>2</sub>O flux showed negative correlations when it was higher than 60% (Figure 5). These results agreed with research showing that N<sub>2</sub>O emission and

SWC presented a parabolic correlation; and when the SWC was high, the strongly reductive condition in the soil allowed the complete reduction to N<sub>2</sub>, resulting in the decrease of N<sub>2</sub>O emission (Ciarlo *et al.* 2007). We propose that soil moisture explains the difference in N<sub>2</sub>O emission between June 2009 and May 2010, and also the higher N<sub>2</sub>O emission of winter (January 2010) than summer (August 2009). However, different results were obtained in a previous field study. A study carried out at riparian northern hardwood forests showed that there was no regular pattern between N<sub>2</sub>O flux and soil moisture (0.67 to 2.8 g H<sub>2</sub>O g dry soil<sup>-1</sup>) and instead flux was strongly related to percent plant cover (Hopfensperger *et al.* 2009). Although many studies have pointed out the importance of SWC to N<sub>2</sub>O flux, further field and laboratory work is needed to explain these inconsistent results.

Soil nutrients were considered to be the most likely explanation for higher emission from the infralittoral zone than the eulittoral zone. Denitrification is the only process that permanently reduces NO<sub>3</sub><sup>-</sup> to N<sub>2</sub> or N<sub>2</sub>O. The NO<sub>3</sub><sup>-</sup> ion is utilized as a terminal electron acceptor by facultative anaerobic bacteria during respiration in the absence of O<sub>2</sub> (Batson *et al.* 2012). Soil nitrogen, carbon, and C/N ratio have been found to be significantly correlated with denitrification potentials (Wu *et al.* 2013). Variation of denitrification primarily depends on nitrate (Song *et al.* 2012) and carbon availability (Song *et al.* 2011). Work at the Meiliang Bay of Taihu Lake, China, suggested hyper-eutrophication contributed to N<sub>2</sub>O emission (Wang *et al.* 2006). Similar results were found in a series of studies at the Yangtze River Delta, Huangpu River, and Chong-ming islands in the Yangtze River estuary (Hefting *et al.* 2003; Tomaszek & Czerwieniec 2003; Dong *et al.* 2004; Wang *et al.* 2009; Wu *et al.* 2009). Basing on such findings, we speculated the higher emission from the infralittoral zone than the eulittoral zone, at least in May, was attributed partly to the 2.1–5.5 times higher SOM and TN (Table 1). Moreover, spring rains may activate microbes (Breuer *et al.* 2000) or simply displace the greenhouse gas already stored in pore spaces (known as the ‘Birch effect’) (Unger *et al.* 2010).

The presence or absence of vegetation may affect emission rates in several ways. In the present study, low biomass was observed during high emission periods of January and May (Figure 2(g)). At these times, plants were either

dormant or commencing growth, and so there is no strong competition with microbes for nitrogen use as there inevitably is during the period of growth (Zhang *et al.* 2009). Thus there would be more available nutrients left for N<sub>2</sub>O production, which might partly contribute to the higher emission of eulittoral and infralittoral zones in winter and spring. A similar observation was also shown in the study by Berglund & Berglund (2011) that N<sub>2</sub>O emission from cultivated peat soils with *Lolium perenne* peaked in springtime. Moreover, decomposition of residues of aquatic plants after the water level drops might supply substrates for N<sub>2</sub>O production, as at the Pantanal tropical wetland (Liengaard *et al.* 2013). Plant decomposition might also be the reason for the pattern of soil nutrients (Table 1). The biomass in the infralittoral zone was largest, where fixed carbon and nitrogen accelerated soil nutrient accumulation (Laurance *et al.* 1999; Sun *et al.* 2011). Although the biomass of the eulittoral zone was higher than that of the supralittoral zone, frequent water level fluctuation might aggravate the loss of nutrients through mineralization, nitrification, denitrification, and volatilization (Reddy *et al.* 1984), since the eulittoral zone is located at the edge of water and the higher ground.

Temporal variation in N<sub>2</sub>O emissions from soil can often be attributed to changes in temperature (Senga *et al.* 2002; Hefting *et al.* 2003; Dhondt *et al.* 2004). Some studies showed that temperature is positively correlated with N<sub>2</sub>O flux (Chen *et al.* 2010, 2011; Cantarel *et al.* 2011). However, negative correlation between N<sub>2</sub>O emission and soil and air temperature was observed in the present study (Table 3), which agreed with at least two other studies, one at a headwater stream and the other at a temperate farmland (Beaulieu *et al.* 2008; Cosentino *et al.* 2012). We propose this uncommon correlation occurs because the positive effect of temperature on decomposition is masked by even stronger effects from SWC and soil nutrients as discussed above.

