Human land uses enhance sediment denitrification and N_2O production in Yangtze lakes primarily by influencing lake water quality

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Abstract

Sediment denitrification in lakes alleviates the effects of eutrophication through removal of nitrogen to the atmosphere as N_2O and N_2. However, N_2O contributes notably to the greenhouse effect and global warming. Human lands uses (e.g., agricultural and urban areas) strongly affect lake water quality and sediment characteristics, which, in turn, may regulate lake sediment denitrification and N_2O production. In this study, we investigated sediment denitrification and N_2O production and their relationships to within-lake variables and watershed land uses in 20 lakes from the Yangtze River basin in China. The results indicated that both lake water quality and sediment characteristics were significantly influenced by watershed land uses. N_2O production rates increased with increasing background denitrification rates. Background denitrification and N_2O production rates were positively related to water nitrogen concentrations but were not significantly correlated with sediment characteristics and plant community structure. A significant positive relationship was observed between background denitrification rate and percentage of human-dominated land uses (HDL) in watersheds. Structural equation modelling revealed that the indirect effects of HDL on sediment denitrification and N_2O production in Yangtze lakes were mediated primarily through lake water quality. Our findings also suggest that although sediments in Yangtze lakes can remove large quantities of nitrogen through denitrification, they may also be an important source of N_2O, especially in lakes with high nitrogen content.

1 INTRODUCTION

As a global environmental issue, land use change has numerous consequences, ranging from changes in global climate to local water quality (Foley et al., 2005). Conversion of natural land use (e.g., forests and wetlands) to human land use (e.g., cropland and urban areas) releases large quantities of pollutants, including nitrogen, and has widespread effects on biodiversity and ecological function of lakes, rivers and other water bodies (Müller et al., 1998). Lake water quality has deteriorated worldwide due to increasing point- and nonpoint-source pollution in watersheds (Nielsen et al., 2012; Baron et al., 2013). For instance, the percentage of Chinese lakes classified as eutrophic has rapidly increased from 41 % in 1980 to 85 % in 2005 (Liu et al., 2010). The effects of watershed land uses on lake water quality are well-documented (Liu et al., 2012; Nielsen et al., 2012). However, relatively few studies have reported the relationship between watershed land uses and lake sediment characteristics, and their results are inconsistent (Müller et al., 1998; Bruesewitz et al., 2011).

Denitrification plays an important role in lake ecosystems by removing excess nitrogen input from watersheds (Seitzinger et al. 2006; McCrackin and Elser, 2010). The denitrification process predominantly occurs under anaerobic or anoxic conditions and involves the successive reduction of nitrate (NO_3^-) to nitrite (NO_2^-), nitric oxide (NO), nitrous oxide (N_2O) and, finally, dinitrogen gas (N_2) (Sirivedhin and Gray, 2006). In lakes, denitrification capacity is greatest in sediments due to the favourable environment for denitrifying bacteria and fungi (McCrackin and Elser, 2010; Baron et al., 2013). Sediment denitrification can account for approximately 82 % of the total nitrate loss in lakes (Kreiling et al., 2011). Despite the significant increases in nitrogen loading in lake ecosystems and the growing attention to water quality degradation and toxic algal blooms, our understanding of large–scale sediment denitrification in lakes remains limited (McCrackin and Elser, 2010; Bruesewitz et al., 2011; Liu et al., 2015).

N_2O is an intermediate product of sediment denitrification and may be released into the atmosphere in substantial amounts under certain conditions (Seitzinger and Kroeze, 1998; Hefting et al., 2006; Freymond et al., 2013). In Lake Taihu of China, both the littoral and pelagic zones can release large quantities of N_2O, especially during the summer algal bloom period (Wang et al., 2007). However, N_2O fluxes in the pelagic regions of shallow boreal lakes are reported to be negligible (Huttunen et al., 2003), which indicates that lakes are only moderate sources of N_2O (Mengis et al., 1997). N_2O is a powerful greenhouse gas that is approximately 300 times stronger than CO_2 and is responsible for approximately 6 % of global warming (IPCC, 2007). Thus, it is important to examine the N_2O production during sediment denitrification. Seitzinger and Kroeze (1998) have indicated that the relative N_2O production (i.e., the $N_2O/(N_2O+N_2)$ ratio) for heavily polluted river and estuary sediments is approximately 0.03. By contrast, the relative N_2O production for lakes is still poorly understood (McCrackin and Elser, 2010).

At the within-lake scale, overlying water quality and sediment characteristics are recognised as the primary determinants of sediment denitrification and N_2O production (Saunders and Kalff, 2001; Hasegawa and Okino, 2004; Zhong et al., 2010; Rissanen et al., 2011; Liu et al., 2015). Recently, several studies have found that aquatic plant communities can also greatly influence sediment denitrification in fresh waters (e.g., Forshay and Dodson, 2011; Wang et al., 2013; Jacobs and Harrison, 2014). Plants may affect denitrification primarily through the modification of carbon inputs and soil redox conditions via oxygen excretion from roots (Sutton-Grier et al., 2013). At the watershed scale, few studies have addressed the indirect effects of watershed land uses on lake sediment denitrification and N_2O production (but see McCrackin and Elser, 2010; Bruesewitz et al., 2011; Liu et al., 2015), and no studies have examined whether the indirect effects are mainly mediated through lake water quality or sediment characteristics. A better understanding of multi-scale determinants of sediment denitrification in lakes is necessary to achieve high nitrogen removal efficiency and low N_2O production rates.

In this work, we investigated sediment denitrification and N_2O production and their relationships to within-lake variables and watershed land uses in 20 lakes from the Yangtze River basin in China. We hypothesised that the human land uses in watersheds would influence sediment denitrification and N_2O production in lakes, primarily via effects on lake water quality. The objectives of this study were (1) to examine if watershed land uses could influence lake sediment characteristics, as well as water quality, (2) to determine the effects of water quality, sediment characteristics and plant community structure on lake sediment denitrification and N_2O production, and (3) to reveal the mechanisms of lake sediment denitrification and N_2O production in response to human land uses in watersheds for the first time.

