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Dissolved nitrous oxide and emission relating to denitrification across the Poyang Lake aquatic continuum

Huaxin Wang[1,3,**], *Lu Zhang*[2,**], *Xiaolong Yao*[2], *Bin Xue*[2], *Weijin Yan*[1,2,*]

1. *Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China*
Email: wanghx.13b@igsnrr.ac.cn
2. *State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China*
3. *Graduate University of the Chinese Academy of Sciences, Beijing 100049, China*

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ABSTRACT

Most aquatic ecosystems contribute elevated N_2O to atmosphere due to increasing anthropogenic nitrogen loading. To further understand the spatial heterogeneity along an aquatic continuum from the upriver to wetland to lake to downriver, the study was conducted on spatial variations in N_2O emission along Poyang Lake aquatic continuum during the flood season from 15 July 2013 to 10 August 2013. The results showed the N_2O concentrations, the ratio of N_2O/dinitrogen (N_2) gases production, N_2O emission and denitrification rates ranged from 0.10 to 1.11 μg N/L, −0.007% to 0.051%, −9.73 to 127 μg N/m^2/hr and 1.33×10^4 to 31.9×10^4 μg N_2/m^2/hr, respectively, across the continuum. The average N_2O concentrations, the ratio of N_2O/N_2 and N_2O emission was significantly lower in wetlands as compared to the rivers and lake ($p < 0.01$). The significantly high denitrification rate and low N_2O emission together highlighted that most N_2O can be converted into N_2 via near complete denitrification in the Poyang Lake wetlands. Our study suggests that the wetlands might impact N_2O budget in an integrated aquatic ecosystems. Moreover, N_2O emission from different aquatic ecosystem should be considered separately when quantifying the regional budget in aquatic ecosystem.

Introduction

The Intergovernmental Panel on Climate Change (IPCC, 2007) reported that N_2O is a key greenhouse gas, potentially having an even greater impact on global warming than CO_2. The atmospheric concentrations of N_2O are increasing at approximately 0.25% per year, being responsible for approximately 5% to 10% of global warming (IPCC, 2007). Human activities have altered nitrogen cycles, causing a transfer of nitrogen inputs from terrestrial ecosystems to aquatic ecosystems leading to a cascade from underground water through rivers, lakes, estuaries and coastal seas (Hinshaw and Dahlgren, 2013). The impacts of this on N_2O emission, potentially impacting global warming, are poorly understood. Thus, it is

* *Corresponding author*. Email: yanwj@igsnrr.ac.cn (Weijin Yan).
** *These authors contributed equally to field observation, lab analysis and writing this article.*

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important to quantify FN_2O and its influence factors from aquatic systems for both global budget studies and national emission inventories.

Previous studies reported that N_2O emission was controlled by three aspects in aquatic ecosystem, including nitrogen level, denitrification rate and hydrological condition. Based on a long time series records, relationship was estimated between the NO_3^- concentration and N_2O production in the coastal upwelling area of Chile (Farias et al., 2015). Musenze et al. (2014) also found that N_2O saturation increase with the nitrogen level. In addition, the microbial denitrification might alter aquatic ecosystem as a source or sink for N_2O under different environmental conditions (Beaulieu et al., 2014; Hou et al., 2015). A study demonstrated that high degree of N_2O reduction during denitrification cause a freshwater riparian fen acting as a sink for N_2O (Blicher-Mathiesen and Hoffmann, 1999). Controls of the hydrological condition on N_2O emission has been discussed recently (Marzadri et al., 2014). The slope, water depth and water velocity had influence on the gas-exchange velocity and N_2O emission (Wang et al., 2015).

Natural wetlands, and in particularly seasonally inundated wetlands located in a river system, can play an important role in regulating N_2O emissions. IPCC (2007) reported that tropical wetlands are considered significant natural sources of N_2O. Many reports in the literature found anthropogenic nitrogen input delivered from terrestrial to wetland may increase N_2O emission in the wetland (Morse et al., 2012; Morse and Bernhardt, 2013; Moseman-Valtierra et al., 2011; Palta et al., 2013; Paludan and Blicher-Mathiesen, 1996). However, a study by Audet et al. (2014) showed that especially under NO_3^- limitation, N_2O emission was low or negative in the wetland because of consumption of N_2O during denitrification and nitrification. Wang et al. (2009) investigated N_2O emission in Taihu Lake and found that algal blooms correspond to high N_2O emission, while the macrophyte correspond to low N_2O emission.

Until now, most studies documented N_2O emission from individual freshwater ecosystem. In China, several studies have been performed on the N_2O emission from rivers (Yan et al., 2012), lakes (Wang et al., 2009; Zhong et al., 2010) and reservoirs (Liu et al., 2011). However, few studies have characterized an integrated aquatic system, spanning the upriver to wetlands to lake to downriver. N_2O and N_2 are incomplete and complete product of denitrification process, respectively. Most studies focused on the N_2O only (Allen et al., 2007; Beaulieu et al., 2010), and there is a limited understanding of the mechanism of N_2O production and emission. N_2O yield (ratio of N_2O to N_2 production) could reflect the relative rates of N_2O production and consumption. Thus, determination of dissolved N_2 and N_2O concentration in aquatic ecosystem contributes to further study of N_2O emission.

As the largest shallow lake in China, the Poyang Lake water system is one of seven aquatic systems in the Changjiang River Basin. Due to the significant fluctuation of seasonal water level in the Poyang Lake, abundant natural wetlands are formed with an area of approximately 2787 km^2 (Hu, 2010). The Poyang Lake and its wetlands are well preserved and slightly polluted. N_2O emission of this area is not well-documented (Liu and Xu, 2016; Liu et al., 2013). It is essential to understand the role of Poyang Lake and its wetlands in influencing the regional N_2O emission budget.

Here, we reported a study on dinitrogen (N_2) and N_2O gases production and emission. Using a Membrane Inlet Mass Spectrometry (MIMS) system and gas chromatography, we directly measured dissolved N_2 and N_2O concentrations ($C_{N2\ dis}$ and $C_{N2O\ dis}$, respectively) in natural wetlands. Measures also included their adjacent aquatic ecosystems along a flow gradient from upriver to natural wetlands to lake to the downriver continuum of the Poyang Lake water system. The measurements were taken during the 2013 flood season (Fig. 1). Our study had three principal objectives. First, to analyze the spatial variability and diurnal patterns of dissolved N_2 and N_2O concentrations denitrification rate and N_2O emission. Second, to determine the spatial variations in the N_2O yield and the relationship between denitrification and N_2O production in wetlands, rivers and lake, respectively. Third, to identify influence factors on the N_2O production and emission in the aquatic continuum.