Temperature is considered to be an important factor affecting N<sub>2</sub>O diurnal variation (Smith *et al.* 1998). However, we found wind speed could be the other factor influencing N<sub>2</sub>O flux (Figure 4). As shown in Figure 4, N<sub>2</sub>O flux totally or partly changed corresponding with wind speed especially in the period with high N<sub>2</sub>O flux. Wind affects air exchange velocity between soil/water and atmosphere (Wanninkhof & McGillis 1999; Reicosky *et al.*

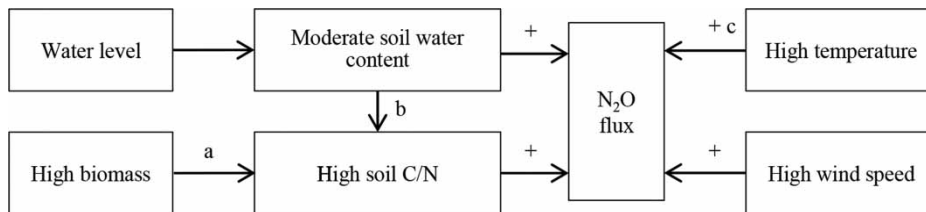
2008), partly by increasing diffusion at the surface and a 'bel-lows effect' causing mass flow between pore spaces and the atmosphere through pressure fluctuations (Reicosky *et al.* 2008). Reicosky *et al.* (2008) showed that wind influences gas concentration at 30 cm depth in the soil which implies that a strong ventilation process is commonplace. Wind also is found to have an influence on soil respiration when using closed dynamic chambers (Bain *et al.* 2005). However, there is a lack of direct and specific research on the influence of wind on the flux measured by static chambers.

The average N<sub>2</sub>O emission rate from the littoral zone during this sampling period was calculated as 0.243 mg m<sup>-2</sup> h<sup>-1</sup> (varying from -0.1 to 1.9 mg m<sup>-2</sup> h<sup>-1</sup>), which was similar to that found in previous studies in the littoral zone of lakes (Wang *et al.* 2006), but it is one to three times higher than the non-littoral areas of other reservoirs (Liu *et al.* 2011; Diem *et al.* 2012). That difference might be caused by the influence of water level and vegetation, as discussed above. This emphasizes the importance of the littoral zone to N<sub>2</sub>O emission from wetlands.

Previous studies provide data on other greenhouse gases from the Miyun Reservoir. Yang *et al.* (2011) found an average CH<sub>4</sub> flux of 3.2 mg m<sup>-2</sup> h<sup>-1</sup> and a CO<sub>2</sub> flux of 315 mg m<sup>-2</sup> h<sup>-1</sup>. Considering the greenhouse effect of N<sub>2</sub>O is approximately 296 times that of CO<sub>2</sub>, and CH<sub>4</sub> is approximately 23 times that of CO<sub>2</sub> (IPCC 2001), nitrous oxide emission at the Miyun Reservoir was equivalent to 72 mg m<sup>-2</sup> h<sup>-1</sup> CO<sub>2</sub>, whereas the observed CH<sub>4</sub> emission at the same site was equivalent to 74 mg m<sup>-2</sup> h<sup>-1</sup> CO<sub>2</sub>, showing the N<sub>2</sub>O emission at Miyun Reservoir represents a similar greenhouse effect as CH<sub>4</sub> at the same zone.

## CONCLUSIONS

Spatial and temporal variations in N<sub>2</sub>O may be influenced by several environmental factors, working singly or in conjunction, including soil water depth, soil nutrition, and biomass (Figure 6). This study provides some evidence of the underlying mechanisms which may help develop better management of the littoral zones at Miyun Reservoir in an effort to reduce greenhouse gas emission; and such management options may be transferred to other reservoir locations. For example, decreased frequency of water level fluctuation



**Figure 6** | Conceptual model of drivers of  $N_2O$  emission. Symbols (+) represent positive effects. (a) Plants have both positive and negative effect on soil nutrients through consumption of nutrients for growth and addition of nutrients resulting from decomposition of tissues. (b) High SWC is conducive to the accumulation of SOM. (c) In this work, we see a negative correlation between temperature and flux; we propose this uncommon correlation occurs because the positive effect of temperature on decomposition is masked by even stronger effects from SWC and nutrients.

can shorten the duration of moderate humidity of soil. Of course, an integrated assessment is required to include other site-specific issues (e.g., water pollution and biodiversity) before implementation. This study recorded a substantial  $N_2O$  flux which had a similar potential greenhouse effect as  $CH_4$  emissions at the same study site. We suggest the contribution of  $N_2O$  to the ‘greenhouse effect’ might be as important as that of  $CH_4$ , at least for the littoral area of a freshwater ecosystem. However, there remain high levels of uncertainty because of great spatiotemporal flux variation and environmental heterogeneity. For further works, more intense field observation and controlled experiments will help in developing a fully effective and workable management program to minimize greenhouse gas emissions from Miyun and other reservoirs.

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