2 METHODS

2.1 Study sites

The Yangtze River, originating from the Qinghai-Tibet Plateau and flowing into the Pacific Ocean in the city of Shanghai, is the largest river in China and has a drainage area of approximately 1.8 million km^2 and a total length of 6300 km (Fig. 1). There are 648 lakes with areas greater than 1 km^2 in the Yangtze River basin (Wang & Dou, 1998). Most of these lakes are concentrated in the middle and lower Yangtze River basin (i.e., the Yangtze floodplain; Fig. 1), where alluvium predominates. Lakes in the Yangtze floodplain are generally shallow with short hydraulic retention times (Liu et al., 2011).

Over the past decades, the watershed land uses of many Yangtze lakes have undergone substantial changes (Liu et al., 2012). Large areas of forests, wetlands and other natural landscapes have been transformed into agricultural, industrial, and urban lands. For instance, the percentage classified as built-up lands in the Chaohu Lake watershed has increased from 6.59 % to 9.58 % during the period of 1979–2008 (Wu, 2011). As a result of watershed land use changes, water quality in many lakes has deteriorated considerably in recent years, and up to 86 % of the Yangtze lakes are classified as eutrophic or even hypereutrophic (Yang et al., 2010).

2.2 Field sampling

During the summer (from July 30 to August 7) of 2012, twenty lakes in the middle and lower Yangtze River basin of China were selected non-randomly on the basis of ease of access (Fig. 1). These selected lakes covered a wide range of physical and chemical conditions and vegetation characteristics (Wang and Dou, 1998). Almost all these lakes are shallow (mean depth < 4 m), except for the Lake Bohu with a mean depth of 4.41 m. The largest lake is Lake Longganhu with an area of 316.20 km^2, while the smallest one is Lake Yiaihu with an area of 3.21 km^2. The name, geographic location and morphology characteristics of these studied lakes are listed in Table S1. To obtain representative samples, one transect from the littoral zone to the lake centre was randomly established in each lake and 3–4 sampling sites were chosen along this transect at regular intervals based on lake area (Yang et al., 2008). Littoral sampling sites were generally located at a water depth less than 1.5 m, approximately 50–100 m away from the lake shore.

At each sampling site, three replicate surface sediments were randomly collected within an approximately 50 m^2 area from a boat using a Peterson dredge and then mixed and homogenised to form a composite sample. For each site, approximately 1 kg of sediment was placed in a sealed plastic bag and stored at approximately 5°C in a refrigerator until return to the laboratory. At each sampling site, a 200-mL unfiltered water sample was collected at a depth of approximately 1 m for use in the denitrification and N_2O production assays. At the same time, we collected an additional 200-mL water sample at the same depth for water quality analyses in the laboratory. Water sampling was performed before sediment collection to prevent sediments from being resuspended and thereby contaminating the water. At each sampling site, submerged macrophytes were sampled using a pronged grab (25 cm × 35 cm) with three replicates, and the species richness (i.e., mean species number recorded in a sampling site) and fresh plant biomass were calculated.

2.3 Denitrification and N_2O production assays

The most common method for measuring sediment denitrification is based on the ability of acetylene to inhibit the reduction of N_2O to N_2 during denitrification (Groffman et al., 1999). Although the acetylene blockage technique has a number of limitations (Yu et al., 2010; Felber et al., 2012), it is still amenable to large-scale comparisons of denitrification, especially for systems with moderate or high NO_3^- concentrations (Groffman et al., 2006). In this study, we measured potential and background denitrification rates of lake sediments using the acetylene blockage technique. The potential denitrification rate was measured under optimal conditions (by supplying an excess of NO_3^- and organic carbon and ensuring anoxia) and thus provided an upper-bound estimate of in situ denitrification. Background denitrification (i.e., unamended denitrification) rate was a conservative estimate of in situ denitrification without carbon and NO_3^- amendments because acetylene also inhibited nitrification (NO_3^- production). The increase in N_2O concentrations was always linear throughout the short assay duration, therefore, short-term incubation (2–4 h) was recommended for measuring sediment denitrification of lakes (Bruesewitz et al., 2012).

For potential denitrification rate assays, 50 g of homogenised sediments from each sampling site were slurried with 30 mL of incubation solution (final concentrations: 0.1 g/L KNO_3, 0.18 g/L glucose and 1 g/L chloramphenicol) in a 250 mL serum bottle. Each bottle was then sealed and purged with N_2 gas for 2 min to induce anoxic conditions. Approximately 10 % of the bottle headspace was replaced with acetylene to block the conversion of N_2O to N_2 during denitrification. We measured background denitrification and N_2O production rates using a similar procedure, but with the addition of 30 mL of unfiltered lake water instead of the incubation solution (McCrackin and Elser, 2012). Parallel incubations with and without acetylene (10 % vol/vol in the bottle headspace) were used to differentiate between background denitrification and N_2O production (McCrackin and Elser, 2010). All bottles were then incubated in the dark for 4 h at 25°C (the approximate in situ water temperature). At the beginning and end of incubation, 5 mL of headspace gas samples were collected from each bottle (after shaking vigorously) using a syringe. The N_2O concentrations were measured using a gas chromatograph (Agilent 7890, Santa Clara, CA, USA) equipped with an electron capture detector.

Potential denitrification, background denitrification and N_2O production rates were calculated as the difference between the initial and final headspace N_2O concentrations (corrected for N_2O dissolved in water, Bunsen coefficient = 0.544) divided by the incubation time (Hayakawa et al. 2012), and was expressed on the basis of dry matter of sediment (ng N g^{-1} h^{-1}). The relative N_2O production was calculated by dividing the N_2O production rate by the background denitrification rate (García-Ruiz et al., 1998; McCrackin and Elser, 2010).

2.4 Sediment characteristics and water quality measurements

Sediment pH was measured in a soil to water ratio of 1: 5 (v/v) using a pH meter and bulk density was determined by weighing soil cores of known volume after drying for 24 h at 105 °C. Sediment moisture was measured gravimetrically (24 h at 105 °C) from 30 g sediment samples. Sediment total nitrogen (STN) was measured using the Kjeldahl method after digesting samples in a digestor using a sulfuric acid/mercuric oxide catalyst. Sediment total carbon (STC) content of air-dried samples was analysed by a TOC analyser (Vario TOC cube, Elementar, Germany).

