

Effects of sediment dredging on nitrogen cycling in Lake Taihu, China: Insight from mass balance based on a 2-year field study

Juhua Yu^{1,2} · Chengxin Fan¹ · Jicheng Zhong¹ · Lu Zhang¹ ·
Lei Zhang¹ · Changhui Wang¹ · Xiaolong Yao¹

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Abstract Sediment dredging can permanently remove pollutants from an aquatic ecosystem, which is considered an effective approach to aquatic ecosystem restoration. In this work, a 2-year field simulation test was carried out to investigate the effect of dredging on nitrogen cycling across the sediment-water interface (SWI) in Lake Taihu, China. The results showed that simulated dredging applied to an area rich in total organic carbon (TOC) and total nitrogen (TN) slightly reduced the NH_4^+ -N release from sediments while temporarily enhanced the NH_4^+ -N release in an area with lower TOC and/or TN (in the first 180 days), although the application had a limited effect on the fluxes of NO_2^- -N and NO_3^- -N in both areas. Further analysis indicated that dredging induced decreases in nitrification, denitrification, and anaerobic ammonium oxidation (anammox) in sediments, notably by 76.9, 49.0, and 89.9 %, respectively, in the TOC and/or TN-rich area. Therefore, dredging slowed down nitrogen cycling rates in sediments but did not increase N loading to overlying water. The main reason for the above phenomenon could be attributed to the removal of the surface sediments enriched with more TOC and/or TN (compared with the bottom sediments).

Overall, to minimize internal N pollution, dredging may be more applicable to nutrient-rich sediments.

Keywords Sediment dredging · Nitrogen cycling · Nitrification · Denitrification · Anaerobic ammonium oxidation (anammox) · Lake Taihu

Introduction

Eutrophication has caused a number of threats to freshwater ecosystems and has become the most prominent problem of lakes all over the world (Qin and Zhu 2006; Fan et al. 2004; Jin 2003; Smith et al. 1999). Many approaches have been applied during the past decades to improve lake water quality (Pan et al. 2012; Gustavson et al. 2008; Yin et al. 2013; Wang et al. 2014; 2015). Sediment dredging is used as a significant eco-environmental engineering method to permanently remove surface-contaminated sediments from aquatic ecosystems. However, the effectiveness of sediment dredging is still debatable because it may not be as ideal as people have expected for unclear reasons (Fan et al. 2004; Pu et al. 2000). At the same time, there are also questions concerning the efficacy of sediment dredging and the degree to which dredging results in reduced risk to biological health and the aquatic environment. In such cases, numerous studies have examined the biological effects of dredging in lakes, which mainly focused on razor fish community characterization (Tuck et al. 2000), scallops (Maguire et al. 2002), benthos and algal-periphyton community composition (Lewis et al. 2001), and the restoration of the benthic habitat (Szymelfenig et al. 2006). To better understand the environmental effectiveness and to elaborate on mechanisms of pollutant removal by dredging, much attention has been paid to the influence of dredging on changes in sediment resuspension and transport processes (Yu et al.

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✉ Jicheng Zhong
jczhong@niglas.ac.cn

¹ State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Number 73 Beijing East Road, 210008 Nanjing, People's Republic of China

² University of Chinese Academy of Sciences, Beijing 100049, China

2012), the release of persistent organic pollutants (POPs) and heavy metals following the removal of contaminated sediments (Bremle and Larsson 1998; Bowman et al. 2003; Van den berg et al. 2001), and the long term effectiveness of dredging on internal contaminant control (Kleeberg and Kohl 1999; Reddy et al. 2007).

Generally, dredging is primarily applied to remove the POPs, heavy metals, and phosphorus in eutrophic lakes (Kleeberg and Kohl 1999; Reddy et al. 2007; Zhong et al. 2008; Yenilmez and Aksoy 2013; Recknagel et al. 1996). Nitrogen in eutrophic lakes is typically one of the limiting nutrients; however, the effectiveness of dredging on nitrogen removal is unknown. In terms of the methodology, most field studies (Fan et al. 2004; Wang and Feng 2007) and laboratory simulations (Spencer et al. 2006; Zhong et al. 2009) have been deployed by monitoring nitrogen content in pore water and overlying water to evaluate the short-term and long-term effects of nitrogen removal after dredging based on flux calculation or instantaneous measurements alone (Fan et al. 2004; Wang and Feng 2007). The results are usually contradictory among various studies (Fan et al. 2004; Yu et al. 2012; Lohrer and Wetz 2003), and these discrepancies may be attributed, to a great extent, to the implications of pollution status of the sediments in which dredging is implemented. Thus, it is not possible to definitively conclude whether variations in water quality of lakes are natural in origin or are a result of dredging practices under different pollution statuses of sediments (Gustavson et al. 2008).

