



Diurnal pattern in nitrous oxide emissions from a sewage-enriched river



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HIGHLIGHTS

- ▶ We observed the rapid degassing of N₂O in a typical sewage-enriched river.
- ▶ Factors controlling the N₂O flux were pH, DO, NH₄⁺, SO₄²⁻, and temperature.
- ▶ Sampling at 19:00 h well represent the daily average N₂O flux at the studied river.

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ABSTRACT

Estimates of N₂O emission based on limit measurements could be highly inaccurate because of considerable diurnal variations in N₂O flux due to rapid transformation of nutrients and diel change of dissolved oxygen (DO). In the present study, the N₂O fluxes, dissolved N₂O concentrations, and the controlling variables were measured hourly for 3 d and night cycles at five sites on a typically sewage-enriched river in the Taihu region. There were no significant diurnal patterns in N₂O emissions and dissolved N₂O saturation, with respective mean value of 56.1 μg N₂O–N m⁻² h⁻¹ (range = 41.1 μg N₂O–N m⁻² h⁻¹ to 87.7 μg N₂O–N m⁻² h⁻¹) and 813% (range = 597–1372%), though distinct diurnal patterns were observed in DO concentration and river chemistry. However, the mean N₂O emissions and the mean dissolved N₂O saturation during the day (61.7 μg N m⁻² h⁻¹ for N₂O fluxes and 0.52 μg N L⁻¹ for dissolved N₂O saturation) were significantly higher than those during the night (50.1 μg N m⁻² h⁻¹ for N₂O fluxes and 0.44 μg N L⁻¹ for dissolved N₂O saturation). Factors controlling the N₂O flux were pH, DO, NH₄⁺, SO₄²⁻, air temperature, and water temperature. Sampling at 19:00 h could well represent the daily average N₂O flux at the studied river.

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1. Introduction

Nitrous oxide (N₂O) plays an important role in stratospheric ozone destruction and climate change (Crutzen, 1981). Rivers and streams constitute a primary pathway for anthropogenic nitrogen (N) transport from land to sea and are potentially important N₂O sources (Seitzinger and Kroeze, 1998). River networks produce at least 0.68 Tg y⁻¹ of anthropogenic N₂O–N, which is equivalent to 10% of the global anthropogenic N₂O emission (Beaulieu et al., 2011). However, the fraction of N₂O emitted relative to the reactive N inputs to rivers is poorly defined, because factors that regulate N₂O production in streams are not fully understood and in situ data are sparse (Mulholland et al., 2008).

Aquatic emitted N₂O is intermittent product of nitrification in oxic environments and byproduct of denitrification in anoxic

environments. Nitrification and denitrification are regulated by substrate supply (Robertson, 1987). Both of these microbially driven processes are affected by river chemistry, such as pH, dissolved oxygen (DO), oxidation–reduction potential, dissolved organic carbon (DOC), chloride (Cl⁻), and sulfate (SO₄²⁻) (Tsuneda et al., 2005). Studies have shown that rivers and streams can rapidly transform river chemicals over a diurnal scale, this may result in diurnal variations in N₂O emissions. Harrison et al. (2005) and Clough et al. (2007) showed that complete reduction and oxidation sequences were happened within only a few hours, which resulted in a diurnal cycle of N₂O emission in streams and rivers. In the Taihu region, N₂O emission rates from surface water varied substantially (Xiong et al., 2006; Wang et al., 2010), however, the patterns were less studied and understood, especially on a diurnal scale.

The diurnal variations in N₂O emissions from rivers and streams are often complex. Significant diurnal pattern of N₂O fluxes are found broadly in streams and rivers (Harrison et al., 2005; Clough

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et al., 2007). The most marked variation was observed in a eutrophic stream with rapid diel change in redox–oxide sequence (Harrison et al., 2005). In another short-term field observation of dissolved N_2O in Lake Taihu, Wang et al. (2010) found that N_2O production was regulated by rapid changes in DO and organic matter at the sediment–water interface. However, Baulch et al. (2012) showed no consistent patterns of N_2O fluxes in streams with several N sources and a more restricted O_2 conditions, such as the streams in Toronto, Canada.

The river network in the Taihu region (30–32°N, 118–122°E) in Eastern China is extensive, and occupies 15% of the total watershed surface area. The total length of the river network is 120 000 km and the river density is 3.24 km km⁻² (Gong and Lin, 2009). The rivers in the region are narrow (5–30 m) and short (0.5–80 km), with flow speed ranging from 0.01 m s⁻¹ to 0.3 m s⁻¹. Taihu region accounts for only 0.4% of the total area of China and 2.9% of the population, but it provides more than 14% of China's gross domestic product (GDP) (Qin et al., 2007). With rapid economic growth, large amounts of N-enriched sewage input into the rivers which may have become N_2O sources. In 2008, the total quantity of sewage reached 2710 million tons, with more than 40% of urban sewage and 80% of rural sewage left untreated and directly discharged into the river water (Zang et al., 2009). Sewage water is characterized by low DO, with saturation as low as 13.0%, and high ion concentrations (e.g., NH_4^+ , NO_3^- , Cl^- , SO_4^{2-} , with their respective concentrations as high as 1.16, 6.73, 173.3, and 184.0 mg L⁻¹) (Wang et al., 2009). Nevertheless, diurnal variations in these parameters and their effects on N_2O emissions have not been studied. Wang et al. (2010) investigated the effects of oxygen and other chemicals on N_2O saturation in Taihu and found that N_2O underwent complete reduction within a few hours. River differs from lake in chemistry because of the direct inputs of fertilizer, sewage, manure, and wastewater, which leads to higher concentrations in the river than in the lake. Therefore, this study aimed to determine: (1) the presence and extent of diurnal variations in river water chemistry and N_2O emissions in a sewage-enriched river in the Taihu region; (2) the potential factors that influence diurnal N_2O variability; and (3) the implications for sampling programs to capture the average daily N_2O fluxes.