1. Materials and methods

1.1. Study area

The Poyang Lake is a subtropics lake, which located in the Jiangxi Province, south of the Changjiang River of China. The mean annual air temperature and precipitation was 17.1°C and about 1500 mm, respectively (Liu et al., 2013). Most precipitation of the year occurs from April to September, especially in July and August. In general, the area of the wetlands is large during this period. Our study was conducted in Poyang Lake during the 2013 flood season. Sample collection occurred over 25 days (from 15 July 2013 to 10 August 2013). The study area spanned the upriver–wetlands–Poyang Lake–downriver aquatic continuum, with the Changjiang River Datong hydrological station serving as the administrative center (Fig. 1).

In this study, three fluvial wetlands of the Poyang Lake were selected based on their wetland-vegetation structure and the water level. Of these wetlands, one wetland is located in the national wetlands protection zone at Nanjishan, and the other two wetlands are located in the national wetlands protection zone at Wucheng (Fig. 1). The Nanjishan and Wucheng wetlands are the two largest natural wetlands in the Poyang Lake region. The Nanjishan wetland (N 28°52′21″–29°06′46″, E 116°10′24″–116°23′50″), which is South of Poyang Lake, is a typical river fluvial wetland that is fed by the middle and southern branches of the Ganjiang River and the Fuhe River. The Wucheng wetland (N 29°05′–29°15′, E 115°55′–116°03′) is located on the river-mouth delta of west branch of the Ganjiang River and the Xiushui River. Based on the wetland–vegetation composition, the selected wetlands fell into four categories (Nanjishan 1 category; Wucheng 3 categories). The first category covered a third of the Nanjishan wetland and was comprised of emergent macrophyte vegetation with *Phragmites australis* and *Triarrhera lutarioriparia L. Liu* being the dominant species. The second category was a wetland transition zone in the Wucheng wetland that accounted for one fifth of the area and was comprised of sparse emergent macrophyte vegetation with *Carex tristachya* being the dominant species. The third category was an open water zone in the Wucheng wetland consisting of small floating and submerged plant structures

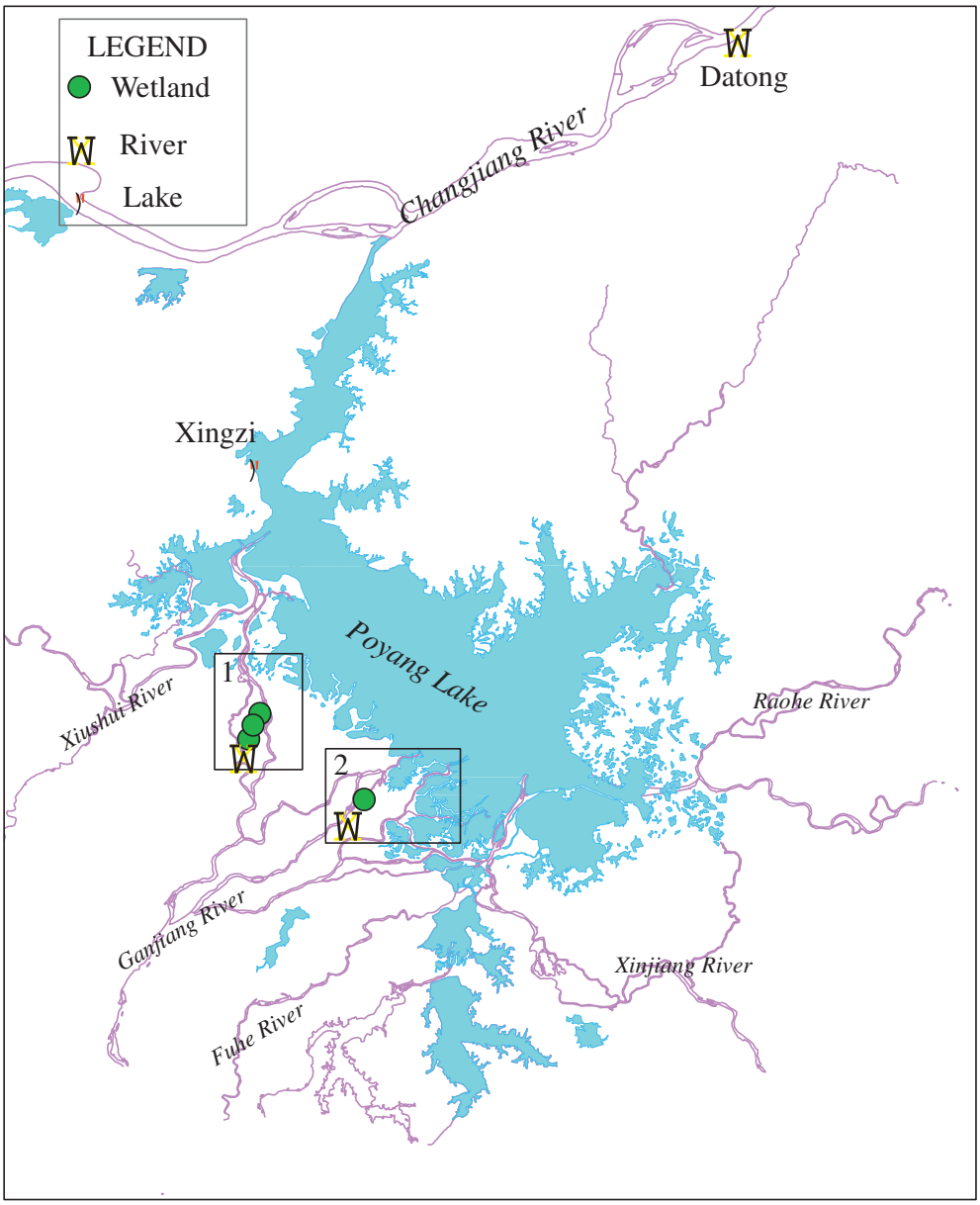


Fig. 1 – Study area across the Poyang Lake water system continuum (1 and 2 represent Wucheng wetland and Nanjishan wetland, respectively. Three circles in 1 represent WE, WI and WO in Wucheng wetland, respectively, and one circle in 2 represent WN in Nanjishan wetland).

with the dominant species of *Vallisneria natans* and *Trapa bispinosa Roxb*. The fourth category occurred in the Wucheng wetland and was dominated by dense *Carex tristachya*. The flooding depth, months of flooding, rainfall and the wind speed of each wetland is presented in Table 1.