Conductivity (Cond), oxidation–reduction potential (ORP), dissolved oxygen (DO), nitrate (NO_3^-) and ammonium (NH_4^+) were measured on site at the sampling depth using a YSI 6920 multiparameter water quality probe (YSI Inc., Yellow Springs, Ohio, USA). Total nitrogen (TN) concentration was analysed using the micro-Kjeldahl method. The concentrations of total carbon (TC) and total organic carbon (TOC) in waters were measured with a TOC analyser (Vario TOC cube, Elementar, Germany).

2.5 Watershed land use calculation

After delineating the watershed boundaries of the 20 studied lakes using a 1 km resolution digital elevation model, overlay functions were used to extract a land use map of each lake's watershed from 100 m resolution national land cover data in ArcGIS 10 software (ESRI, Redlands, California, USA). The national land cover data, interpreted from recent Landsat TM images, were obtained from the Data Sharing Infrastructure of Earth System Science in China (http://www.geodata.cn/).

The original land use classes were further grouped into four main categories: (1) vegetation, including forest and grassland; (2) cropland, including dry land and paddy fields; (3) built-up land, including urban areas, rural settlements and others such as industrial areas, roads, and airports; and (4) water bodies, including lakes, rivers, streams, reservoirs, ponds, and wetlands (Fig. 1). Bare land was not included in our analysis because its area in watersheds was very small. The percentages of land use types in each lake's watershed were calculated in ArcGIS 10 software. Cropland and built-up land can be considered as human-dominated land uses (HDL; Abdullah and Nakagoshi, 2006). We used the HDL here, because the correlations between in-lake characteristics (i.e., water quality and sediment characteristics) and the percentage of cropland or percentage of built-up land were weak.

To quantify the vegetation cover, we calculated the normalised difference vegetation index (NDVI) of each watershed from the Moderate-resolution Imaging Spectroradiometer (MODIS) red and near-infrared bands. The 250-m resolution

MODIS data for the years 2010 and 2011 was obtained from the USGS EROS Data Center. The NDVI is a reflection of the biophysical condition of a watershed's vegetation cover, which, in turn, may affect the water runoff and water quality (Griffith et al., 2002).

2.6 Statistical analyses

For each studied lake, we calculated the mean values for sediment denitrification, N_2O production, water quality, sediment characteristics and plant community structure (Table S2 and Table S3). Before statistical analyses, we tested data for normal distribution using a Shapiro–Wilk test. When possible, non-normally distributed data were natural log or square root transformed to reach a normal distribution. To examine the relationships among watershed land uses, water chemistry, sediment characteristics, plant community structure, sediment denitrification and N_2O production at the lake level, we performed Pearson correlation and regression analyses using PASW Statistics 18 software (IBM SPSS Inc., Chicago, USA).

Structural equation modelling (SEM) with Bayesian estimation was used to study the indirect effects of watershed human land use on lake sediment denitrification and N_2O production. We used results from our correlation analysis to select the promising explanatory variables to include in the SEM models. Given the positive correlations between TN and NO_3^- ($r = 0.86$, $P < 0.01$) and between TN and NH_4^+ ($r = 0.45$, $P < 0.05$), the TN was not included in the SEM analyses to simplify the final models. We expected that NO_3^- and NH_4^+ were likely correlated with DO, because when aerobic condition was present, NH_4^+ might easily be converted into

NO_3^- by nitrifying organisms in waters (Peterson et al., 2001). The software Amos 20 (IBM SPSS Inc., Chicago, USA) was used to design the SEM models and calculate path coefficients, squared multiple correlations, direct and indirect effects. Indirect effects, i.e., effects mediated by other variables, were calculated by multiplying the standardised path coefficients (i.e., estimates) of the direct effects of the variables involved in the total pathway. We used the chi-square (χ^2) test and the comparative fit index (CFI) to evaluate the overall fit of the SEM models. According to Kline (1998), a good fit was indicated by an insignificant χ^2 statistic ($P > 0.05$) and a CFI value > 0.90. The χ^2 provides an estimate of how closely the proposed SEM model matches the structure of the actual data. Thus, an insignificant χ2 statistic ($P > 0.05$) indicates that the structure of the proposed model is no different than the structure of the data.

3 RESULTS

3.1 Within-lake characteristics and watershed land uses

The highest TC and TN concentrations in water were found in Lake Makouhu (31.40 mg L^{-1}) and Lake Yiaihu (2.87 mg L^{-1}), respectively (Table S2). The mean TC and TN contents in sediments were 25 and 0.51 mg g^{-1}, respectively (Table 1). Submerged plants were only found in 5 lakes, with maximum biomass (2.43 kg m^{-2}) recorded in Lake Haikouhu (Table S2).

The percentage of HDL in watersheds varied from 34 % to 80 % with a mean of 61 % (Table 1). Among water quality and sediment characteristics, DO, NO_3^-, NH_4^+, TN and STN were significantly correlated with the percentage of HDL (Table 2). There was no significant relationship between plant community structure and watershed land uses (Table 2).

3.2 Denitrification and N_2O production

Potential denitrification rates varied widely across the 20 lakes and ranged from 11 ng N g^{-1} h^{-1} in Lake Chaohu to 125 ng N g^{-1} h^{-1} in Lake Makouhu (Table 1). Background denitrification rates were lowest in Lake Huangdahu at 0.67 µg N g^{-1} h^{-1}, and highest in Lake Linghu at 19 ng N g^{-1} h^{-1} (Table 1). N_2O production rates ranged from 0.05 to 0.71 ng N g^{-1} h^{-1}, with the largest value recorded in Lake Caizihu where the relative N_2O production was 0.13 (Table S3).

There was no significant relationship between potential denitrification rate and background denitrification rates (R = 0.09, P = 0.70), N_2O production rate (R = 0.22, P = 0.36) or relative N_2O production (R = 0.24, P = 0.31; figures not shown). N_2O production rates increased with increasing background denitrification rates (R^2 = 0.61, P < 0.01; Fig. 2).