2. Materials and methods

2.1. Study site

The present study was carried out along the Xin'an Tang river in Changshu County of the Taihu region. The site has a mean annual temperature of 15.6 °C, and a mean annual precipitation of 1043 mm. The river drains a 39 km² suburban agricultural area and flows in a south–north direction for 14.3 km, before it drains into the Changjiang river. The river is mixed sand and mud-bottomed, and is turbid with poor light penetration. The surface sediment is black, which is indicative of iron sulfides. The growth of benthic algal is limited and no algal blooms occurred during our sampling period. Planting and riparian vegetation flourishes around the riverbank. The major sources of water pollution in this river are untreated urban and rural sewage, as well as animal excreta directly discharged into the water bodies (Xing et al., 2001; Xie et al., 2007). Therefore, the surface water is highly eutrophic, i.e., rich in minerals and organic nutrients that promote the proliferation of plant life.

The sampling site (32°33'N, 118°42'E) is 8 km downstream of the headstream. The average water depth at the site is 2.5 m, and the river width is 14 m. The average water velocity of the Xin'an Tang river during our observation period was 0.1 m s⁻¹ and the discharge rate was 3.5 m³ s⁻¹.

2.2. Sample collection

Water surface N_2O fluxes were measured using the closed chamber method on five sites across the river plane every 2 h from 15:00 h on 3 October to 15:00 h on 6 October 2010. Five closed chambers made from round polypropylene “cake” containers (No. 201120210689.2, ISSAS, China) shaped as columnar frustra (0.0064 m³ volume, 0.07 m² surface area of the opening) were aligned and floated using attached Styrofoam annuli. The chambers were suspended from a simply constructed bridge, and projected only 1–2 cm into the water column to minimize water surface disruption. The chamber lids were covered with silver paper (SD, Moaihua, NJ, China) to prevent heating inside the chamber. The headspace temperature was monitored in the chamber to verify that heating did not exceed ambient temperatures. A 22 mL gas sample for each closed chamber was collected every 10 min during a 40-min period (total of five gas samples per chamber) using a syringe from a port located at the top center of the chamber. Testing of the closed chambers showed that the N_2O concentration increased linearly for up to 40 min sampling times in the study river. The samples were stored in glass vials (Venoject; No. 1365, Terumo Corp., Tokyo, Japan) pre-evacuated and sealed with silicon rubber stoppers (Surflo, No. S346, Terums Corp., Tokyo, Japan). The gas samples were analyzed for N_2O concentrations within 2 weeks after collection. Time-serial measurements indicated that the N_2O samples remained stable in the vials for at least 4 weeks. Three ambient air samples were also collected during each sampling interval to determine the background N_2O concentrations.

The N_2O emission rate from the river surface to the closed chamber, E_c ($\mu\text{g N m}^{-2} \text{h}^{-1}$), was calculated using the following equation:

$$E_c = M(N) \times \frac{\Delta c}{\Delta t} \times \frac{PV}{ART} \quad (1)$$

where $M(N)$ is the molar mass of N (14.0 g mol⁻¹), $\frac{\Delta c}{\Delta t}$ is the linear increase in N_2O concentration in the chamber during the sampling period (ppm L⁻¹ h⁻¹), V is the chamber volume (L), A is the enclosed surface area (m²), P is the atmospheric pressure (atm), R is the universal gas constant (0.0821 L atm K⁻¹ mol⁻¹), and T is the air temperature (K).

Water samples were collected at the surface (0.05 m depth) at 2 h interval from five points across the river plane at the sampling site for DO, DOC, inorganic N, SO_4^{2-} , Cl^- , and pH analyses. Water and air temperatures were also recorded in situ at each sampling.

The dissolved N_2O concentration was determined according to Terry et al. (1981). Up to 5 mL of river water collected 5 mm below the river surface using a syringe, injected into five 29 mL pre-evacuated serum vials, and corresponding ambient air was allowed to flow into the vials. The vials were immediately placed on ice in a portable cooler for transport to the laboratory. The vials were shaking for 24 h at 4 °C to equilibrate. Subsequently, 7 mL of headspace gas samples were drawn using a syringe, and analyzed for the N_2O concentration. The N_2O concentration in the solution in the liquid phase of the bottle was calculated based on the Bunsen absorption coefficient of 1.06 at 5 °C (Terry et al., 1981; Xiong et al., 2006).

2.3. Sample analyses

The N_2O concentrations in the gas samples were determined using a gas chromatograph (Agilent 7890 A) equipped with an electron capture detector (ECD) and back-flush controlled by a 10-port valve. A 1 m × 2 mm chromatograph column packed with Poropak Q (80/100) and a 3 m × 2 mm column packed with Poropak Q (80/100) were used. The ECD temperature was 330 °C and the column temperature was 55 °C. Highly pure Ar and CH₄ (95% Ar, 5% CH₄) were used as carrier gases in the ECD (35 cm³ min⁻¹).

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