1.2. Sampling sites design

As shown in Fig. 1, samples were collected at 25 sites spanning the entire water flow gradient continuum (upriver–wetlands–Poyang Lake–downriver) as follows: two sites are located in uprivers (UR) to the lake, where one site at the middle branch of Ganjiang River, and another one at the west branch of Ganjiang River; 9 sites were located in the Nanjishan wetland (WN); 4 sites were located in the Wucheng eulittoral wetland (WE), 3 sites are located in the Wucheng wetland in the infralittoral zone (WI), 3 of the sites were located in the Wucheng wetland with open water (WO), 4 of the sites were located in the Poyang Lake (PL); and 1 site was located at the Datong Hydrological Station (DR) on the Changjiang River. The sampling sites were distributed along both the middle branch (UR–WN–PL–DR, see Fig. 2A) and west branch (UR–WE–WI–WO–PL–DR, see Fig. 2B) of the Ganjiang River, respectively. During the non-flooding seasons, the range of water depths in the Wucheng wetland (WE, WI and WO) was greater than that in Nanjishan wetland (Fig. 2 and Table 1).

For characterizing diurnal variations of N_2O along the aquatic system gradient, 5 sites were selected in UR (the

Table 1 – Physiochemical characteristics of the studied areas across upriver-wetland- lake-downriver aquatic continuum (mean ± standard deviation).

Sampling location*	Water depth (m)	Water temperature (°C)	Wind speed (m/sec)	$NO_3^-N+NO_2^-N$ (mg N/L)	NH_4^+-N (mg N/L)	pH	Flood depth[A] (m)	Flood time	Rainfall[B] (mm)
UR	1.5–2.5	30.0–31.5 (31.0) [a]	0.4–6.0	1.24–1.49 (1.29)[a]	0.04–0.18(0.12) [a]	6.40–6.93 (6.66)	(15.4 ± 1.73)	Apr–Aug	(113 ± 108)
DR	18–22	26.0–34.8 (28.3) [b]	0.4–2.1 (1.13)	0.83–1.45 (1.09) [a]	0.10–0.30(0.18) [a]	7.11–7.6 (7.32)			
WN	0.5–1.1	29.8 ~ 33.7(31.4) [a]	0.3–6.0	0.02–0.14 (0.05) [b]	0.03–0.10(0.06) [b]	5.49–6.35 (5.92)	(15.4 ± 1.73)	Apr–Sep	(113 ± 108)
WE	1.0–1.7	28.1–31.7 (29.6) [a]	1.5–3.6	0.004–0.06 (0.02) [c]	0.03–0.07(0.04) [c]	5.69–7.21 (6.42)	(15.5 ± 0.93)	Apr–Sep	(133 ± 71)
WI	1.5–2.0	28.1–31.7 (29.5) [a]	1.75–2.6 (2.31)	0.02–0.06 (0.03) [c]	0.05–0.19(0.09) [a]	5.69–7.15 (6.23)	(15.5 ± 0.93)	Apr–Sep	(133 ± 71)
WO	1.8–3.5	29.4–30.0 (29.8) [a]	4.83–7.02 (6.19)	0.02–0.06 (0.03) [c]	0.04–0.06(0.05) [b]	6.12–7.21 (6.75)	(15.5 ± 0.93)	Apr–Sep	(133 ± 71)
PL	2.8–8.4	28.2–31.3 (29.8) [a]	0.1–3.6 (1.38)	0.23–0.93 (0.56) [d]	0.04–0.21(0.13) [a]	6.56–8.45 (7.50)	(12.1 ± 2.89)	May–Aug	(101 ± 92)

*UR: upriver, DR: downriver, WN: Nanjishan wetland, WE: Wucheng wetland with eulittoral zone, WI: Wucheng wetland with infralittoral zone, WO: Wucheng wetland with open water, and PL: the Poyang Lake. A and B are obtained from Poyang Lake Wetland Research Station. Data in parenthesis indicates average value among sampling site. Values in the same column with different superscripts within each group differ significantly ($p < 0.05$).

middle branch), WN, WI, PL and DR, respectively. For each site, samples were taken every 4 for 24 hr period for one time.

For all sampling sites, NO_3^-, NH_4^+, dissolved organic carbon (DOC), dissolved oxygen (DO), N_2O concentration, pH, water temperature and wind speed were measured.

1.3. Sample collection

The dissolved N_2O and N_2 within water column from all 25 sites (including 4 and 6 groups in the middle and west branch, respectively) were sampled and measured spanning the entire water flow gradient continuum. During each sampling session, surface (0.5 m) water samples were collected, and the water flow rate, water and air temperature, wind speed and DO were measured in situ. Samples for N_2O analysis were collected by filling 60-mL glass serum bottles from the sampler. Samples for dissolved N_2 measurements were collected using 100-mL glass digests. Samples were preserved by adding 0.5 mL of saturated $ZnCl_2$ solution to each bottle immediately to stop microbial activity before sealing. A surface water sample was collected in a 100-mL glass bottle to determine the NO_3^- and NH_4^+ concentrations. For DOC concentration measurement, an additional water sample was collected in a 100-mL glass bottle and filtered immediately through precombusted (450°C) 0.7 μm glass fiber filters (Whatman, GF/F) to determine DOC concentrations. Samples were transported to the lab on ice for further analysis. Water temperature and DO were measured using a portable meter (YSI 550A) pre-calibrated for DO using an iodometric method in the lab. All samples were collected in triplicates.

1.4. Analytical methods

N_2O were measured using a Gas Chromatography equipped with an Electron Capture Detector (GC-ECD) on a Shimadzu GC-2014 gas chromatograph (Japan) according to the headspace-equilibrium method (Huttunen et al., 2002). Specifically, 10-mL of highly purified N_2 (purity >99.999%) was injected into the sampling bottle using an airtight syringe, displacing a 10-mL water sample. The bottles were then shaken vigorously for 10 min and equilibrated for 4 hr. A 5-mL air sample was then taken for N_2O analysis, and the gas chromatograph was calibrated using a standard air sample. DOC concentration was measured using a total organic carbon analyzer (TOC-VCPH, Shimadzu, Japan). For both NO_3^- and NH_4^+ concentrations water samples were filtered through 0.45-μm membranes and then subjected to analysis in a Flow Injection Analyzer (FIA-3100, Titan, China).

