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Denitrification occurring on suspended sediment in a large, shallow, subtropical lake (Poyang Lake, China)☆

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ABSTRACT

Many lakes and rivers are enriched with high levels of suspended sediments (SPS). Denitrification occurring on suspended sediments (DSS) may play an important role in nitrogen removal in water columns with high SPS concentrations. Poyang Lake, with dramatic hydrologic variations, has high spatial and seasonal variation of SPS, and we hypothesized that DSS and nitrogen removal in this lake would vary similarly. DSS in Poyang Lake was determined by the traditional acetylene-inhibition method combined with a batch mode assay. Laboratory simulation experiments were also conducted to examine the factors controlling denitrification occurring on SPS. Seasonally, DSS rates at 15 sampling sites in Poyang Lake were 0.63 ± 0.24, 0.29 ± 0.17, 0.25 ± 0.18, and 0.52 ± 0.37 μmol $N \cdot L^{-1} \cdot d^{-1}$, respectively in spring, summer, autumn, and winter. Spatially, average DSS rates were higher in the northern lake area, which is connected to the Yangtze River, than in the upstream and central lake area. Lowest DSS rates occurred in semi-closed bay and dish lakes. Spatial and seasonal variations of DSS rates were affected by a combination of factors, in which nitrate concentrations, SPS composition, and concentrations of organic-SPS were the most important. These influencing factors were seasonally dependent, with nitrate concentrations having stronger effects on DSS during wet seasons than dry seasons. Results from a multiple stepwise regression model also demonstrated that DSS tended to occur on fine particles (e.g., clay particles, <4 μm). Evaluation of annual nitrogen loss by DSS was estimated according to the seasonal water budget and DSS rates in Poyang Lake. The total nitrogen loss by DSS was estimated to be 10800 ± 6090 t, which accounted for 2.8–9.9% of the nitrogen input, and this proportion was comparable to nitrogen removal by sediment denitrification. This result confirms that DSS was an important nitrogen sink in this large, turbid lake.

1. Introduction

Anthropogenic addition of bioavailable nitrogen to ecosystems has increased in recent decades (Galloway et al., 2004; Mulholland et al., 2008; Vitousek et al., 1997). The excessive input of nitrogen to aquatic systems may fuel high rates of plant growth and lead to eutrophication (Qu et al., 2005; Seitzinger, 2008). However, not all of the anthropogenic nitrogen reaching streams and lakes is discharged into coastal systems (Mulholland et al., 2008), and there is a net loss of N (Hill, 1979, 1983). Thus, freshwater ecosystems, including lakes, streams, rivers, and wetlands, are generally a large sink for reactive nitrogen (Finlay et al., 2013). Denitrification, which transforms nitrates into gaseous products, such as molecular dinitrogen (N_2) and nitrous oxide (N_2O) gas (Seitzinger, 2008), is the dominant mechanism for substantial removal of fixed nitrogen from the biosphere (Altabet et al., 1995; Finlay et al., 2013). In sediments and biofilms, denitrification is restricted to a narrow zone immediately below the aerobic-anaerobic interface, and the depth distribution of denitrification activity is related to NO_3^- diffusion (Christensen et al., 1989; Nielsen et al., 1990a,b). Studies of denitrification in aquatic systems are mostly focused on bottom-sediments, which are thought to be the dominant nitrogen removal location in aquatic systems, due to the anoxic environment, nitrate, and availability of electron donors (Christensen et al., 1989). Thus, denitrification rates across the sediment-water

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interface (SWI) in different types of aquatic systems around the world are well documented (Bruesewitz et al., 2011; Christensen et al., 1990; Garcia-Ruiz et al., 1998; Seitzinger, 1988). Furthermore, some researchers have studied gaseous nitrogen removal at the water-air interface through direct measurement of excess dissolved N_2 and/or N_2O and calculation of net water-air N_2 and/or N_2O fluxes (Chen et al., 2014a, 2014b; Yan et al., 2012; Zhao et al., 2015). Release of excess dissolved N_2 was attributable mainly to denitrification in sediments underlying deep water (Chen et al., 2014a), and the role of suspended sediments (SPS) was often neglected (Liu et al., 2013). Increasing SPS accelerated denitrification in both SPS + water systems and bottom-sediment + SPS + water systems in the turbid Yellow River (Liu et al. 2013). Suspended matter in natural waters is a complex mixture of organic and inorganic detritus, Fe-Mn oxyhydroxides, clay minerals, carbonates, phytoplankton, zooplankton, bacteria, and other organisms (Odman et al., 1999; Turner and Millward, 2002). SPS may support enhanced bacterial activity (heterotrophic and autotrophic) (Crump and Baross, 1996; Ochs et al., 2010; Plummer et al., 1987) and play an important role in the reactivity, transport and biological impacts of substances in aquatic environments (Turner and Millward, 2002). Therefore, SPS and the water column may provide similar essential conditions for denitrification occurring on suspended sediments (DSS), such as low-oxygen microsites, SPS-associated denitrifying bacteria, organic matter and nitrate (Garneau et al., 2009; Liu et al., 2013; Michotey and Bonin, 1997; Turner and Millward, 2002). Compared with bottom-sediments, SPS may be more advantageous for denitrification due to access to nitrate for denitrifying bacteria on SPS (Liu et al., 2013).