The cycling of nitrogen in sediments is driven by the rate and lability of organic matter deposition, sediment accumulation rates and bioturbation/bioadvection, and rates of microbial nitrogen transformation (Cornwell and Owens 2011). Dredging is one type of large-scale, anthropogenic disturbance associated with high costs, and it often destroys the balance of the sediment-water interface, including the physical environment (soil texture, particulate deposition, suspension, and advection), chemical processes (mineralization, adsorption, and desorption), and biological processes (bioturbation, bioirrigation, and excretion) (Pu et al. 2000; Qin et al. 2004). Taken together, these processes must inevitably influence nutrient cycling processes, reaction rates, diagenetic properties, transport in surface sediments, and nutrient exchange across the sediment-water interface (SWI) (Falcão et al. 2003; Yu et al. 2012; Qin et al. 2004). To date, there is little information available concerning the effect of dredging on micro-interface nitrogen cycling. The study published by Jing et al. (Jing et al. 2013) measured the variations of nitrogen fractions in overlying water from undredged, fresh-dredged, and post-dredged sediments to evaluate the effects of dredging on nitrogen cycling across the SWI based on nitrogen diffusive flux from sediment to overlying water. Cornwell and Owens (2011) used sediment pore water NH_4^+ -N chemistry, adsorbed NH_4^+ -N concentrations, sediment-water NH_4^+ -N exchange,

and N_2 -N flux to quantify the mass of labile N that can be released during dredging in the Chesapeake Bay. However, published studies have not addressed the elaborate mechanisms of nitrogen removal processes and furthermore have not distinguished the nitrogen release induced by dredging from the environmental effects of natural processes and have not quantified the nitrogen releases under the condition of overall N cycling in aquatic ecosystems. Only fragmentary studies conducted by Zhong et al. (2010a, b) and Graca et al. (2004) noted that dredging can reduce the denitrification rates by changing the pattern of inherent cycling of nitrogen. However, the basic microbial processes to which nitrogen is subjected in the sediment are denitrification, nitrification, ammonification, anammox, and dissimilatory NO_3^- -N reduction to ammonium (DNRA), of which the denitrification, anammox, and DNRA processes can permanently remove nitrogen from aquatic ecosystems. In addition, denitrification seems to be the most significant process for removing nitrogen from freshwater bodies (Liu et al. 2015; Hanaki et al. 1990; Zhong et al. 2010a, b; Trimmer et al. 2003). Therefore, it is important to identify the specific microbial processes of nitrogen release induced by dredging under different pollution states of sediments in aquatic ecosystem restoration.

After dredging, we hypothesized that N cycling would be slowed down, particularly the dissimilatory microbial processes of nitrogen removal, owing to lower values of TOC and/or TN and of microbe abundance. We further hypothesized that the NO_3^- -N and NO_2^- -N removal from aquatic ecosystem would be weakened because the de nitrogen processes were inhibited substantially. Therefore, the main objectives of this study were first to determine the effect of dredging on inorganic nitrogen releases from sediments across the SWI; second, to construct rough budgets for the nitrogen removal induced by dredging on the basis of nitrification, denitrification, and anammox; and third, to further understand the implications of dredging projects to nitrogen mass balance under different pollution states of sediments based on the results of the first two objectives. To better interpret the effects of dredging on nitrogen cycling, this in situ microcosm study was deployed in the lake for 2 years.