Dissolved N_2 in river water were determined using a Membrane Inlet Mass Spectrometry (MIMS) system (HPR40, Hiden Analytical, UK). Dissolved N_2 was measured by the N_2:Ar method. For details on the analysis and calculation of dissolved N_2 concentrations, see the method described by Yan et al. (2004).

1.5. Calculation

1.5.1. N_2O and N_2 emission

The N_2 and N_2O production (μg N/L) within water columns are defined as the difference between the measured concentrations ($C_{N2Omeas}$ (μg N/L), C_{N2meas} (mg N_2/L)), and the atmospheric equilibrium concentrations of river water (C_{N2Oequ} (μg N/L), C_{N2equ} (mg N_2/L)):

$$N_2O\,\text{production} = C_{N2Omeas} - C_{N2Oequ} \quad (1)$$

$$N_2\,\text{production} = C_{N2meas} - C_{N2equ}. \quad (2)$$

N_2O and N_2 emissions (or flux) between the water surface and atmosphere are calculated based on the gas transfer velocity and gas net production (Garnier et al., 2006; Raymond and Cole, 2001):

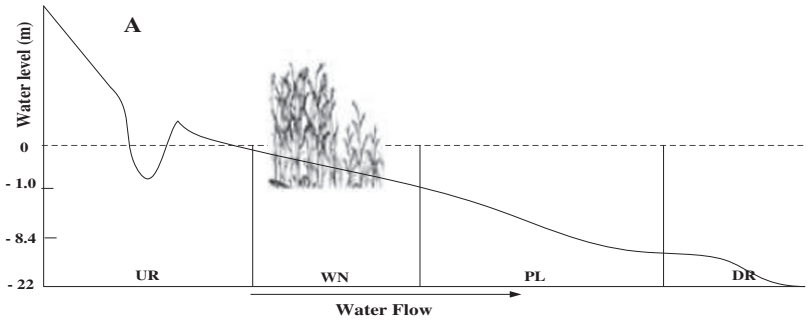
$$N_2O\,\text{emission} = N_2O\,\text{production} \times k_{gas} \quad (3)$$

$$N_2\,\text{emission} = N_2\,\text{production} \times k_{gas} \quad (4)$$

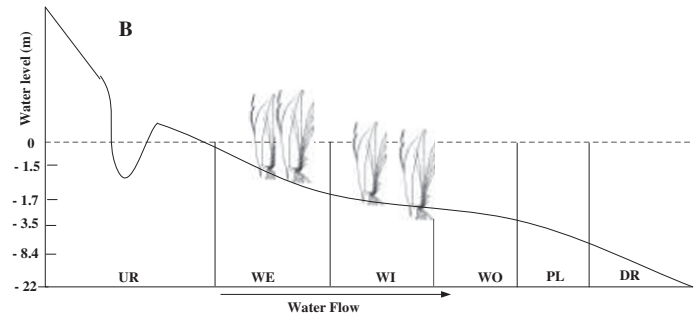
where, N_2O emission (μg N/m^2/hr) and N_2 emission (μg N_2/m^2/hr) are the interfacial flux of N_2O and N_2, respectively and k_{gas} (cm/hr) is the gas transfer velocity, calculated by Eq. (5):

$$k_{gas} = k_{600} \times (Sc_{gas}/600)^{-n} \quad (5)$$

where, Sc_{gas} is the Schmidt number for N_2O or N_2 and is calculated according to Wanninkhof (1992) at the *in situ*



	UR	WN	PL	DR
N_2O production (μg N/L)	0.84 ± 0.07[a]	-0.02 ± 0.05[b]	0.22 ± 0.09[c]	0.20 ± 0.04[c]
N_2O emission (μg N/m^2/hr)	84.9 ± 38.9[a]	-1.55 ± 3.90[b]	23.0 ± 9.70[c]	14.4 ± 7.79[d]
Denitrification (1×10^5 μg N_2/m^2/hr)	1.98 ± 0.19[a]	2.06 ± 0.59[a]	1.44 ± 0.09[b]	0.13 ± 0.05[c]
N_2O yield (%)	0.045 ± 0.013[a]	-0.001 ± 0.003[b]	0.021 ± 0.004[c]	0.011 ± 0.06[d]



	UR	WE	WI	WO	PL	DR
N_2O production (μg N/L)	0.79[a]	0.02 ± 0.05[b]	0.10 ± 0.05[c]	0.14 ± 0.07[d]	0.22 ± 0.09[e]	0.20 ± 0.04[f]
N_2O emission (μg N/m^2/hr)	127[a]	1.80 ± 8.50[b]	8.93 ± 3.72[c]	15.2 ± 8.68[d]	23.0 ± 9.70[e]	14.4 ± 7.79[d]
Denitrification rate (1×10^5 μg N_2/m^2/hr)	1.52[a]	2.06 ± 0.06[b]	1.52 ± 0.27[a]	1.41 ± 0.07[a]	1.44 ± 0.09[a]	0.13 ± 0.05[c]
N_2O yield (%)	0.049[a]	0.001 ± 0.002[b]	0.006 ± 0.003[c]	0.010 ± 0.002[d]	0.021 ± 0.004[e]	0.11 ± 0.06[f]

Fig. 2 – The profile of sampling sites along upriver (UR, middle tributary of Ganjiang River) — Nanjishan wetland (WN) — lake (PL) — downriver (DR) (A), and the profile of the sampling sites along upriver (UR, west tributary of Ganjiang River) — Wucheng wetland (WE,WI and WO) — lake (PL) — downriver (DR) (B). Dished lines represent the average water depth. Tables showed the spatial variation of N_2O production, N_2O emission, denitrification rate and N_2O production/N_2 production across the sampling sites. Values in the same row with different superscripts within each group differ significantly ($p < 0.05$) in each table.

temperature, n is the Schmidt number coefficient (2/3 for smooth surfaces, decreasing to 1/2 in the presence of breaking waves), k_{600} is gas (N_2O or N_2) transfer velocity at the Schmidt number of 600 in fresh water, and k_{600} can be calculated by Eq. (6) accounting for both wind speed and water flow rate (Borges et al., 2004):

$$k_{600} = 1.0 + 1.719(w/h)^{0.5} + 2.58\mu_{10} \quad (6)$$

where, w (m/sec) is the water flow rate, h (m) is the depth of the river water column and μ_{10} (m/sec) is the wind speed at a 10-m height.