Poyang Lake (28°22′ to 29°45′N, 115°47′ to 116°45′E) is located at the south bank of the middle reaches of the Yangtze River in China. As the largest freshwater lake in China, Poyang Lake covers a watershed area of 1.622×10^5 km^2, and the annual discharge of the lake is approximately 1.457×10^{11} m^3, which accounts for 15.6% of the average Yangtze River runoff (Shankman et al., 2006; Wu et al., 2013). Poyang Lake receives water from five inflows, including the Ganjiang, Fuhe, Xinhe, Xiushui, and Raohe Rivers, and it freely exchanges water with the Yangtze River (Fu et al., 2003; Wang et al., 2011; Zhang et al., 2014b). There is considerable seasonal variation (more than 10 m) of lake water level due to the combined effects of catchment inflows, interaction with the Yangtze River, rainfall in the local area (Shankman et al., 2006; Wu et al., 2013; Zhang et al., 2014b), and/or the Three Gorges Dam (Zhang et al., 2012). As a result, the surface area of Poyang Lake fluctuates substantially throughout the year; it can exceed 4000 km^2 during the summer wet season and decrease to less than 1000 km^2 during winter (Zhang, 1993). The significant seasonal fluctuations in Poyang Lake create extensive wetlands across an ephemeral region of some 3000 km^2, which provides vital habitats for many species, including rare or endangered birds (Feng et al., 2013; Wang et al., 2013; Zhang et al., 2014b). SPS in Poyang Lake are mainly affected by runoff of water and sands from the five inflow rivers, sands from dredging/mining activities, interaction with the Yangtze River, and other human activities (Feng et al., 2012; Wang et al., 2012; Zhang, 2012). With rapid population growth and economic development in the watershed, nitrogen and phosphorus inputs to Poyang Lake have increased in recent years and caused eutrophication in some lake regions. According to Liu et al. (2012), an average of 170 thousand tons of nitrogen enters Poyang Lake every year. However, there is scant data on nitrogen removal in Poyang Lake. Particularly, there is no information on nitrogen removal rates from the turbid water column of this lake, which has hindered understanding of nitrogen budget.

Our objectives in this study are to: (1) investigate the spatial and seasonal variability of DSS rates; (2) analyze the relationship between DSS rates and SPS characteristics and other environmental factors, such as nitrate, SPS concentrations, and dissolved organic matter; and (3) estimate the nitrogen loss by DSS and its proportion of the annual nitrogen input in Poyang Lake. Our research on DSS in Poyang Lake will be valuable for constructing the nitrogen budget and determining the ecological role of SPS in this ecosystem.

2. Materials and methods

2.1. Site description and sample collection

Sampling sites included four main areas: upstream, main lake, downstream, and wetland (Fig. 1). Downstream and upstream sites were connected to inflowing rivers or the Yangtze River and often had higher SPS concentrations due to higher flow rates and sand dredging/mining activities (Zhang, 2012). Wetland sites had very low SPS in summer, due to low flow rates and hydrological isolation. Main lake sites had intermediate flow rates due to dissipation over the large surface area (Zhang et al., 2014a).

Water and SPS samples from 15 regular sites (PY1-15) were collected four times (spring = April, summer = July, autumn = October, winter = January) from April 2014 to January 2015. Water and SPS samples from additional sites PYW1-9 were collected during summer and winter (PYW9 only in winter). The water level of Poyang Lake was lowest from December to March and increased from April to June (Liu et al., 2012). High water level maintained from July to September and decreased from late October (Liu et al., 2012). Hence, winter and spring were the dry seasons for Poyang Lake, while summer and autumn were wet seasons.

Water samples were collected using a polymethyl methacrylate water sampler (UWITEC, Austria) at 0.5 m below the water surface to avoid contamination by floating pollutants, and 200 mL of water was drawn into 318 mL sterile glass serum bottles. Water samples were collected in triplicate at each site and sealed with airtight rubber plugs. In addition, a 15 L polyethylene container was used to collect water and SPS samples for the experimental analysis of physical and chemical properties of water and SPS. pH, water temperature (WT), dissolved oxygen (DO) and salinity were measured *in situ* using a multi-parameter water quality analyzer (YSI Professional Plus, 6600V2, USA). Water depth (WD) was determined using a Speedtech SM-5 Portable Depth Sounder (USA). Water transparency was determined using a Secchi disk. All samples were kept cool at 4 °C and shaded immediately after sampling. Experimental analysis was conducted within 48 h after sampling.

2.2. Denitrification experiment

Denitrification rates caused by SPS were determined using the acetylene-inhibition method combined with batch mode assay. An air headspace of 20 mL was extracted from each serum bottle and replaced with an equivalent volume of C_2H_2 using an air-tight syringe. C_2H_2 in the headspace accounts for 16.9% (v/v) and is sufficient to inhibit the reduction of N_2O to N_2. The serum bottles were continuously shaken at 120 rpm in an oscillator at field temperature. After 12 h, 1 mL of head space (for t0) from each serum bottle was sampled using a 1 mL air-tight glass syringe and injected into a GC (Shimadzu GC-2014C, Japan). The headspace was then sampled every 24 h for at least 7 days.

Concentrations of dissolved N_2O in headspace were calculated using Dalton's Law (see SI). Assuming that equilibrium exists between the headspace and water column, the concentration of dissolved N_2O in the water column was calculated from concentrations measured in the headspace (Weiss and Price, 1980)