Methods and materials

Study site

Lake Taihu is the third largest freshwater lake located in the southern part of the Delta of the Yangtze River in China (Fig. 1). Since the mid-1980s, Microcystis algal blooms have occurred every summer in the northern part of the lake, with increasing severity in recent years. Subsequently, drinking water supply has often been limited, which has caused massive economic loss (Qin et al. 2006). Some dredging projects

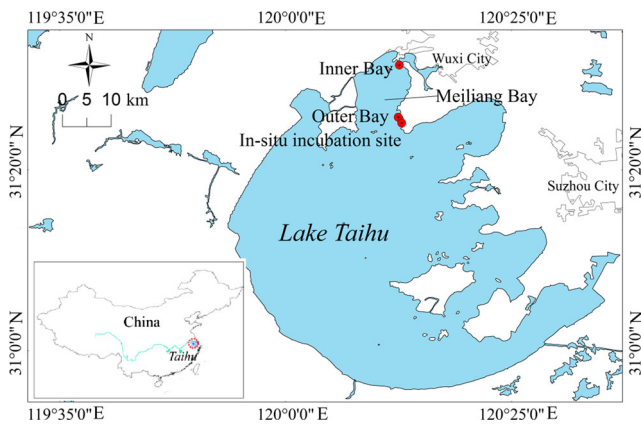


Fig. 1 The sampling sites and in situ incubation location

have been conducted starting in 2009, and many more dredging plans are being developed or will be put into practice for pollution control of Lake Taihu. Meiliang Bay is located at the north of Lake Taihu with an area of 130 km². The water at the study site has vital biological functions, not only as landscape water used for navigational purposes and fishing but also as a source of drinking water for the city of Wuxi. Two stations, in the inner bay and the outer bay, were selected considering the pollution state characteristics of Meiliang Bay. The inner bay (31° 31' 33.9" N, 120° 12' 35.2" E) is enriched with TOC, TN, and TP in sediments by receiving a greater proportion of municipal and industrial waste water from the Liangxihe River (yearly average values of overlying water NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N were 3.92, 0.66, and 0.12 mg L⁻¹ from January 2006 to January 2007, respectively), and the TOC and TN content were much greater than those of paddy soil in the Taihu region (ranging from 0.18 to 2.8 % and 0.047 to 0.093 % of TOC and TN) (Xu et al. 1980). The outer bay (31° 25' 49.3" N, 120° 12' 27.7" E) is situated at the mouth of the Meiliang Bay with a distance of 1 km away from the Taihu Laboratory for Lake Ecosystem Research (TLLER) (31° 25' 10.78" N, 120° 12' 50.7" E), with less TOC, TN, and TP than the inner bay, and with TOC and TN contents less than those of the paddy soil of the Taihu region (Xu et al. 1980; Zhong et al. 2010a, b).

Experimental set-up of in situ dredging simulation study

A total of 30 intact sediment cores were collected with a large caliber core sampler (Φ110 mm, length 500 mm, Rigo Co., Japan) at either site. To decrease the heterogeneity of sediments, the cores were systematically collected in a grid within the 2×2 m square. The cores collected were generally more than 400 mm deep, and they were filled up with near-bottom water from the same location and closed with rubber stoppers to avoid sediment oxidation during sampling and transportation. With this sampling procedure, the surfaces of sediment core remained undisturbed. Simultaneously, some subsamples

of surface sediments (0–2 cm) were rapidly sliced onboard within a few minutes to minimize changes resulting from exposure of sediment surfaces to the atmosphere. Overlying water samples were also collected at each location using 25 L polyethylene bottles.

In the laboratory, sediment cores from either site were treated as follows. The overlying water of all sediment cores was siphoned. The simulating dredging depth was 25 cm sediment based on practical dredging projects in Lake Taihu, and the sediment height used in the study was approximately 12 cm. Subsequently, the surface sediments of three cores for each site were removed by suction pump, and the underlying 12 cm layer of sediment was filled into the tubes undisturbed as the dredged treatments. As for the control treatment, the top 12 cm sediment layers of three cores for each site were cut and carefully transplanted into the Plexiglas cylinders (11 cm inner diameter, 17 cm height). A total of 48 cores were selected to determine the rates of nitrification, denitrification, and anammox over the course of 1 year, and sampled in October 2011, and January, April, and July 2012. The overlying water was carefully dropped onto the sediment cores immediately. To better mimic the environmental conditions in the lake, undredged cores and dredged cores were incubated on the lake bottom, situated at the observation platform of the TLLER. All prepared cores were put into the stainless shelf with six units for each shelf. In addition, each core was covered completely with fine mesh (ca. 0.0374 mm) nylon screening to restrict access of large aquatic fauna. Then, the shelf was fastened to the bridge of the observation platform using nylon line, which was put down to the sediment surface of the lake (ca. 1.5 m). In the incubation period from August 2011 to July 2013, sampling was carried out six times (at 3, 30, 90, 180, 360, and 720 days after dredging). At the end of the experiments, the sediment cores were sliced at 1 cm intervals, and the sediment subsamples were freeze-dried and sieved through 100 mesh sieves for further analysis.