3.3 Relationship of denitrification and N_2O production to within-lake characteristics

Denitrification and N_2O production rates were found to be significantly related to several water quality parameters (Table 3). However, sediment characteristics and plant community structure had no significant relationships with denitrification and N_2O production rates. Potential denitrification rate was negatively correlated with DO (Table 3). We determined that both background denitrification rate and N_2O production rate were enhanced at higher NO_3^- and TN concentrations and lower ORP and DO concentrations (Table 3).

3.4 Relationship of denitrification and N_2O production to watershed land uses

Watershed land uses were not significantly related to potential denitrification rates or relative N_2O production (R = 0.11, P = 0.61; figures not shown). However, background denitrification rate was found to be positively correlated with the percentage of HDL in watersheds ($R^2 = 0.29$, $P < 0.05$; Fig. 3A). We also found that N_2O production rate was significantly related to the percentage of water bodies in watersheds ($R^2 = 0.27$, $P < 0.05$; Fig. 3B).

3.5 Indirect effects of human land uses on denitrification and N_2O production

The results of the SEM analysis indicated that the fit of the data to the models was acceptable (Fig. 4). In the potential denitrification model ($\chi^2 = 4.272$, $df = 5$, $P = 0.511$, CFI = 1.000), the total indirect effect of watershed HDL on potential denitrification (0.23) was small and statistically insignificant (Table 4, Fig. 4A). In the background denitrification model ($\chi^2 = 2.846$, $df = 5$, $P = 0.724$, CFI = 1.000), almost all of the indirect effects of HDL on background denitrification (0.73) was mediated through water quality (principally via NO_3^-; Table 4, Fig. 4B). In both the N_2O production model ($\chi^2 = 4.818$, $df = 5$, $P = 0.439$, CFI = 1.000) and relative N_2O production model ($\chi^2 = 4.349$, $df = 5$, $P = 0.500$, CFI = 1.000), HDL indirect effects were largely transmitted via water quality (Table 4).

4 DISCUSSION

4.1 Effects of watershed human land use on in-lake characteristics

Numerous studies have revealed that human modification of the land uses plays a key role in determining the water quality of adjacent aquatic ecosystems (e.g., generation of point and non-point source pollutants and expansion of impervious surface area)(Crosbie and Chow-Fraser, 1999; Arbuckle and Downing, 2001; Taranu and Gregory-Eaves, 2008). In this study, human land use in watersheds was strongly and positively related to water N concentrations in lakes (Table 2). Built-up land, including urban, industrial areas and rural settlements, is the leading source of point source pollution in the Yangtze River basin (Liu et al., 2012). Chaohu Lake, one of the lakes included in the present study, received 2.17×10^{8} t of industrial wastewater and domestic sewage discharged from adjacent Hefei City every year (Shang and Shang, 2005). Cropland is recognised as the most important source of non-point source pollution affecting lakes in many countries. Approximately half of the area of the middle and lower Yangtze River basin is covered by agriculture land, especially paddy fields (Liu et al., 2011). In the Yangtze River basin, the annual consumption of nitrogen fertiliser on agricultural lands increased from 94 kg ha^{-1} in 1980 to 509 kg ha^{-1} in 2000 (Liu et al., 2008). Moreover, the nitrogen use efficiency decreased considerably from 0.31 to 0.23 between 1980 and 2000. Most of the excess nitrogen in paddy fields was discharged into aquatic environments through surface runoff.

Relatively few studies have reported on the relationship between watershed landscapes and lake sediment characteristics and their results are inconsistent (Müller et al., 1998; Bruesewitz et al., 2011). Bruesewitz et al. (2011) found that the percentage of catchment agriculture was positively related to sediment carbon but not nitrogen. These researchers found that agricultural landscapes can increase carbon inputs to aquatic ecosystems as a result of increased soil erosion and organic carbon transport or through increased lake productivity resulting in higher rates of organic matter deposition in sediments. However, Müller et al. (1998) indicated that TN concentration in lake sediments strongly increased with increasing agricultural land use in watersheds. Our study also found a significant relationship between watershed human landscapes and STN. It is noteworthy that the correlation between human landscapes and water TN was greater than that between human land uses and STN (Table 2). One possible explanation is that water TN level in lakes primarily reflects recent land use patterns in watersheds. Sediment TN content, however, reflects sedimentation and deposition of nutrients that have occurred over several decades. Any modelling of water and sediment nutrient contents using recent land use data is likely to show stronger correlations with water than sediment nutrient levels (Houlahan and Findlay, 2004).

4.2 The role of sediment denitrification in nitrogen removal in Yangtze lakes

Sediment denitrification is recognised as the most important pathway of nitrogen removal in lakes, followed by nitrogen sedimentation and uptake by aquatic macrophytes (Saunders and Kalff, 2001). Scaled on an areal basis using the bulk density of the top 7 cm of sediment, background denitrification rate ranged from 353.6 to 1.097×10^4 kg N km^{-2} year^{-1}, with a mean of 1065 kg N km^{-2} year^{-1}. This mean rate is smaller than the global average of lakes (3421 kg N km^{-2} year^{-1}) estimated by a NiRReLa model (Harrison et al., 2009). Background denitrification rate can be considered as a conservative estimate of actual in situ denitrification. Therefore, according to the areal background denitrification rate and the surface area of the 20 studied Yangtze lakes (Table S1), we find that Lake Chaohu and Lake Caizihu can at least remove 6.019×10^5 and 5.538×10^5 kg of N every year, respectively. It has been reported that denitrification rate of lake sediments shows a marked seasonal variation with higher values in summer and lower values in winter (Christensen and

Sørensen, 1986). Therefore, further studies are needed to investigate the temporal variations of sediment denitrification in the Yangtze River basin to estimate the N removal capacity of lake sediments more accurately.

