



Quantification of N₂O and NO emissions from a small-scale pond-ditch circulation system for rural polluted water treatment

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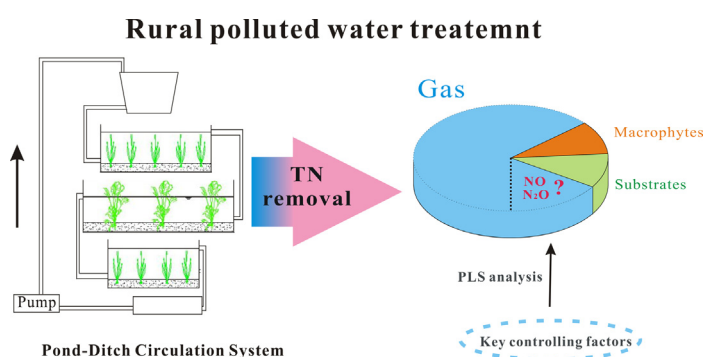
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HIGHLIGHTS

- N₂O and NO fluxes from PDCS were both quantified.
- N₂O and NO emission from the PDCS accounted for 0.17–4.32% of the TN removal.
- Partial least squares approach and Pearson's correlation coefficients were combined.
- W-NO₃⁻-N, W-DO, DO, W-pH, pH, TKN, DOC were dominant factors affecting N₂O emission.
- W-NO₃⁻-N, DO, NO₂⁻-N were dominant factors affecting the NO emission.

GRAPHICAL ABSTRACT



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ABSTRACT

The pond-ditch circulation system (PDCS) is an efficient and economical solution for the restoration of degraded rural water environments. However, little is known about nitrous oxide (N₂O) and nitric oxide (NO) emissions in the microbial removal process of nitrogen in PDCSs, and their contribution to nitrogen removal. The aim of this study was to quantify N₂O and NO emissions from the PDCS, evaluate their capacities, and elucidate the key environmental factors controlling them. The results showed that N₂O and NO fluxes were in the ranges 1.1–2055.1 μg N m⁻² h⁻¹ and 0.1–6.8 μg N m⁻² h⁻¹ for the PDCS, respectively. Meanwhile, the N₂O and NO fluxes from the two ponds in the PDCS were significantly higher than those in the static system. Moreover, the amount of N₂O and NO emissions in the PDCS accounted for 0.17–4.32% of the total nitrogen (TN) removal. According to the partial least squares (PLS) approach and Pearson's correlation coefficients, nitrate nitrogen in water (W-NO₃⁻-N), dissolved oxygen in water (W-DO), dissolved oxygen in sediment (DO), pH in water (W-pH), pH in sediment (pH), total kjeldahl nitrogen (TKN), and soil organic carbon (SOC) significantly affected the N₂O flux ($p < 0.05$), whereas W-NO₃⁻-N, DO, and nitrite nitrogen in sediment (NO₂⁻-N) significantly affected the NO emission ($p < 0.05$).

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1. Introduction

In rural areas of China, a large amount of domestic wastewater often is poorly collected and thus directly discharged into rivers and lakes, which greatly threatens aquatic environments and public health

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(Yu et al., 2012). The main sources of this wastewater are domestic and agricultural production. Hence, an effective nutrient removal method must be developed for wastewater treatment to improve water quality in rural areas. In view of rural undeveloped economies, energy shortages, and a lack of specialized staff, several decentralized treatment approaches, such as constructed wetlands (CWs) (Calheiros et al., 2015; Wu et al., 2008), tower vermifiltration (Kumar et al., 2015), and combined bioecological systems (Wu et al., 2013), have been developed and widely applied to significantly improve water quality in many rural areas of China. Recently, our group developed a new, on-site, decentralized system, designated the pond-ditch circulation system (PDCS) (Ma et al., 2015a). The PDCS has been proven a promising technology for in situ treatment of decentralized domestic wastewater owing to its low cost, easy maintenance, and energy efficiency (Ma et al., 2015a). Via the combined effect from the connected rural polluted water, plants, and microbes, the PDCS can increase the mobility and improve the self-purification ability of wastewater (Ma et al., 2015b), and hence, significantly improve water quality (effectively removing nitrogen and phosphorus) in ecological systems.

Nitrogen removal in the PDCS is an integrated process containing bacterial decomposition, plant uptake, and substrate affinity. Among these, bacteria are considered the main pathway for biological nitrogen removal from wastewater (Ma et al., 2016). Nitrification couple with denitrification, which needs both aerobic and anaerobic environments, plays a pivotal role in this nitrogen removal process (Wu et al., 2015). Meanwhile, gaseous products, such as nitric oxide (NO), nitrous oxide (N₂O), and N₂, are released during biological nitrogen removal, resulting in permanent removal of N (Alcántara et al., 2015). However, NO and N₂O, as important greenhouse gases, contribute significantly to global warming and destruction of stratospheric ozone (Lan et al., 2013; Sabba et al., 2015). Recently, increasing attention is being paid to N₂O emissions from various types of natural and engineered systems, because high N₂O fluxes were observed in many wastewater treatment processes such as those occurring in denitrifying bioreactors (Wu et al., 2014), freshwater marshes (Lu et al., 2012), CWs (Al-Isawi et al., 2017), and high rate algal ponds (Alcántara et al., 2015). Furthermore, NO emissions from wastewater treatment plants are often ignored, owing to their low flux. However, whether NO and N₂O emissions exist in the PDCS remains unclear. What are their roles in and contributions to the total nitrogen (TN) removal of the PDCS? Therefore, investigations revealing more details about the N₂O and NO emission fluxes from the PDCS are necessary for a better understanding of the underlying mechanisms of nitrogen removal concerning such a system.