Collection of pore water

The pore water sample was collected by using a nondestructive method to portray the nutrient profiles in interstitial water at different dredging times. Rhizon soil moisture samplers (Rhizon SMS: Rhizosphere Research Products, the Netherlands) were used to extract successive pore water samples from the microcosm sediments at depths of 0.5, 1.5, 2.5, 3.5, 5, 7, and 9 cm. Holes for Rhizon SMS Samplers in the Plexiglass tube walls were sealed in advance with hydrophobic tape. Owing to the pore water samples separated using negative pressure extraction, the overlying water in the sediment cores were extracted using a syringe before sampling to avoid the diffusion of overlying water into the interstitial water under negative pressure. At each site, approximately 5 mL of pore water was obtained through suction and was filtered in

the field by 0.45 μm aperture GF/F filter membranes. Then, the pretreated samples were immediately transported to the laboratory in ice boxes for determination of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and $\text{NO}_2^-\text{-N}$ using a Skalar flow-injection analyzer (Skalar Sanplus, the Netherlands).

Nitrification potential rate measurement

The potential nitrification rate of sediments was measured by using the modified method (Dollhopf et al. 2005; Hall, 1984) with the specific inhibitor of nitrification, allylthiourea (ATU), which blocks the $\text{NH}_4^+\text{-N}$ to the NH_2OH step of ammonium oxidation to ensure that nitrification does not occur. Each subsample of surface sediment was sampled and homogenized, and one aliquot of sediment (ca. 5 g of wet sediment) was placed into 250 mL Erlenmeyer flasks. Each treatment had three replicates, and the known amount of $\text{NH}_4^+\text{-N}$ was added as $(\text{NH}_4)_2\text{SO}_4$ to obtain the initial $\text{NH}_4^+\text{-N}$ concentration of 14 mg L^{-1} ($1000\ \mu\text{mol L}^{-1}$). The ATU was added to the flasks of the inhibited group to attain a final ATU concentration of 10 mg L^{-1} , and the uninhibited group only contained the $(\text{NH}_4)_2\text{SO}_4$ solution. Subsequently, all of the flasks were capped with gauze to exclude external bacteria, then covered with aluminum foil and incubated in a shaker incubator at $25\text{ }^\circ\text{C}$. There were five sampling times carried out at 0, 6, 12, 18, and 24 h during the whole incubation. After incubation, the overlying water of each flask was filtered through previously ignited and washed Whatman GF/F filters, and the $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ concentrations were further determined. Potential nitrification rates were calculated according to the $\text{NH}_4^+\text{-N}$ concentration differences between the inhibited and uninhibited flasks as follows:

$$NP = \frac{\rho h (V_1 + V_2) (K_1 - K_0)}{1000} w \quad (1)$$

where NP is the potential nitrification rate ($\text{mmol m}^{-2}\text{ h}^{-1}$), ρ is the bulk density of sediment (kg m^{-3}), h is the sampling depth of sediment (m), and V_1 and V_2 are solution volume and pore water of sediment (L), respectively, k_1 and k_0 represent the variations of $\text{NH}_4^+\text{-N}$ between the inhibited and uninhibited group with time ($\mu\text{mol L}^{-1}\text{ h}^{-1}$), and w is the dry weight of sediment used in incubation.