Net N_2O production during sediment N cycles is gaining increased attention because N_2O is a powerful greenhouse gas (Hefting et al., 2006). The mean net N_2O production rate (i.e., N_2O production in the absence of acetylene) of the studied Yangtze lakes was 0.27 ng N g^{-1} h^{-1} or 142.9 kg N km^{-2} $year^{-1}$. This value is approximately 2.1 times greater than the average rate in Norwegian lakes (68.60 kg N km^{-2} $year^{-1}$) calculated by McCrackin and Elser (2010). By using the static chamber method, Wang et al. (2007) reported that the N_2O flux in the pelagic zones of a eutrophic Yangtze lake (i.e., Lake Taihu) was approximately 182.2 kg N km^{-2} $year^{-1}$. It should be noted that N_2O production in the absence of acetylene may be derived from both denitrification and nitrification processes (García-Ruiz et al., 1998; Xu et al., 2008). Nitrification, the oxidation of NH_4^+ to NO_3^-, can produce N_2O as an intermediate product. Beaulieu et al. (2014) found that denitrification and nitrification were likely occurring simultaneously when dissolved oxygen in water column was > 5 mg/L. (Beaulieu et al., 2014).

In the present study, N_2O production rates significantly increased with increasing background denitrification rates (Fig. 2). Although our study did not quantify the respective contributions of denitrification and nitrification to N_2O production, the positive relationship between N_2O production and background denitrification rates might imply that sediment denitrification produces more N_2O than nitrification in the Yangtze lakes. Our result also indicates that lake sediments may be significant sinks for terrestrial NO_3^- but may also become a significant source of N_2O if NO_3^- input is to increase. However, the available information on the N_2O production and consumption of the Yangtze lakes is still rather limited (Wang et al. 2007). Further direct measurements of N_2O emission are needed to better understand the contribution of Yangtze lakes to regional N_2O fluxes.

The present study found that the average of relative N_2O production was 0.17, similar to the value (0.18) reported by Garcia-Ruiz et al (1998). However, our results contrast with those found in other investigations, the majority of which give values less than 0.05 (e.g., Seitzinger, 1988; Beaulieu et al., 2011). This result might be explained by the fact that oxygen in overlying water of shallow lakes could suppress the N_2O reductase enzyme, which reduces N_2O to N_2 during denitrification (Garcia-Ruiz et al., 1998). It is known that this enzyme is the most oxygen sensitive of all enzymes involved in denitrification process, and slurries in the present study were made with unfiltered lake water. It should be noted that some studies have found that the relative N_2O production can be larger than 1 and explained that in such cases nitrification, but not denitrification, is the major source of N_2O (Xu et al., 2008).

4.3 Local and regional determinants of sediment denitrification in lakes

Environmental variables that affect sediment denitrification can be categorised as proximal or distal regulators (Saggar et al., 2013). Proximal regulators, including NO_3^- concentration, oxygen supply or water content, carbon availability and temperature in sediment and overlying water, affect the instantaneous rate of sediment denitrification. Consistent with previous studies conducted in streams and rivers (García-Ruiz et al., 1998; Inwood et al., 2005), we found that only overlying water quality had significant relationships with sediment denitrification and N_2O

production in Yangtze lakes. Our study suggests that nitrogen availability in water is the primary factor limiting sediment denitrification in Yangtze lakes. Water NO_3^- concentration explained approximately 70 % of the variability in denitrification rates in a meta-analysis (Piña-Ochoa and Álvarez-Cobelas, 2006). Some studies also reported a significant relationship between denitrification and sediment carbon concentration (e.g., Bruesewitz et al., 2011). However, this relationship is not significant in the Yangtze lakes, most likely because sediment TC concentration was high (25 mg g^{-1}) and was not limiting to sediment denitrification.

Distal regulators affect sediment denitrification indirectly by acting on the proximal controls (Saggar et al., 2013). These regulators include factors such as catchment land use and regional climate. Few studies have examined the relationships between watershed human land uses and sediment denitrification in lakes (but see Bruesewitz et al., 2011; Liu et al. 2015). Bruesewitz et al. (2011) found that potential denitrification but not background denitrification was positively related to the proportion of catchment agriculture. Similarly, Liu et al. (2015) reported that the proportion of agricultural and urban land uses in watersheds was positively related to potential denitrification rate, but not to background denitrification rate and N_2O production rate. However, consistent with previous observations regarding stream sediment denitrification (e.g., Arango and Tank, 2008), we only found a significant positive relationship between sediment background denitrification and human land uses.

Although many studies have investigated the sediment denitrification in lakes worldwide (e.g., Hasegawa and Okino, 2004; Bruesewitz et al. 2011), to our knowledge, no study to date has examined the indirect effects of catchment land uses on sediment denitrification in lakes. Using a conceptual model, Inwood et al. (2007) predicted that land use could indirectly affect sediment denitrification and N_2O emission in headwater streams in Michigan, USA by influencing the stream water quality or sediment characteristics. SEM, a comprehensive statistical approach which allows for testing and estimating indirect effects among a set of associated variables, is widely used in ecological studies in recent years (e.g., Sutton-Grier et al., 2010). As we hypothesised, although both lake water quality and sediment characteristics were significantly affected by human land use, the indirect effects of human land use in watersheds on sediment denitrification and N_2O production in Yangtze lakes were more likely driven through changes in lake water quality than through changes in sediment characteristics (Table 4). SEM analysis further demonstrated that positive relationships between human land uses and sediment background denitrification were primarily driven by water nitrogen loading (Fig. 3A, Table 4). Areas of urban in the Yangtze River basin have expanded dramatically in the last three decades, and this change is considered a severe threat to lake ecosystems (Liu et al., 2012). Although our results suggest that human-dominated land uses can enhance the sediment denitrification in lakes, the quantity of nitrogen exported from watersheds should be controlled to improve the water quality and reduce the N_2O production of eutrophic lakes in the Yangtze River basin.

5 CONCLUSIONS

Increased human land uses in watersheds have led to elevated nitrogen concentration in both water and sediments in Yangtze lakes. Among the water quality parameters, ORP, DO, NO_3^- and TN were significantly related to sediment background denitrification and N_2O production rates. Only the background denitrification rate was found to be positively correlated with the percentage of human land uses in the watersheds. The results of SEM analysis supported the hypothesis that sediment denitrification and N_2O production in the Yangtze lakes were indirectly associated with watershed human landscapes, primarily through the effect of land use on lake water quality, but not sediment characteristics.

