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Does carbon-nitrogen ratio affect nitrous oxide emission and spatial distribution in subsurface wastewater infiltration system?

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ABSTRACT

In order to evaluate the effects of carbon-nitrogen ratio (CNR) on nitrous oxide (N_2O) emission and quantify N_2O spatial distribution in subsurface wastewater infiltration system (SWIS), layered sampling method was introduced. Results showed that low N_2O emission rate (1.43 mg/m^2·h) and conversion rate (0.1% accounting for influent TN) were obtained when CNR increased up to as high as 10. The highest N_2O emission (3.14 mg/m^2·h) was observed at CNR of 6. Instead, independent of CNR variations, 0–75 cm was the main contributor for N_2O emission. The results indicated that layered sampling method is necessary in revealing N_2O spatial distribution in soil layers. Carbon source availability and nitrogen load and their ratio (i.e. CNR) determined N_2O emission rate. CNR of medium level leads to an increase in N_2O emission rate.

1. Introduction

Subsurface wastewater infiltration system (SWIS) is finding its wide application as an alternative to the conventional activated sludge processes in sewage treatment (Pan et al., 2016). After pretreatment, the sewage flows into SWIS under gravity via distribution pipes (beneath the soil surface to reduce potential threat from bacteria and virus) and stores in trough where anaerobic denitrification process occurs. Then, the water climbs up under capillary force and reaches the aerobic area. Finally, the wastewater infiltrates through the substrate and being collected by collection pipes. Contaminants in sewage can be removed (or trapped) by the combined actions of soil-microbial-plant. SWIS has excellent removal performance for organics, nutrients and viruses, and it has lower operation costs, easier management and better adaptability to load shock (Zheng et al., 2016).

Nitrous oxide (N_2O), a potent greenhouse gas, has a significant impact on the global environment. It is a powerful greenhouse gas with 200 times higher greenhouse effect than that of the same amount of CO_2. There are several factors involved in N_2O emission in SWIS (pH, ORP, NO_3^--N and NO_2^--N concentrations). Particularly, influent carbon-nitrogen ratio (namely COD/TN concentration, CNR) representing the relative amount of organic carbon source in a SWIS, was recognized as the crucial factor in nitrogen transformation by affecting the microbial nitrification and heterotrophic denitrification significantly. On one hand, the biological denitrification requires organic carbon source to provide electron donor (He et al., 2016). Furthermore, with the increase of CNR, soil hydraulic conductivity decreases restricting nitrification due to the increase of biomass and carbohydrate content (Fan et al., 2013). Unfortunately, the correlations between CNR and N_2O emission are controversial. Most of researchers support the views that CNR has negative effect on N_2O production. Research by Heil et al. (2015) revealed that at least some soils have the potential to oxidize NH_2OH to N_2O in a purely abiotic reaction and CNR is one of the factors to possibly explain the capacity. Then in the same year, Zhang et al. (2015) found that certain organic substrates might be the dominant factors adversely controlling the conversion rate of N_2O. He et al. (2016) and Wang et al. (2017) reported similar results in biological aerated filter (BAF) and conventional biological nitrification and denitrification reactors. Instead, some researchers hold the opposite opinions. The study by Kong et al. (2016) informed that increase CNR is beneficial to SWIS denitrification and promotes the release of N_2O. Wu et al. (2017) also reported that N_2O emission is the lowest when CNR is 5 in the wetland system, comparing with CNR 8 and 10. The controversy arises partly due to the fact that different ecosystems were getting involved, such as forests (Zhang et al., 2015), agricultural systems (Heil et al., 2015), wetlands (Zhu et al., 2014; Maucieri et al., 2017) and SWIS (Jiang et al., 2017). More importantly, static chamber method was used to measure N_2O emission rate and gross N_2O production was achieved. Given the fact that several biotic reactions (ammoxidation, nitrification and denitrification) have been identified leading to N_2O emission, stable chamber method can hardly reveal their individual contributions and N_2O spatial distribution as well. In order

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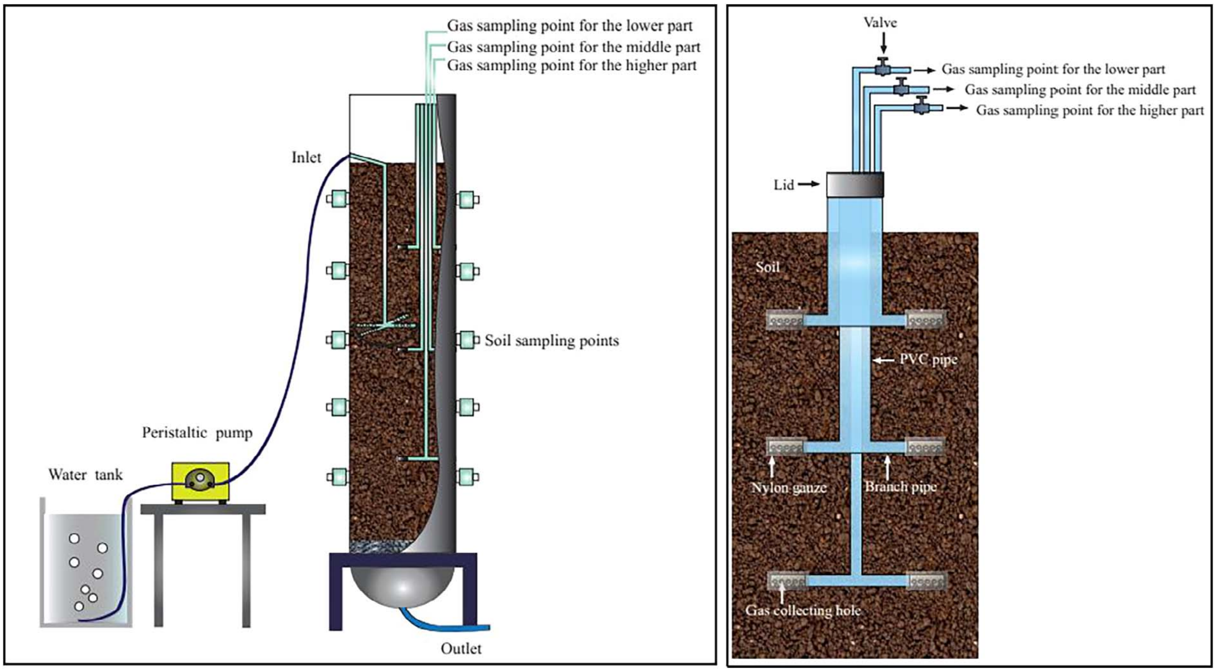