The N_2O saturation (concentration relative to that expected under water-atmosphere equilibrium) was calculated by:

$$N_2O\,\text{saturation} = \left(C_{N2Omeas}/C_{N2Oequ}\right) \times 100\% \quad (7)$$

where $C_{N2Omeas}$ (μg N/L) is the measured N_2O concentration and C_{N2Oequ} (μg N/L) is the saturation concentration of atmospheric N_2O at the given water temperature (Weiss and Price, 1980).

1.6. Statistical analysis

The data was analyzed using the SPSS 16.0 software package. One-way analysis of variance (ANOVA) combined with the independent-samples *t*-test was used to test for statistically significant difference between group mean values. Pearson correlation analyses were performed to test the relationships between N_2O emission and environmental factors. Stepwise regression analyze was also used to quantify the relationship between N_2O emission and influence factors. The statistical significance was determined at the 95% confidence level.

2. Results

2.1. Spatial variations of water chemistry across the Poyang Lake wetlands continuum

Water chemistry characteristics across the continuum are provided in Table 1 and Fig. 3. The water temperature and wind speed varied from 26.0 to 34.8°C and 0.1 to 6.0 m/sec, respectively. NO_3^- and NO_2^-, NH_4^+ concentrations ranged from 0.004 to 1.49 mg N/L and 0.03 to 0.30 mg N/L, respectively. NO_x^- concentration was significantly higher in rivers and lake than that in wetlands (ANOVA, $p < 0.01$). The mean water depth was 0.8, 1.4, 1.8 and 2.7 m in WN, WE, WI and WO, respectively. The pH and DO concentration ranged from 5.49 to 8.45 and 0.70 to 8.52 mg/L, respectively. Dissolved inorganic nitrogen (DIN) ($NO_2^- + NO_3^- + NH_4^+$) ranged from 0.04 to 1.55 mg N/L. DIN concentration was significantly higher in rivers and lake than that in wetlands (ANOVA, $p < 0.01$). The DOC concentrations averaged (3.02 ± 1.64) mg/L, and it varied greatly among sampling sites, ranging from 1.18 to 7.43 mg/L. DOC concentration was higher in wetlands than that in the rivers and lake (ANOVA, $p < 0.01$).

2.2. Spatial and diurnal variations of N_2O and N_2 across the Poyang Lake wetlands continuum

The spatial patterns of N_2O concentration, N_2O production and N_2O emission were extremely variable (Fig. 2). They decreased rapidly from UR to WN and then showed an increasing trend from WN to PL, but the N_2O in DR was slightly lower than those in PL. The N_2O concentration, N_2O production and N_2O emission ranged from 0.10 to 1.11 μg N/L, −0.10 to 0.93 μg N/L and −9.73 to 127 μg N/m/hr, respectively. The associated mean values for the three forms of N were 0.39 ± 0.25 μg N/L, 0.20 ± 0.25 μg N/L and 17.5 ± 30.3 μg N/m^2/hr, respectively. N_2O saturation ranged from 50.4% to 597%. All sampling sites were over saturated with the exception of WN (87% ± 25%). Together, both N_2O production and N_2O emission were significantly lower in WN than in any of the other sites (ANOVA, $p < 0.01$ and see Fig. 2A). N_2O emission was almost below the level of detection in WN. For wetlands, N_2O emission was just slightly greater in WE and WI than in WN (Fig. 2). In addition, for rivers and lake, both N_2O production and N_2O emission were significantly lower in PL and DR than those in UR (ANOVA, $p < 0.01$). Across the Wucheng wetland flow gradient (Fig. 2B), N_2O production was lower in wetlands (WE, WI and WO) than in the other 3 zones (UR, PL, DR) of the flow gradient (ANOVA, $p < 0.05$). N_2O emission was relatively high in WO, we attribute this to high wind speed that may facilitate N_2O emission.

N_2O concentration, N_2O production and N_2O emission had no diurnal variability during the 24 hr sampling period at the

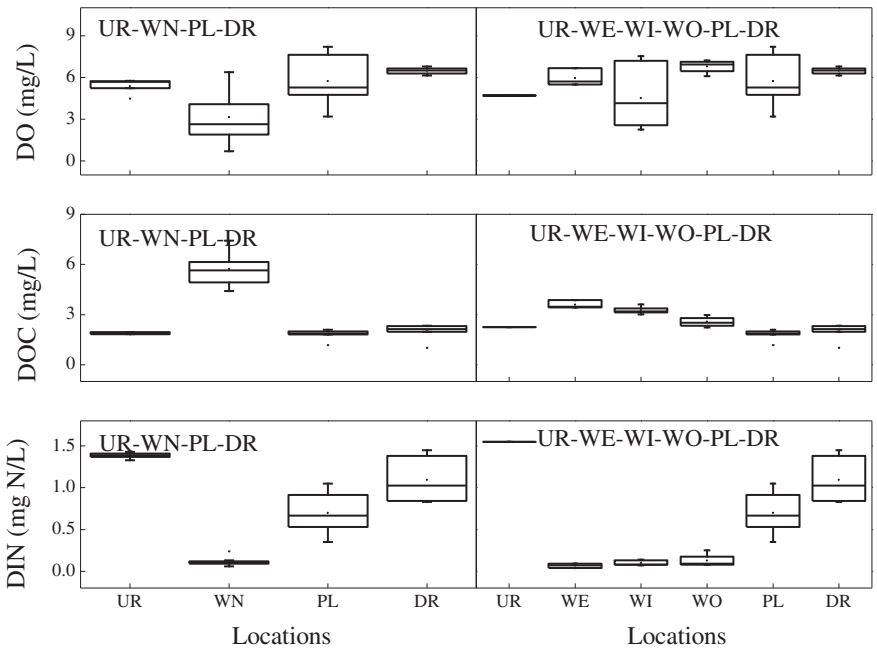


Fig. 3 – Spatial variation of physiochemical characteristics (dissolved oxygen (DO), dissolved organic carbon (DOC) and dissolved inorganic nitrogen (DIN)) across the Poyang Lake aquatic continuum.

5 sites (ANOVA, $p > 0.01$). Moreover, the diurnal variation of water physiochemical characters (*i.e.*, DO, water temperature, pH and wind speed) could not explain the diurnal variation of N_2O concentration, N_2O production and N_2O emission.