Simultaneous determination of denitrification and anammox

The system described by Nishio et al. (1982) and Usui et al. (2001) was used to simultaneously measure denitrification and anammox with some modifications. Specifically, the intact sediment cores were capped with a Plexiglas lid with an O-ring to avoid leaving headspace in the cores, were wrapped with aluminum foil to keep light out, and were placed in a

temperature-controlled incubation water bath under in situ temperature. The continuous flow-through chamber setup consisted of an intake water vessel, Teflon flow tubes, a peristaltic pump, and a sample collection vessel. The influent water was sampled from in situ near-bottom lake water, of which the concentration was adjusted to $100\ \mu\text{mol L}^{-1}$ using $^{15}\text{NO}_3^-\text{-N}$ (final % ^{15}N ca.90–99 %, depending on the background nitrate concentration). Then, the cores were incubated under in situ conditions for approximately 24 h to achieve an equilibrium state. During the course of incubation, the intake water was pumped into the chamber using a peristaltic pump. The inlet vessel was lower than that of the output vessel, which was located at the position approximately 1 cm above the SWI. Resulting from horizontal and vertical flow in cores, the intake water could be mixed completely. As for field conditions, it was crucial to select the appropriate water flow used in the experiment mainly based on the oxygen consumption of the overlying water and the characteristics of SWI. In this study, according to the monthly field observation in Lake Taihu (Peter et al. 2005), to avoid air introduction to the mixing chamber, total flow rates of waters into the mixing chamber were set at 1 mL L^{-1} . Oxygen saturation was retained ranging from 7 to 29 %, with a mean value of 16 % (7.62 mg L^{-1} inlet water, 5.40 mg L^{-1} output water), and oxygen concentration in the influent and effluent were determined simultaneously. Incubation temperature was set at $15\text{ }^\circ\text{C}$ in spring and autumn, $30\text{ }^\circ\text{C}$ in summer, and $5\text{ }^\circ\text{C}$ in winter. The experiments were continued for 24 h, and the influent water and overflowed effluent samples were collected at the 12-h interval using gas-tight polyethylene bottles with 250 μL of 50 % ZnCl_2 solution and analyzed within 6 h. At that moment, several samples were collected and filtered by baked Whatman GF/F filters to determine the $\text{NO}_3^-\text{-N}$ concentration in the water samples. Dissolved N_2 compound were determined by membrane inlet mass spectrometry (MIMS) with a detection limit of $0.06\ \mu\text{mol L}^{-1}$ (Yin et al. 2015).

The rates of the denitrification and anammox processes across the SWI were calculated from the differences in concentrations of dissolved nitrogen gases between influent and effluent samples, cross-sectional area of the core, and flow rates, which were expressed as micromoles of N per square meter per hour. Both the calculations of denitrification rates and anammox rates adopted the method used by Yin et al. (2015) and Nielsen (1992) and Thamdrup and Dalsgaard (2002).

Quantification of physicochemical characteristics

To determine the sediment core oxygen microprofiles, oxygen concentration was measured by an optical fiber O_2 microelectrode with a tip diameter of less than 0.1 mm (Presens, Germany). Sediment water content was determined by drying fresh sediment to a constant weight at $105\text{ }^\circ\text{C}$. Porosity and

bulk density were measured with the use of a cutting ring (Graca et al. 2004). TN and TP in sediments were measured by the potassium persulfate oxidation method, and the TOC in sediments was measured by the method of potassium dichromate oxidation-ferrous sulfate titrimetry (Jin and Tu 1990). The $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and $\text{NO}_2^-\text{-N}$ concentrations in surface water and pore water were filtered (Whatman GF/F) and measured by a flow-injection autoanalyzer (Skalar Sanplus, The Netherlands).

Calculation of nutrient fluxes

Under steady state conditions, the nutrient fluxes across the SWI occurred mainly through chemical diffusion induced by the concentration gradient. The amounts of nutrient fluxes across the SWI can be calculated according to Fick’s first law of diffusion (Ullman and Aller 1982):

$$F = \varnothing D_s \frac{\partial C}{\partial x} |_{x=0} \tag{2}$$

where \varnothing is the porosity of surficial sediment, and $\frac{\partial C}{\partial x} |_{x=0}$ is the concentration gradient between the surficial sediment and bottom water, determined as the concentration difference between the interstitial water sample and the bottom lake water. The elaborated operation for the concentration gradient was adopted from the study performed by Zhong et al. (2008). D_s is the sediment diffusion coefficient for various nutrients.

There is an empirical equation provided by Ullman and Aller (1982) for the relationship between the real diffusion coefficient and the porosity:

$$D_s = \varnothing D_0 \quad \varnothing < 0.7 \tag{3}$$

$$D_s = \varnothing^2 D_0 \quad \varnothing > 0.7 \tag{4}$$

where D_0 is the ideal diffusion coefficient of the infinite dilution solution. The diffusion coefficients of NH_4^+ , NO_3^- , and NO_2^- at 25 °C are $17.6 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, $19.0 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, and $19.1 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, respectively. The D_s value was corrected for the field temperature and was calculated using the means of three replicates of porosity in the upmost 2 cm depth sediments.