Fig. 1. Diagram of SWIS and N_2O layered sampler embedded in SWIS.

to solve this problem, the present study introduced layered sampling method for N_2O sampling. The key objective was to explore the effect of CNR variations on N_2O emission rate and investigate N_2O spatial distribution.

2. Materials and methods

2.1. System description

Four parallel soil columns made of plexiglass were constructed (height 130 cm and internal diameter 30 cm) and operated indoors. Wastewater was distributed via a 2-cm-diameter perforated water distribution pipe placed at 50 cm depth below the soil surface in each infiltration system. 10 cm of gravel (10–20 cm, diameter) wrapped up the distribution pipe to protect clogging. Filled substrate from top to bottom was farmland soil (0–30 cm), bio-substrate (30–110 cm), coarse sand (110–120 cm) and gravel (120–130 cm). Dissolved oxygen (DO) electrodes were placed in the center of each SWIS at 30, 60, 90, 110 and 120 cm depths. Polarographic electrodes were employed (VisiPro DO 225, purchased from Hamilton Bonzduz AG, Shanghai). The response time of electrodes was 90 s. At each sampling day, 3 parallel readings for each point were recorded and the averages were calculated. The bio-substrate consisted of sludge, slag and farmland soil mixed evenly with volume ratio of 1:2:7. The physico-chemical characteristics of the substrate were listed in previous reports (Li et al., 2011). 10 cm of deep gravel (10–20 cm, diameter) was prepared at the bottom to support infiltration system and evenly distributed the treated water. The treated water was gathered at the bottom.

Vertically, each soil column was equipped with a N_2O *in-situ* sampler in the center, which was considered to be casing string design (Fig. 1). The plexiglass tubes were 25, 50 and 75 cm in length and 2, 3 and 4 cm in internal diameters from the inner layer to the outer layer, respectively. There were uniform sampling holes wrapped in nylon mesh at both ends of horizontal plexiglass tubes. N_2O could be sampled from upper layer (0–35 cm), middle layer (35–75 cm) and lower layer (75–100 cm) simultaneously.

2.2. Wastewater and operation

Domestic wastewater was pretreated in a septic tank prior to being flowed through each SWIS. The ranges of wastewater after pretreatment were as follows: COD 156–220 mg/L, BOD_5 103–189 mg/L, NH_4^+-N 19–37 mg/L, TN 21–39 mg/L, TP 3.3–4.0 mg/L and pH 6.9–7.2.

Two runs were arranged. During the first stage (0–72 h), static chamber method was used to characterize gross N_2O emission with four soil columns operated at four CNR (sampling and calculation method was listed below). Pollutant removal efficiencies were also evaluated during this stage. For the next 10 weeks (the second experimental stage), static chambers were removed. N_2O layered samplers were embedded in the center of each column. During the whole experimental period, wet and dry alternation (i.e. distribution and "rest" alteration) was adopted as ratio of 1:1. That means four SWISs were fed with pretreated wastewater continuously for 12 h (between 8:00 a.m. and 8:00 p.m.) followed by a drying period of 12 h. When the system is "drying", a stopper has been used to cut air pathway into the column from the outlet. So the air cannot be drawn into the column.

The lawn water demand was employed as the design basis of the hydraulic load, which primarily reflects in the lawn evapotranspiration (Zhang et al., 2015). The average potential evapotranspiration of the lawns in Shenyang ranged from 0.05 to 0.17 $m^3/m^2 \cdot d$. Hence, hydraulic load in this study was designed to be 0.10 $m^3/m^2 \cdot d$. Tests were conducted from June 5 to August 16, 2016 (room temperature 25.7 ± 1.2 °C). But the SWISs were operated one month before sampling to allow microbial biomass maturation.

2.3. Sampling and analytical methods

Gas sampling was done at 0, 30, 60, 90 and 120 min after opening the valve of the sampler. For each sampling time point, 36 gas samples (4 soil columns, sampling from upper, middle and lower layer, 3 parallel) were collected using 25-mL plastic syringes. Then samples were stored in preevacuated 20-mL vials. The concentration of N_2O was