Spatial variabilities in N_2 production and denirtification rate were examined between wetlands and rivers and lake across the continuum (ANOVA, $p < 0.01$) (Fig. 4). Spatial changes were also observed among the wetlands (ANOVA, $p < 0.01$). N_2 production and denitrification rate ranged from1.19 to 2.76 mg N_2/L and 1.33×10^4 to 31.9×10^4 μg N_2/m^2/hr, respectively. The corresponding average values for the two measures were (1.34 ± 0.77) mg N_2/L and $(17.4 \times 10^4 \pm 45.6 \times 10^4$ μg N_2/m^2/hr, respectively, across the continuum. The denitrification rate was highest in WN, while was lowest in DR. Denitrification rate decreased along the water flow from the eulittoral zone to the open water zone in the Wucheng wetland. N_2O emission was negatively correlated with denitrification rate in wetlands ($p < 0.05$), while was positively correlated with denitrification rate in rivers and lake ($p < 0.05$) (Fig. 5).

The N_2O yield varied spatially across the Poyang Lake wetlands system (Fig. 2). A relatively high N_2O yield was measured in UR and averaged 0.045% ± 0.006%. In WN, most N_2O yield values were less than zero and averaged −0.001% ± 0.003%. In the Wucheng wetland (WE, WI and WO), the N_2O yield values ranged from −0.002% to 0.014%, with a relatively low mean value of 0.006% ± 0.005%. Furthermore, the N_2O yield increased along the water flow path gradient from the eulittoral zone to the open water zone in the Wucheng wetland (Fig. 4).

2.3. N_2O production and N_2O emission and the influence factors

Table 2 shows the Pearson correlation between N_2O concentration, N_2O production, N_2O emission and environmental factors.

NH_4^+ did not correlate with N_2O concentration and N_2O production for river, lake and wetlands, respectively ($p > 0.01$). The correlation coefficient was high between N_2O concentration and N_2O production and NO_3^- ($p < 0.01$) in rivers and lake, and was high with DOC concentrations ($p < 0.01$) in wetlands. N_2O emission was only positively correlated with NO_3^- concentrations ($p < 0.01$) and wind speed ($p < 0.05$) in rivers and lake. Besides, our results showed N_2O was correlated with increased pH in the wetlands, but with decreased pH in the rivers and lake. The N_2O seems to be more related to the DO in the wetlands.

Stepwise regression analyses were used to test the relationship between N_2O concentration, N_2O production and N_2O emission and environmental factors (Table 3). NO_3^- concentration accounted for 80% and 73% of the variance in N_2O production for the continuum, rivers and lake, respectively. pH, water temperature in combination with DO accounted for 62% of the variance in N_2O production for wetlands. N_2O concentration and wind speed accounted for 88% and 97% of the variance in N_2O emission for the continuum, rivers and lake, respectively. N_2O production accounted for 88% of variance in N_2O emission in wetlands.

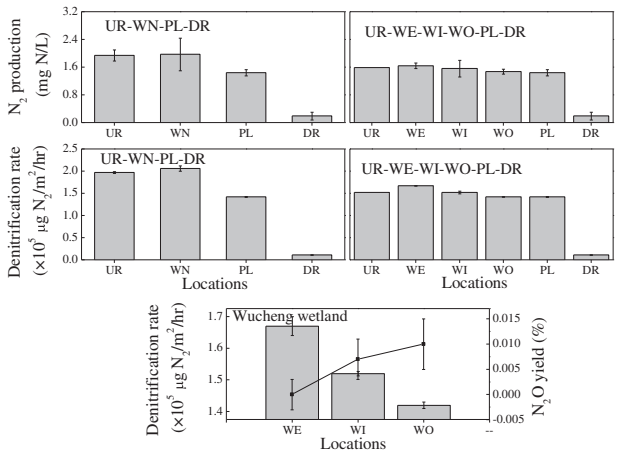


Fig. 4 – Spatial variation of N_2 production, denitrification rate and the ratio of N_2O production/N_2 production across the Poyang Lake aquatic continuum.

3. Discussions

3.1. N_2O production, N_2O emission and its influence factors across the Poyang Lake wetlands continuum

The findings of this study suggest that along the wetlands water flow gradient, the N_2O production and N_2O emission decreased from uprivers to wetlands, and that the concentrations were relatively lower in the wetlands than those in the rivers and lake. Rivers and lake were strong sources of atmospheric N_2O, while wetlands were the sink of that.

N_2O is created in the process of nitrification and denitrification. The NH_4^+ concentration was low (averaged: 0.11 ± 0.30 mg N/L), and the relationship between and N_2O was weak. As a result, nitrification process wasn't dominating the turnover of nitrogen and N_2O production. It was reported that the oxic environments (Wang et al., 2009) and elevated NH_4^+ supply (DeSimone et al., 2010) may lead to nitrification resulting in the N_2O production.

NO_3^- and DOC concentration can regulate denitrification rate and control the production of N_2O and N_2. Previous studies demonstrated that the coupling process of N and C could affect the N cycle in wetlands (Garnier et al., 2010; Wang et al., 2014). In general, incomplete denitrification occurs with limitation of carbon substrate (Israel et al., 2009). N_2O was generated as the main product in rivers and lake where NO_3^- concentration was high. The wetlands received large organic carbon inputs from macrophytes, resulting in the complete denitrification, and N_2 is the main product. In addition, NO_3^- concentrations were unusually low, and in some water dissolved N_2O concentrations were below the ambient level especially in WN. This indicates that denitrifying consumption of N_2O may have occurred. Low N_2O concentration, N_2O production and N_2O emission but high denitrification rate in the wetlands of Poyang Lake could contribute to the high DOC concentrations. As electron donors of denitrifiers, organic carbon provided energy. Wetland potentially provides optimum conditions for complete denitrification (*i.e.*, available carbon) and subsequently low N_2O emissions.

