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Effect of particle size and composition of suspended sediment on denitrification in river water

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HIGHLIGHTS

- Denitrification rate was negatively related to particle size of SPS.
- Denitrification in the <20 μm SPS had the highest N_2 emission rate of 0.27 mg-N/m^3 d.
- $^{15}N_2O$ production in the system with SPS of 20–50 μm was 14.8 times that of 100–200 μm.
- The denitrifying bacteria population increased with TOC content of SPS.
- Different oxic/anoxic conditions existed around SPS with different particle size.

GRAPHICAL ABSTRACT

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ABSTRACT

Rivers with high suspended sediment (SPS) concentration are common worldwide, and previous studies reported the occurrence of denitrification on SPS. In this work, effect of particle size and composition of SPS on denitrification in river water was studied in laboratory. The ^{15}N isotope tracer technique was used to investigate the denitrification in water containing 8 g L^{-1} SPS with different particle sizes, including <20 μm, 20–50 μm, 50–100 μm, and 100–200 μm. The results showed that the denitrification rate was negatively related to particle size, and the SPS with particle size below 20 μm had the highest $^{15}N_2$ emission rate of 0.27 mg-N/$m^3 \cdot$d, which was twice that of 100–200 μm. The denitrifying bacteria population in the system decreased with the increase of particle size, which was positively correlated with denitrification rate ($p < 0.05$). There was a positive correlation between organic carbon content of SPS and denitrifying bacteria population ($p < 0.01$), indicating that organic carbon is a key factor influencing denitrifying bacteria. Different from the $^{15}N_2$ production, $^{15}N_2O$ emission rate reached the highest of 1.02 μg-N/$m^3 \cdot$d in the system containing SPS of 20–50 μm, which was 14.8 times that of 100–200 μm. This was due to the difference in denitrifying bacteria species in different systems due to different oxic/anoxic conditions around SPS. This study suggests that not only the SPS concentration but also the SPS size and composition should be considered in studying the nitrogen cycle in river systems, especially for the production of N_2O.

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1. Introduction

Nitrogen (N) is a fundamental and vital element for the composition of organisms and ecosystems (Galloway et al., 2004; Howarth and Marino, 2006; Hsiao et al., 2014), but the excess discharge of nitrogen caused by wide but unreasonable application has led to eutrophication, deterioration of water quality, decrease of biodiversity, and destruction of aquatic habitats (Rabalais, 2002). Denitrification is a biological process that can permanently remove nitrogen from ecosystems by reducing nitrate (NO_3^-) to dinitrogen (N_2) in hypoxic conditions; it plays a significant role in controlling the fate of nitrogen in rivers (Alexander et al., 2000). During denitrification, NO_3^- is reduced to nitric oxide (NO), nitrous oxide (N_2O) via nitrite (NO_2^-), then to N_2. As an intermediate during denitrification, N_2O may release to and affect atmosphere by acting as an important ozone-depleting substance as well as greenhouse gas (Crutzen, 1970; Cicerone, 1987), and its global warming potential is 310-fold higher than carbon dioxide.

River is an important link between terrestrial and aquatic ecosystems for nitrogen cycling, and SPS commonly exists in rivers worldwide because of the external input (Sivakumar, 2002; Putnam and Pope, 2003; Yang et al., 2008; Yellow River Sediment Bulletin, 2013). Our previous study found the occurrence of denitrification on SPS in rivers and the increase of SPS concentration can accelerate the denitrification rate (Liu et al., 2013a). This is due to the fact that low-oxygen microsites exist around SPS, which can provide circumstances for the denitrifying bacteria to conduct denitrification. The low-oxygen conditions around SPS are probably caused by the following processes. Lots of aerobic heterotrophic organisms tend to attach on the SPS particle surface (Netzband et al.,1999; Xia et al., 2009; Ochs et al., 2010), and oxygen will be consumed by the biodegradation of organic matters which is mediated by these organisms on SPS. Moreover, plenty of reducing inorganic ions (NH_4^+, Fe^{2+}) are easily adsorbed on the particle surface (Sweerts et al., 1991; Xia et al., 2004; Wang et al., 2012), and oxidation of these ions will consume oxygen.

Several main factors controlling denitrification have been identified in previous studies, including the supply of nitrate, organic carbon, and oxygen concentrations (Knowles, 1982; Seitzinger, 1988). For example, Zimmerman and Benner (1994) reported that the differences in sediment denitrification among the examined estuaries of Galveston Bay are attributed mainly to sediment organic carbon content. SPS with different particle sizes and composition may have different low-oxygen microsites and provide different electron donors for denitrification, which may affect denitrification on SPS. Therefore, we hypothesized that particle size and composition of SPS play a significant role on denitrification in river waters.