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Table 1 Summary statistics for sediment denitrification variables, within-lake characteristics and watershed land uses (N = 20).

	Unit	Min.	Max.	Mean	S.E.
Denitrification variables					
Potential denitrification rate	ng N g^{-1} h^{-1}	11	125	47	6.5
Background denitrification rate	ng N g^{-1} h^{-1}	0.67	19	3.0	0.90
N_2O production rate	ng N g^{-1} h^{-1}	0.05	0.71	0.27	0.04
Relative N_2O production		0.02	0.62	0.17	0.03
Water quality					
Cond	µS cm^{-1}	128	360	253	17
ORP	mv	-81	153	101	12
DO	mg L^{-1}	4.5	8.1	6.0	0.27
NH_4^+	mg L^{-1}	0.020	0.50	0.19	0.025
NO_3^-	mg L^{-1}	0.090	1.5	0.53	0.077
TN	mg L^{-1}	0.31	2.9	1.1	0.17
TC	mg L^{-1}	8.5	31	19	1.3
TOC	mg L^{-1}	2.5	6.6	4.2	0.29
Sediment characteristics					
pH		5.5	8.2	7.2	0.18
Bulk density	g cm^{-3}	0.64	0.94	0.86	0.014
Moisture	%	27	71	61	2.1
STN	mg g^{-1}	0.18	1.7	0.51	0.10
STC	mg g^{-1}	5.4	54	25	2.9
Plant community structure					
Species richness		0.00	1.0	0.15	0.072
Fresh biomass	kg m^{-2}	0.00	2.4	0.47	0.19
Watershed land uses					
Percent of vegetation	%	1.4	56	22	3.2
Percent of water body	%	8.9	43	17	1.9
Percent of HDL	%	34	80	61	2.5
NDVI		0.34	0.58	0.47	0.016

HDL: Human-dominated land uses;

NDVI: Normalised difference vegetation index.

Table 2 Pearson correlation coefficients between watershed land uses and within-lake characteristics (*N* = 20).

	Watershed land uses			
	Vegetation (%)	Water body (%)	HDL (%)	NDVI
Water quality				
Cond	-0.27	0.17	0.33	-0.49[a]
ORP	0.07	0.07	-0.11	0.02
DO	0.13	0.30	-0.45[a]	-0.11
NH_4^+	-0.61[b]	0.38	0.59[b]	-0.48[a]
NO_3^-	-0.46[a]	-0.11	0.72[b]	-0.18
TN	-0.47[a]	-0.13	0.75[b]	-0.17
TC	-0.17	0.26	0.08	-0.31
TOC	-0.34	0.10	0.38	-0.09
Sediment characteristics				
pH	-0.02	0.50[a]	-0.34	-0.48[a]
Bulk density	-0.06	0.17	-0.03	0.01
Moisture	0.11	0.10	-0.15	0.10
STN	-0.61[b]	0.47[a]	0.46[a]	-0.30
STC	0.03	0.06	-0.06	-0.14
Plant community structure				
Species richness	-0.01	0.34	-0.31	-0.18
Fresh biomass	0.09	0.12	-0.26	-0.01

[a] Significant at the 0.05 level;

[b] Significant at the 0.01 level.

HDL: Human-dominated land uses;

NDVI: Normalised difference vegetation index.

Table 3 Pearson correlation coefficients between sediment denitrification variables and within-lake characteristics (*N* = 20).

	Potential denitrification rate	Background denitrification rate	N_2O production rate	Relative N_2O production
Water quality				
Cond	0.32	-0.01	0.01	0.04
ORP	-0.10	-0.60[b]	-0.50[a]	0.37
DO	-0.48[a]	-0.52[a]	-0.56[a]	0.15
NH_4^+	0.05	0.38	0.03	-0.45[a]
NO_3^-	0.34	0.79[b]	0.62[a]	-0.32
TN	0.23	0.72[b]	0.56[a]	-0.33
TC	0.36	-0.02	0.12	0.32
TOC	0.32	0.16	0.29	0.14
Sediment characteristics				
pH	-0.01	-0.31	-0.35	0.26
Bulk density	0.30	0.16	0.12	-0.05
Moisture	0.39	0.01	0.10	0.11
STN	0.18	0.30	-0.11	-0.36
STC	0.38	-0.07	-0.08	0.14
Plant community structure				
Species richness	-0.28	-0.34	-0.33	0.10
Fresh biomass	-0.32	-0.17	-0.10	0.05

[a] Significant at the 0.05 level;

[b] Significant at the 0.01 level.

Table 4 Indirect effects of human-dominated land uses on sediment denitrification in lakes mediated by lake water quality and sediment characteristics (*N* = 20).

	Potential denitrification rate		Background denitrification rate		N_2O production rate		Relative N_2O production	
	Indirect effects	Contribution (%)	Indirect effects	Contribution (%)	Indirect effects	Contribution (%)	Indirect effects	Contribution (%)
Water quality								
NH_4^+	0.00	0	-0.01	1	-0.01	1	0.00	0
DO	0.21	74	0.15	20	0.22	27	-0.02	5
NO_3^-	-0.03	10	0.58	78	0.43	52	-0.21	58
Sediment characteristics								
STN	0.05	16	0.01	1	-0.16	20	-0.13	36
Total Indirect effects	0.23	100	0.73	100	0.48	100	-0.35	100

Note: Indirect effects were calculated by multiplying the standardized path coefficients of the direct effects of the variables involved in the total pathway. Percentage of contribution was calculated using the sum of absolute values for contributing effects.