Mass balance in individual sediment cores

The rough budgets constructed for N loss from sediments induced by dredging was mainly including the diffusion fluxes of inorganic N from pore water and the microbial denitrogen processes associated with denitrification and anammox processes. Simultaneously, we also took the N fixation process into consideration, because the N fixation rate in Lake Taihu was an important source compared with microbial

denitrogen mass (Zhang 2012). The loss of N from sediments caused by dredging can be calculated as follows:

$$M = [(R_1 + R_2 + R_3 - R_4) - (r_1 + r_2 + r_3 - r_4)] \times S \times 365 \tag{5}$$

where M represents the total amount of N loss from sediments induced by dredging, $R_1, R_2, R_3,$ and R_4 are the denitrification rates, anammox rates, diffusion fluxes of inorganic N, and N fixation rates in control sediments, respectively; $r_1, r_2, r_3,$ and r_4 represents the denitrification rates, anammox rates, diffusion fluxes of inorganic N, and N fixation rates in dredged sediments. S is the area, and the constant 365 represents 1 year.

Statistical analysis

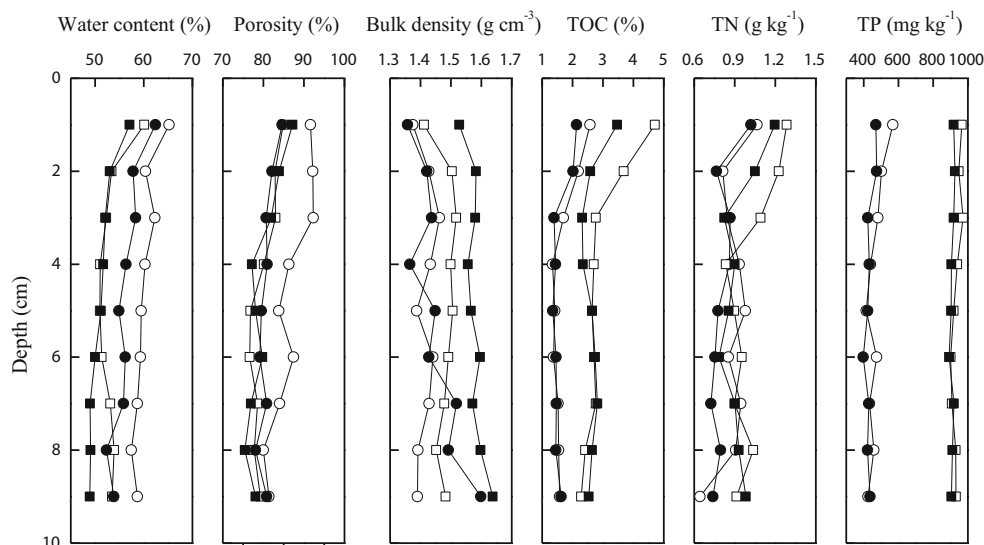
The data shown in this study were represented as the means and standard deviations of three replicates. The differences of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and $\text{NO}_2^-\text{-N}$ concentrations of pore water in different experimental treatments, sampling times, and sediment depths were tested by two-way repeated measures analysis of variance (RM-ANOVA) with the time as the repeated factor. If a significant difference was determined, Tukey’s post hoc test was used to detect the differences between treatments and depths. Likewise, the differences in $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and $\text{NO}_2^-\text{-N}$ fluxes across the SWI were also tested by one-way RM-ANOVA with time as the repeated factor. The differences in the nitrification rates, denitrification rates, denitrification rates of nitrate from coupled nitrification (Dn), and from the overlying water (Dw) and anammox rates across SWI were detected by one-way RM-ANOVA, with time as the repeated factor. Pearson correlation analysis was used to test the correlation between the denitrification rates and anammox rates. The paired sample *t*-test was used to test for significant differences in the variables of the mass of nitrogen loss between undredged and dredged samples. All statistical analyses in this study were performed by SPSS 13.0 (SPSS, USA).