N₂O and NO emissions are affected by a number of environmental factors, such as soil carbon, nitrogen substrate availability, oxygen concentration, temperature, moisture, pH, anaerobic conditions, and ammonium concentrations (Elke and Lex, 2006; Lesschen et al., 2011; Li et al., 2013). Many previous studies have reported that the concentration of DO is considered a very important parameter in controlling N₂O emissions during nitrification, with lower DO concentrations resulting in higher emissions (Kampschreur et al., 2008; Zheng et al., 1994). Schulthess et al. (1995) reported that high nitrite concentrations during denitrification led to a lower denitrification rate and accumulation of N₂O and NO. Meanwhile, low temperatures, high salinity, and lower pH repeatedly have been correlated to increased N₂O emissions (Kampschreur et al., 2009). However, the dominant factors responsible for N₂O and NO emissions need to be researched for PDCS. Hence, discerning the influence of environmental variables on the gas emissions of the PDCS is needed to better estimate and mitigate such emissions.

To fill in the knowledge gap regarding the PDCS, NO and N₂O emissions in the PDCS were continuously monitored for two months. The objectives of this study were to explore the N₂O and NO emission fluxes and their contributions to TN removal and to elucidate the underlying relationships between N₂O/NO emissions and environmental factors.

This will be beneficial for improving the efficacy of nitrogen removal in the PDCS.

2. Materials and methods

2.1. Experimental systems

Two small-scale PDCS (termed S1 and S2) were built, each having a water-distribution apparatus, pond 1, a ditch, and pond 2 (Fig. S1, Table S1). The sizes of the two ponds were as follows (cm; length × width × height): 60 × 40 × 50 (pond 1) and 50 × 30 × 40 (pond 2), with 10 cm deep sediment placed in the bottom. The trapezoidal ditch measured 35 × 25 × 30 (cm; top base × bottom base × height) in cross-section and had a length of 80 cm, and a gravel layer 10 cm deep was placed inside. Moreover, to simulate natural pond and ditch ecosystems, *Ceratophyllum demersum* L. was planted in the ponds, whereas *Iris* and *Acorus calamus* were planted in the ditch. The circulation settings of S1 and S2 were every other 4 h (3.6 L/h) and static, respectively. Meanwhile, experimental water and sediment were collected from four sites: A–D (Fig. S2). In addition, the initial physicochemical characteristics of the overlying water and basic properties of the sediments for this experiment are shown in Tables S2 and S3, respectively.

2.2. Wastewater sampling and analysis

Water samples were collected 12 times from S1 and S2 over a 60-day period, on days 1, 4, 7, 10, 14, 18, 22, 26, 30, 40, 50, and 60. TN, total phosphorus (TP), ammonia nitrogen (NH₄⁺-N), nitrate nitrogen (NO₃⁻-N), nitrite nitrogen (NO₂⁻-N), and total suspended solids (TSS) were monitored following standard procedures (State EPA of China, 2002). Chemical oxygen demand (COD) was analysed with a spectrophotometer (DRB 200, Hach, USA), and Chlorophyll a (Chl-a) was determined spectrophotometrically (Holm-Hansen and Riemann, 1978). Turbidity was measured via a turbidimeter (2100P, Hach, USA). Moreover, salinity, pH, dissolved oxygen (DO), oxidation-reduction potential (ORP), and water temperature (W-temp) were monitored using a portable Orion Star meter (520M-01A, Thermo, USA) equipped with pH/ORP/DO/Conductivity electrodes.

2.3. Gas samplings and measurements

N₂O and NO gas emissions were estimated using the static chamber technique (Uggetti et al., 2012). In this technique, each chamber covered a soil area of 0.009 m², and the internal chamber volumes for pond 1, the ditch, and pond 2 were 0.63, 0.45, and 0.54 L, respectively. The chamber, which is equipped with a thermometer, was placed in the middle of each unit (two ponds and the ditch) and pressed into the soil to a depth of around 5 cm to ensure airtightness.

Samples were always collected between 8:30 and 9:30 am, and the gas emission fluxes were measured for the same days on which the water sampling occurred, for a period of 2 months. The air temperature per chamber was simultaneously gauged by digital thermometer (WSS-413, Hangzhou, China) for subsequent flux corrections of N₂O and NO. The deployment time was set to 0, 20, 40, and 60 min. For each sampling, a 50 mL gas sample was collected from the headspace of each chamber via 60 mL plastic syringes and stored in a pre-evacuated gas-bag (E-Switch, Shanghai, China). All samples were transported to the laboratory and analysed within 48 h.

N₂O concentrations were determined by an Agilent 7890A GC system configured with a 63Ni electron capture detector (μECD), and helium gas was used as the carrier gas, at a flow rate of 6.5 mL min⁻¹. The temperatures of the injector, column, and detector were 100, 70, and 320 °C, respectively. The NO concentration was measured using a Model 42i NO-NO₂-NO_x analyser (Thermo Scientific, USA). N₂O and NO fluxes were calculated as the rate of increase in N₂O/NO in the

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