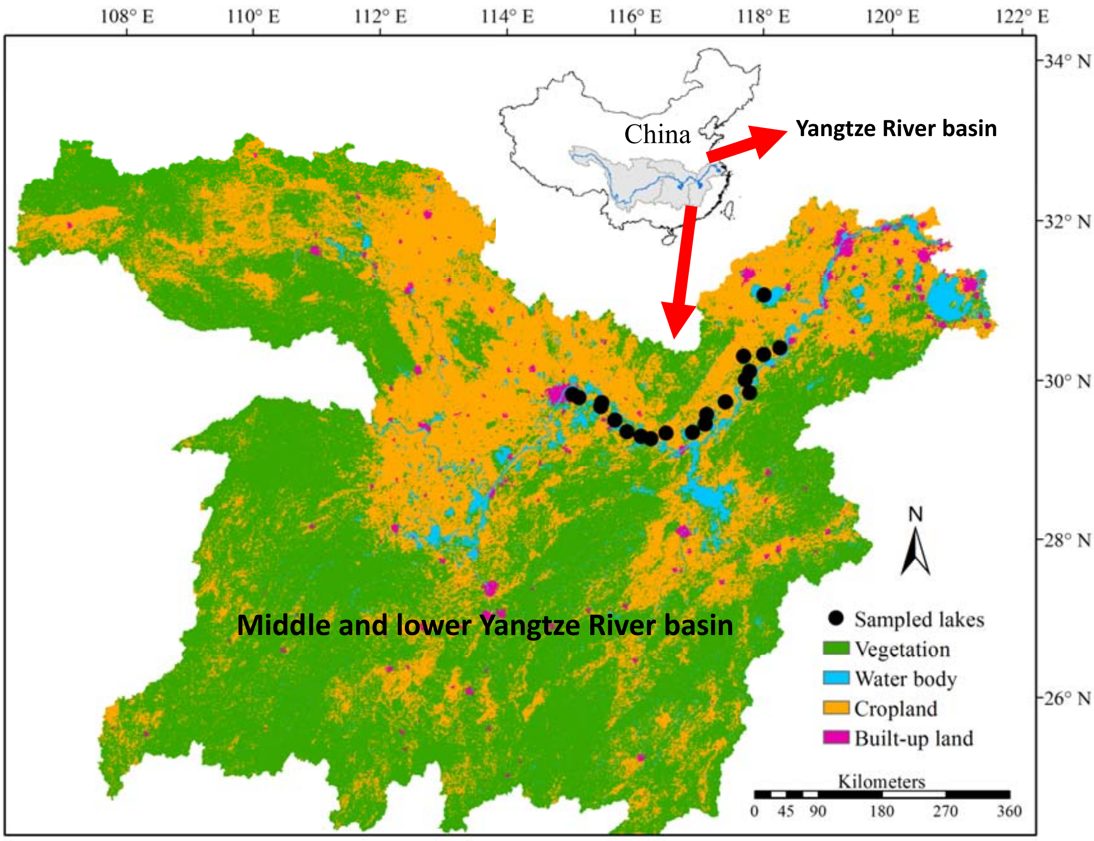

Figure 1. Locations of the Yangtze River basin and 20 studied lakes in China.

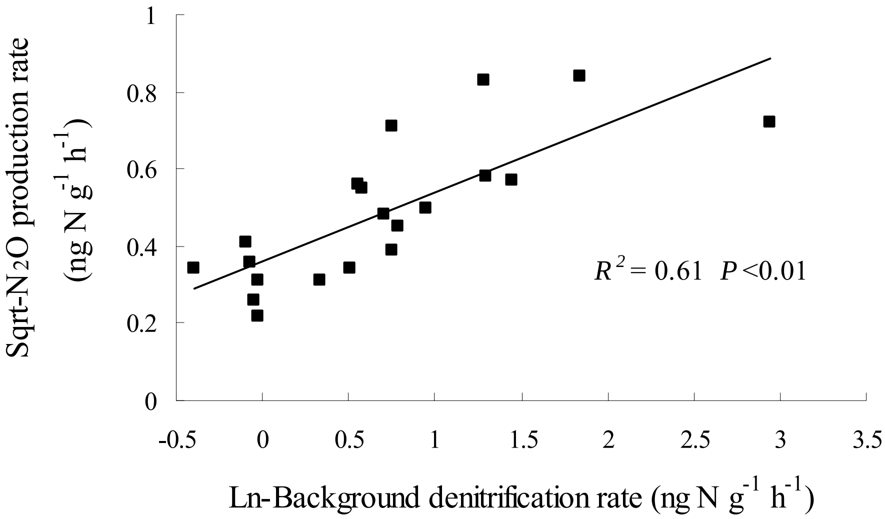

Figure 2. Relationships between background denitrification rate and N_2O production rate of the Yangtze lakes (N = 20).