Results

Sediment characterization

The data for undredged and dredged sediments showed large spatial variances between the inner bay and the outer bay; the TOC, TN, and TP contents of the inner bay were generally much greater than those of the outer bay (Fig. 2). The tendencies of water content, porosity, TOC, TN, and TP of sediments generally decreased with depth, whereas the bulk density increased with depth in both the inner bay and the outer bay. Elevated water content and porosity in the outer bay were recorded in the undredged treatment all over the depths. The

Fig. 2 Physicochemical characteristics of different layer sediments after dredging in the inner bay (*square*) and the outer bay (*circle*). The hollow and filled symbols represent the undredged and dredged cores, respectively



bulk density, TOC, TN, and TP in the outer bay exhibited higher values in undredged samples at depths of 0–4 cm than those of the dredged samples and changed with a reciprocating oscillation trend at depths of 4–9 cm. In the inner bay, porosity and bulk density in dredged sediments were higher at depths of 2 and 9 cm, respectively, compared with undredged sediments, and others were similar to the changes in the outer bay, which indicated an opposite effect of dredging in these two sites for porosity and bulk density. The penetration depths of oxygen in dredged sediments were generally shallower than those in undredged samples (Fig. 3).

Fluxes of inorganic nitrogen release across the SWI

The inorganic nitrogen fluxes were calculated on the basis of Fick's first law. During the experiment, sediment acted as a

nitrogen source to the water column, and the nitrogen release was dominated by $\text{NH}_4^+\text{-N}$ (Table 1). In the inner bay, the release rates of $\text{NH}_4^+\text{-N}$ in undredged sediments were much higher than those in dredged sediments (Tukey's HSD test, $P < 0.05$), and the peak fluxes were observed after dredging at day 720. In the outer bay, the dredging significantly increased the release of $\text{NH}_4^+\text{-N}$ from day 3 to day 180 (Tukey's HSD test, $P < 0.05$). However, sediment was a sink of $\text{NO}_3^-\text{-N}$ in overlying water, except for the $\text{NO}_3^-\text{-N}$ fluxes on day 720. The $\text{NO}_2^-\text{-N}$ fluxes were similar between different treatments (Tukey's HSD test, $P > 0.05$), and the sediment served as a sink initially and then served as a source in both sites (Table 1).

Nitrification

Nitrification rates of undredged sediments were much greater than those of dredged sediments (Fig. 4), and the differences between control and dredged sediments were significant in the inner bay ($P < 0.05$) but not for the outer bay (Table 2). As for the chronological changes in nitrification rates, the nitrification rates of dredged sediments attained the peak value after dredging for 180 days at both sites, whereas the values of the control group had no similar pattern.

Denitrification

Figure 5 shows the denitrification variations of the control and dredged treatments with time ongoing. The denitrification rates of undredged and dredged sediments increased gradually from day 90 to day 270 in both the inner bay and the outer bay (Table 2) but drastically decreased on day 360 (Fig. 5). In principle, the denitrification rates of the undredged treatment group were significantly greater than those of the dredged

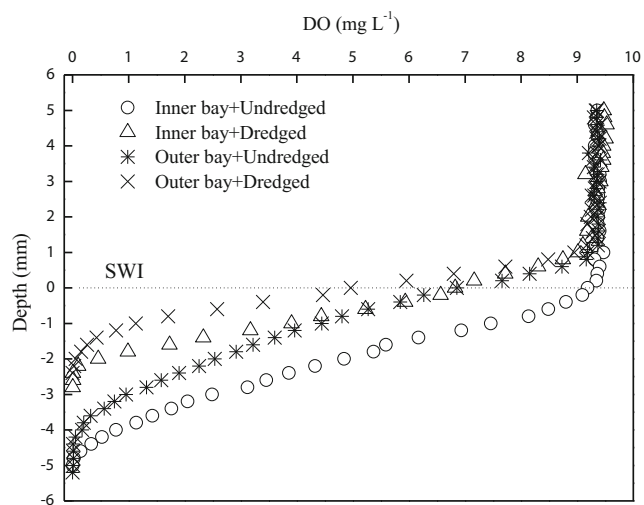


Fig. 3 Oxygen profiles in different sediment cores on day 40 after dredging. The dotted line at 0-mm depth represents the sediment-water interface (SWI)