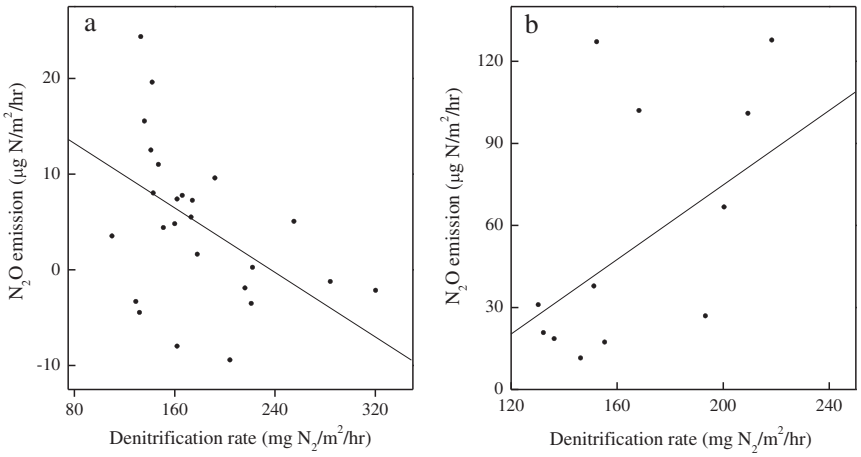


Fig. 5 – Relationship between N_2O emission and denitrification rate in wetlands (a), rivers and lake (b).

Based on our study, NO_3^- and DOC were the most significant variables contributing to N_2O production and emission. However, other factors such as the DO may also have important influences on N_2O production. As an intermediate product of denitrification, N_2O could be produced and reduced simultaneously under oxygen-limited conditions (Minamikawa et al., 2015). Several publications demonstrated a negative relationship between DO and N_2O production (Jacobs and Harrison, 2014; Wang et al., 2015). Similarly, we found a weakly negative relationship between DO and N_2O production in rivers and lake. Contrasting this relationship in rivers and lake, this study found a positive relationship between DO and N_2O production in wetlands. DO was particularly low especially in WN and this was most likely related to the decomposition of abundant DOC (Zarnetske et al., 2011). In conditions where DO concentrations are extremely low, such as in WN (DO range: 1–4 mg/L) the product of denitrification was N_2 rather than N_2O. These low DO findings are in agreement with the published findings of other laboratories, and which most likely reflect, first, that the highest rates of N_2O production are observed between oxic and anoxic states, and second that the oxygen supply can increase N_2O emission (Burgin and Groffman, 2012; Hernandez and Mitsch, 2006). Therefore, DO has a dual role in regulating N_2O emission, since it could promote or inhibit N_2O emission.

A substantial body of research has documented that river is a source of N_2O with increased nitrogen loading (Venkiteswaran et al., 2014). In our study, N_2O emission was positively correlated with NO_3^- and N_2O production and demonstrated the impact of stream nitrogen loading on N_2O emission. In addition, wind speed was positively correlated with N_2O emission in rivers and lake ($p < 0.01$). Wind speed influenced gas velocity of the air–water interface, and thereby modulated N_2O emission (Musenze et al., 2015). But there was no correlation between wind speed and N_2O emission in wetlands. This could be explained by N_2O concentration in wetlands water column of wetlands being so low that the impact of wind speed on N_2O emission was weak.

Table 2 – Pearson correlation coefficient between N_2O and environmental variables.

	NO_3^-	NH_4^+	DOC	pH	DO	WS	WT	N_2O conc.	N_2O pro.
All									
N_2O conc.	0.93^{**}		-0.64^{**}	0.22^{ns}	0.21^{ns}		-0.07^{ns}		
N_2O pro.	0.93^{**}		-0.63^{**}	0.21^{ns}	0.21^{ns}		-0.05^{ns}		
N_2O emission	0.82^{**}		-0.51^{**}	0.09^{ns}	0.15^{ns}	0.35^{*}	-0.08^{ns}	0.88^{**}	0.88^{**}
Wetlands									
N_2O conc.	-0.31^{ns}	0.02^{ns}	-0.68^{**}	0.54^{**}	0.48^{*}		-0.47^{*}		
N_2O pro.	-0.33^{ns}	0.02^{ns}	-0.68^{**}	0.56^{**}	0.52^{**}		-0.40^{*}		
N_2O emission	-0.30^{ns}	-0.01^{ns}	-0.64^{**}	0.63^{**}	0.52^{**}		-0.39^{*}	0.94^{**}	0.94^{**}
Rivers and lake									
N_2O conc.	0.92^{**}	-0.17^{ns}	0.33^{ns}	-0.70^{**}	-0.30^{ns}		0.49^{*}		
N_2O pro.	0.92^{**}	-0.17^{ns}	0.32^{ns}	-0.71^{**}	-0.31^{ns}		0.51^{*}		
N_2O emission	0.81^{**}	-0.13^{ns}	0.41^{ns}	-0.59^{**}	-0.22^{ns}	0.74^{**}	0.21^{ns}	0.84^{**}	0.84^{**}

N_2O conc.: N_2O concentration, N_2O pro.: N_2O production, WS: wind speed, WT: water temperature, ns: not significant.

* Significance of $p < 0.05$.

** Significance of $p < 0.01$.

Table 3 – Regression analysis between N_2O and environmental variables.

Best regression formula	R^2	p	F	n
All				
N_2O conc. = 0.51 NO_3^- + 0.21	0.80	0.000	121	45
N_2O conc. = −0.10 DOC + 0.71	0.41	0.000	20	32
N_2O emission = 0.51 NO_3^- + 0.024	0.80	0.000	122	45
N_2O pro. = −0.10 DOC + 0.52	0.40	0.000	20	32
N_2O emission = 107 N_2O conc. − 21.43	0.86	0.000	188	45
N_2O emission = 107 N_2O conc. + 3.21 WS − 28.6	0.88	0.000	109	45
N_2O emission = −9.09 DOC + 49.3	0.26	0.003	10	32
Wetlands				
N_2O conc. = −0.04 DOC + 0.41	0.47	0.000	18	23
N_2O conc. = 0.16 pH − 0.41 WT + 0.02 DO + 1.28	0.64	0.000	13	27
N_2O conc. = −0.04 WT + 0.03 DO + 1.40	0.64	0.000	20	27
N_2O pro. = −0.04 DOC + 0.21	0.46	0.000	18	23
N_2O pro. = 0.02 pH − 0.04 WT + 0.02 DO + 0.92	0.62	0.000	12	27
N_2O pro. = −0.04 WT + 0.03 DO + 1.05	0.61	0.000	18	27
N_2O emission = 87.0 N_2O pro. + 1.09	0.88	0.000	158	27
N_2O emission = −3.40 DOC + 19.5	0.41	0.001	14	23
N_2O emission = 10.7 pH − 2.43 WT + 12.4	0.56	0.000	15	27
Rivers and lake				
N_2O conc. = 0.80 NO_3^- + 0.09	0.74	0.003	20	18
N_2O conc. = −0.26 pH − 2.50	0.49	0.001	15	18
N_2O pro. = 0.80 NO_3^- − 0.28	0.73	0.003	19	18
N_2O pro. = −0.27 pH + 2.34	0.50	0.001	15	18
N_2O emission = 135 N_2O conc. − 44	0.79	0.001	27	18
N_2O emission = 105 N_2O conc. + 17.0 WS − 58	0.97	0.000	106	18
N_2O emission = 120 NO_3^- − 68	0.66	0.000	31	18
N_2O emission = −32 pH + 273	0.35	0.009	8.7	18

N_2O conc.: N_2O concentration, N_2O pro.: N_2O production, WS: wind speed, WT: water temperature.