As the largest turbid river around the world, the Yellow River was selected as a case to explore the effect of SPS characteristics on denitrification by using ^{15}N isotope tracer method. Compared with traditional acetylene inhibition method, ^{15}N isotope tracer method has been adopted by more and more researches for its advantage of high sensitivity and convenience (Mulholland et al., 2009; Hsiao et al., 2014; Hou et al., 2014). The main objectives of the present study were: 1) to identify the denitrification rate, expressed as $^{15}N_2$–N emission rate, in systems containing SPS of different particle sizes; 2) to compare the effect of SPS particle size and composition on the production of N_2 and N_2O; 3) to investigate the influencing mechanisms of particle size on denitrification by analyzing the population and species of denitrifying bacteria, the content of TOC in SPS with different particle sizes, and the oxic/anoxic conditions around SPS, hoping to fill some gaps in denitrification occurring on SPS in rivers.

2. Materials and methods

2.1. Sample collection

As the second longest river in China, the Yellow River is 5464 km long and has a basin area of 7.52×10^4 km^2. The average SPS concentration of the Yellow River is 26 g L^{-1} (Yellow River Sediment Bulletin, 2013). The sampling site was located at Aishan Hydrological Station (116°18′08.4″E, 36°16′10.4″N) in the downstream of the Yellow River, 402 km away from the river mouth, where the SPS concentration ranged between 5.91 and 17.6 g L^{-1} (Yellow River Sediment Bulletin, 2013). Water was sampled at 0.2 m below the surface of the water, and surface sediment (0–10 cm depth) was sampled using a grab sampler, and then put into sterilized polypropylene bottles. Temperature, pH, dissolved oxygen content of water and redox potential were measured on site. All samples were collected in May 2014, kept on ice in a freezer and then transported back to laboratory for further analysis, which was conducted within 72 h. Nitrate, nitrite, and ammonium concentrations in the water sample were measured. Four groups of the collected sediment with particle size in the range of <20 μm, 20–50 μm, 50–100 μm, 100–200 μm were obtained in laboratory using wet-sieving method, and the size distribution of each group was determined. To avoid the loss of bacteria during the wet-sieving process, the remaining water after wet-sieving was filtered through a 0.7 μm filter to remove the sediment, and then collected for the preparation of the water in simulation experiments.

2.2. Laboratory simulation experiment

A set of chambers composed of polymethyl methacrylate (PMMA) columns were prepared for experiments, and the end was sealed by a rubber stopper which is combined with a mechanical agitator to simulate the state of SPS under hydrodynamic conditions (details in Liu et al., 2013a). A total of 6.4 g (dry weight) SPS with different particle sizes were respectively added into the chambers containing 800 ml of the collected water and 5 mg L^{-1} $^{15}NO_3^-$–N as $K^{15}NO_3$ (99.0 atm% ^{15}N), which was in agreement with [NO_3^-] (3.13 ± 0.04 mg L^{-1}) in the water sample collected from Aishan station (Table S1, Supporting Information). A total of 5 mg-C L^{-1} glucose was added into the system to simulate the dissolved organic carbon concentration in river waters. In order to maintain the suspension of sediment, the rotation speed was set to 200 rpm, and the incubation temperature was 25 °C. The gas samples were collected at predetermined intervals to determine the yield of $^{15}N_2$ and $^{15}N_2O$. After each sampling, temperature, pH, dissolved oxygen content of water and redox potential were determined, and then 30-min aeration to the water phases of the chambers was conducted to purge the old $^{15}N_2$ and $^{15}N_2O$ from the chambers and guarantee oxygen saturated in the process of incubation (Liu et al., 2013a). In addition, at the 7th day of the incubation, 2–3 ml water samples containing SPS were collected from each column to analyze the O_2 flux around the SPS. Determination of denitrifying bacteria and SPS size was conducted at the end of the experimental period. In addition, two sets of control experiments were conducted, one was implemented with sterilized SPS sample and water, and mercuric chloride was added to the system at a final concentration of 0.5% to inhibit the microbial activities; the other was conducted with the water containing bacteria but without SPS to explore denitrification in water. Each experimental set was conducted in triplicate.

2.3. Chemical analysis

Gas samples of $^{15}N_2$ and $^{15}N_2O$ were analyzed for ^{15}N with a Delta V Advantage isotopic ratio mass spectrometer (Thermo Fisher Scientific), which is attached with an automated PreCon unit. ^{15}N content were expressed by the $\delta^{15}N$ notation as $\delta^{15}N(‰) = (^{15}N/^{14}N_{sample}/^{15}N/^{14}N_{std} - 1) \times 1000$, and the analytical precision of the $\delta^{15}N$ measurement was ±0.3‰.

SPS size characteristic was determined using a laser particle analyzer (Microtrac S3500, USA), and the results showed that the size of SPS after the experiment was almost the same as that before the experiment (Table S2 in Supporting Information). To determine the moisture content of SPS, the weight difference between wet and dried (100 °C