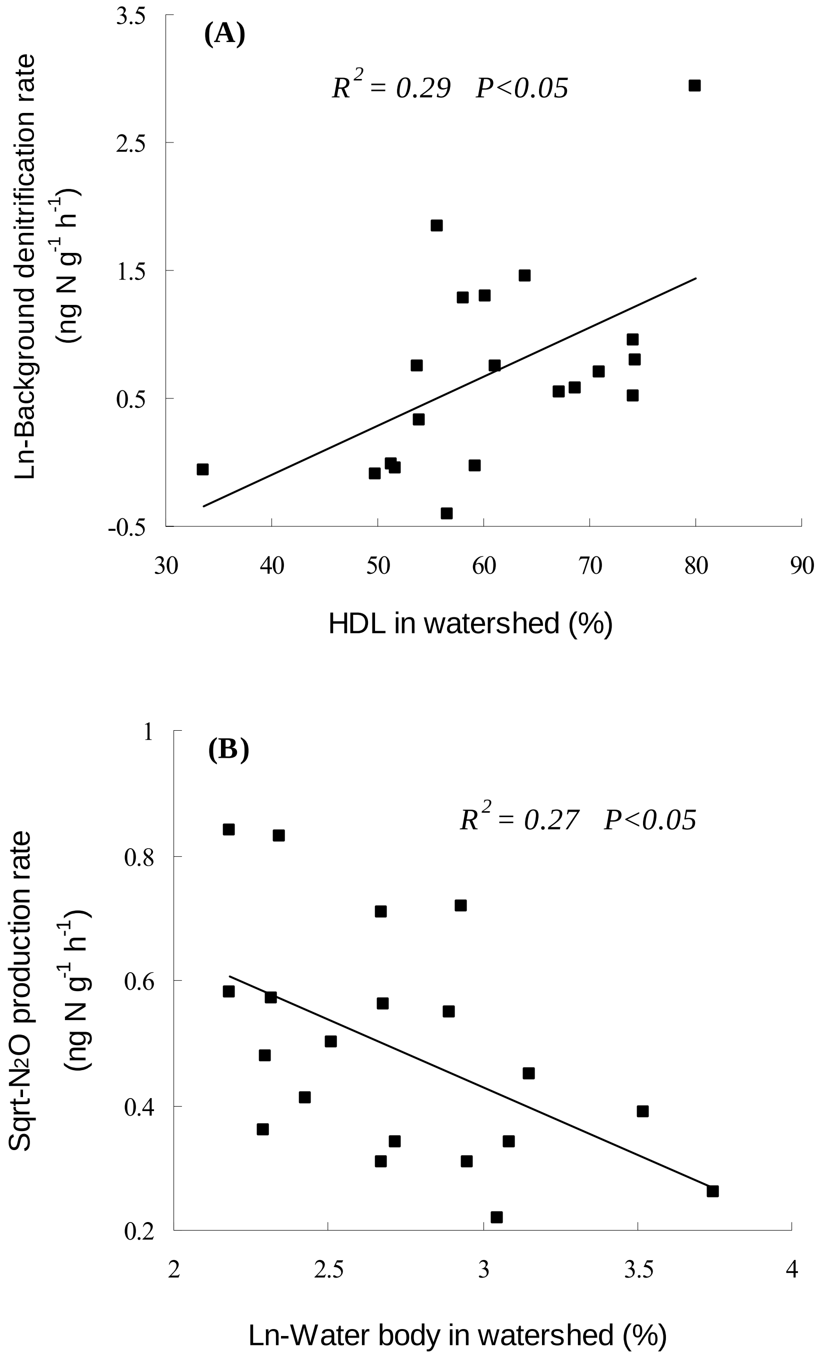

Figure 3. Relationships between human-dominated land uses (HDL) in watershed and sediment background denitrification rate (A) and between water body in watershed and N_2O production rate (B) of the Yangtze lakes (*N* = 20).

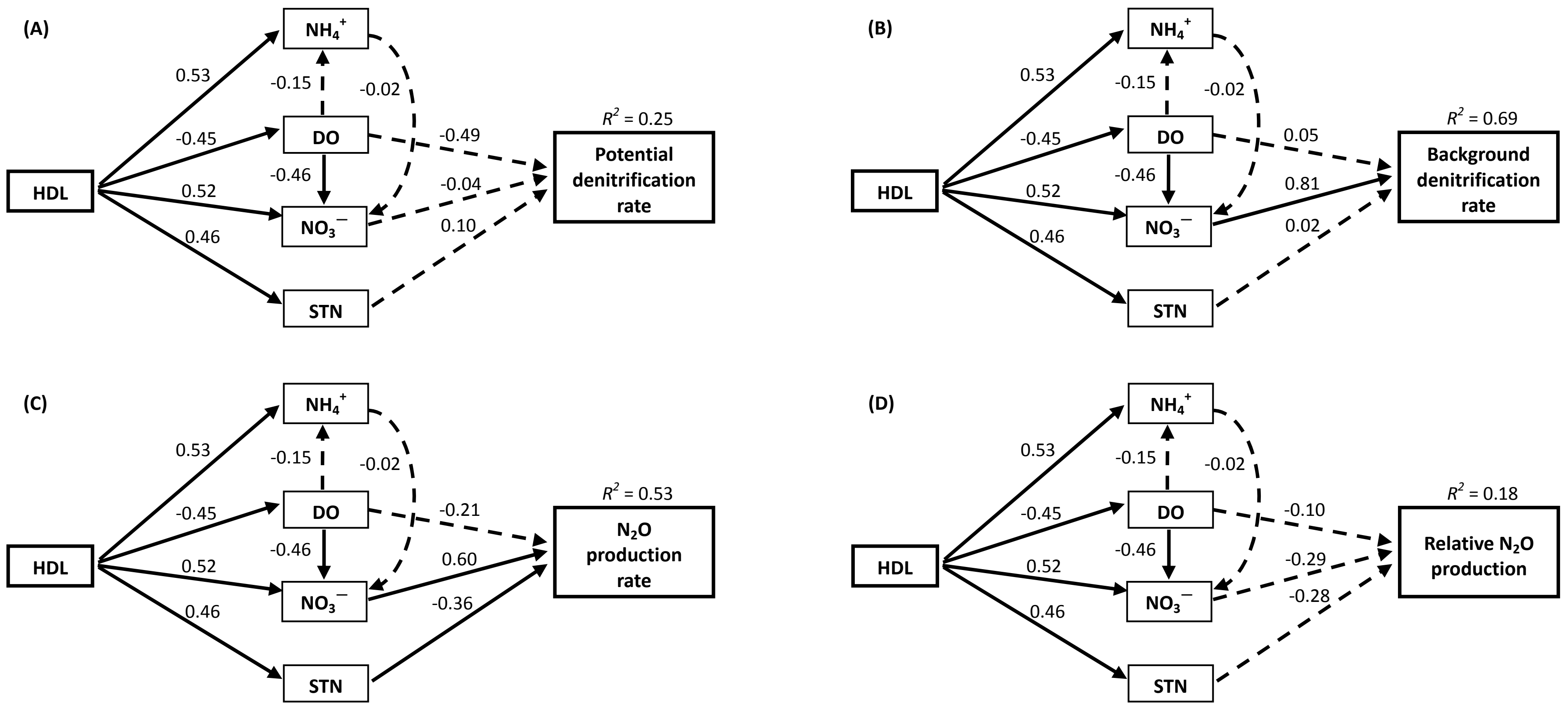

Figure 4. Structural equation models depicting the indirect effects of human-dominated land uses (HDL) in watersheds on the sediment potential denitrification rate (A), background denitrification rate (B), N_2O production rate (C) and relative N_2O production (D) of the Yangtze lakes (N = 20). Solid lines indicate significant ($P < 0.05$) positive or negative effects, and dashed lines indicate insignificant positive or negative effects. Numbers adjacent to the lines are standardised path coefficients.