3.2. The relationship between denitrification and N_2O emission across the Poyang Lake aquatic continuum

N_2O emission was positively related to the denitrification rate in rivers and lake, while N_2O emission was negatively related to the denitrification rate in wetlands. The products of denitrification included N_2 and N_2O, where N_2 was the end product of denitrification and N_2O was the intermediate product (Beaulieu et al., 2014; Chen et al., 2014b). The composition of denitrification products, described as the ratio of N_2O production/N_2 production, can reflect (nitrate reduction degree) the nitrogen removal and N_2O emission (low ratio means complete nitrate reduction, while high ratio means incomplete reduction). And this ratio is defined as the N_2O yield (Beaulieu et al., 2011; Silvennoinen et al., 2008). A low N_2O yield existed with a relatively high denitrification rate in the wetlands. Compared with N_2O yield of 53 streams across the USA, which range from 0.05% to 5.6% (Beaulieu et al., 2011), the N_2O yield in wetlands (mean value: 0.003% ±0.005%) of Poyang Lake was extremely low. Abundant DOC in wetlands can provide energy and anaerobic condition for denitrification processes that further stimulated N_2O conversion into N_2 and that resulted in low N_2O yield in wetlands. Previous studies have documented that under some conditions, the reduction of N_2O to N_2 is favored during denitrification in wetland (Audet et al., 2014; Knowles, 1982; Koegel-Knabner et al., 2010).

A negative value for N_2O emission in WN, suggested that WN was a sink for N_2O. Furthermore, the relatively low N_2O emission in the Wucheng wetland implied that there existed low production and emission of N_2O. Low N_2O emission and high denitrification rate suggested that wetlands could remove DIN and reduce N_2O emission *via* complete denitrification.

3.3. N_2O emission in other aquatic ecosystems in the literature

Compared to reports about other aquatic ecosystems (Table 4), Yan et al. (2012) reported N_2O emission (1.87–40.8 μg N/m^2/hr) in lower reach of the Changjiang River, and this was much higher than that in wetlands of our study. With relatively low nitrate concentrations, the N_2O emission level in the uprivers of this study were far away lower than that in the South Platte river (McMahon and Dennehy, 1999), where enriched DIN (0.25–22 mg N/L) was derived from waste water and agricultural irrigation water. Both NO_3^- concentrations and N_2O production in this study were lower than those documented by Chen et al. (2014a), but N_2O emission was relatively high. These disparities between our findings and those of Chen et al. may be explained by the high wind speed (average: 2.62 m/sec, 1.61 m/sec, Chen et al., 2014a) in this study. N_2O emission were much lower in the wetlands of the Poyang Lake than that of a wetland in Denmark, which is located in an agricultural area and enriched in NO_3^- (Paludan and Blicher-Mathiesen, 1996).

4. Conclusions

Significantly spatial variations of N_2O concentration, N_2O production and N_2O emission were observed across the Poyang Lake water flow gradient. Relatively low N_2O concentration (0.10–0.40 μg N/L) and N_2O emission (−9.37–24.4 μg N/m^2/hr) was observed in wetlands. In addition, our study also demonstrated N_2O concentration, N_2O production and N_2O emission increased along the gradient from the eulittoral zone to the open water zone in the Wucheng wetland. The diurnal study suggested no significant differences in N_2O concentration and N_2O emission over the consecutive 24 hr periods. Diurnal fluctuations in N_2O in different aquatic ecosystem require further study.

Factors governing the variability of N_2O concentration, N_2O production and N_2O emission were identified. In the rivers and lake, the N_2O concentration, N_2O production and N_2O emission levels were correlated with the NO_3^- concentration. But in wetlands, they were correlated with the DOC and DO concentration. Stepwise regression analyses demonstrated the NO_3^- explained 73% of the N_2O production variance in rivers and lake. In wetlands, N_2O production variance could be explained by pH, DO and water temperature. N_2O concentration and wind speed together could explain 97% of the N_2O emission variance in rives and lake. In wetlands, N_2O emission could be predicted by N_2O production.

A high denitrification rate and low or negative value of N_2O emission in the Poyang Lake wetlands suggested that the well preserved wetlands may be the sink for N_2O in the Poyang Lake or may reduce the transfer of N_2O emission to the atmosphere due to the near complete denitrification. We suggested that

Table 4 – N_2O emission in rivers and wetlands in the literature.

	Name	Observed date	N_2O emission ($\mu g\ N/m^2/hr$)	Method	Reference
River	JiuLong Jiang (China)	2010–2011	0.67–9.94	Modeled K	(Chen et al., 2014a)
River	South platte river (USA)	Fall, winter and summer from 1994 to 1995	3.75–1358	Chamber	(McMahon and Dennehy, 1999)
River	The Changjiang River (China)	August 2012	1.87–40.8	Modeled K	(Yan et al., 2012)
River	Spring-fed LII river (New Zealand)		52–140/13–52	Chamber/modeled K	(Clough et al., 2007)
Wetland	Lamprey River	From January 2005 to October 2006	0.6	Chamber	(Flint and McDowell, 2015)
Wetland	Prairie pothole wetland (Canadia)	2004–2005	2.08–11.7	Chamber	(Badiou et al., 2011)
Wetland	Wetlands (Denmark)	From July 1993 to June 1994	5.88–263	Chamber	(Paludan and Blicher-Mathiesen, 1996)
Wetland	Coastal plain wetland (USA)	From July 2007 to June 2009	−0.01-1.59	Chamber	(Morse et al., 2012)

when quantifying the regional N_2O emission budget in an integrated aquatic ecosystem, N_2O emission from river, lake and wetlands should be considered separately.

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