Table 1 Diffusive fluxes of inorganic nitrogen across the sediment-water interface based on Fick’s first law. Positive fluxes are out of sediment, and values are expressed as the mean±SD of three replicates

Flux (mmol m ⁻² day ⁻¹)	Time (day)	Inner bay		Outer bay	
		Undredged	Dredged	Undredged	Dredged
NH ₄ ⁺ -N	3	5.35±1.35	4.91±1.18	6.97±2.38	10.64±1.64
	30	7.87±2.14	8.05±1.49	4.24±0.57	10.32±2.07
	90	5.43±2.13	5.28±1.31	4.34±1.68	6.94±2.37
	180	6.54±0.37	4.86±1.45	6.25±2.04	8.29±1.57
	360	12.06±2.58	4.89±2.03	10.32±1.72	8.09±0.83
	720	12.30±4.13	11.91±2.75	13.19±0.93	10.79±3.28
NO ₃ ⁻ -N	3	-1.60±0.13	-1.44±0.12	-2.04±0.21	-1.65±0.06
	30	-1.69±0.18	-1.70±0.11	-2.09±0.07	-1.75±0.24
	90	-0.65±0.06	-0.67±0.01	-0.73±0.02	-0.69±0.02
	180	-0.43±0.03	-0.26±0.02	-0.15±0.02	-0.73±0.05
	360	-0.15±0.02	-0.25±0.01	-0.38±0.01	-0.27±0.01
	720	1.03±0.28	1.37±0.16	1.73±0.12	0.82±0.11
NO ₂ ⁻ -N	3	-0.06±0.01	-0.04±0.01	-0.09±0.00	-0.06±0.01
	30	-0.03±0.00	-0.02±0.00	0.01±0.00	-0.02±0.00
	90	-0.04±0.00	-0.04±0.01	-0.04±0.00	-0.05±0.00
	180	0.03±0.00	0.03±0.00	0.04±0.00	0.00±0.00
	360	0.01±0.00	-0.02±0.01	-0.02±0.01	0.00±0.00
	720	0.04±0.00	0.05±0.00	0.07±0.00	0.00±0.00

treatment over the whole experiment in the inner bay (Table 2; Fig. 5a) but not for the outer bay (Fig. 5b).

In the inner bay and the outer bay, Dn and Dw rates were significantly correlated with both the denitrification and anammox rates ($P < 0.01$; Figs. 5 and 6). Both the rates of Dn and Dw increased from day 90 to day 270 but sharply decreased on day 360, and the maximum values and much lower values were achieved on day 270 and days 90 and 360 in the inner and outer bays (Fig. 5). There was a significant difference in Dn between the undredged and dredged

sediments but no significant difference in Dn in the outer bay and Dw in both the inner and outer bays (Fig. 5).

Anammox

Anammox rates changed in a similar pattern to denitrification rates (Figs. 5 and 6). Significant differences in anammox rates were determined between the control and dredged treatments, and the anammox rates of the undredged group were generally greater than those of the dredged treatment (Fig. 6). The

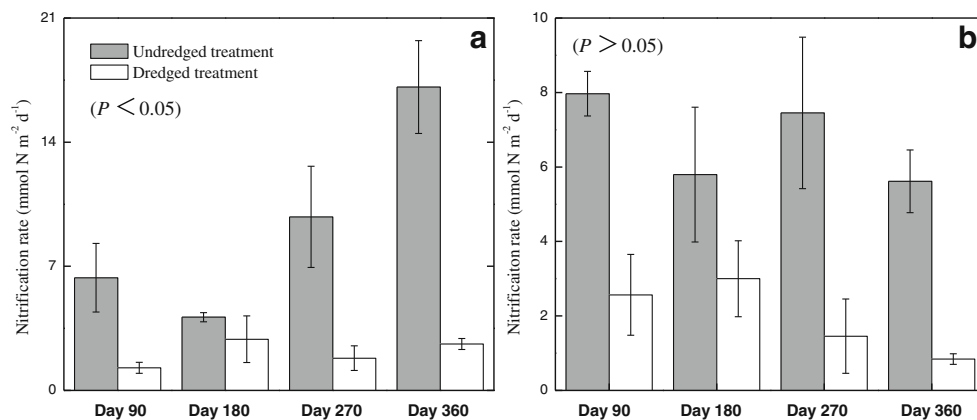
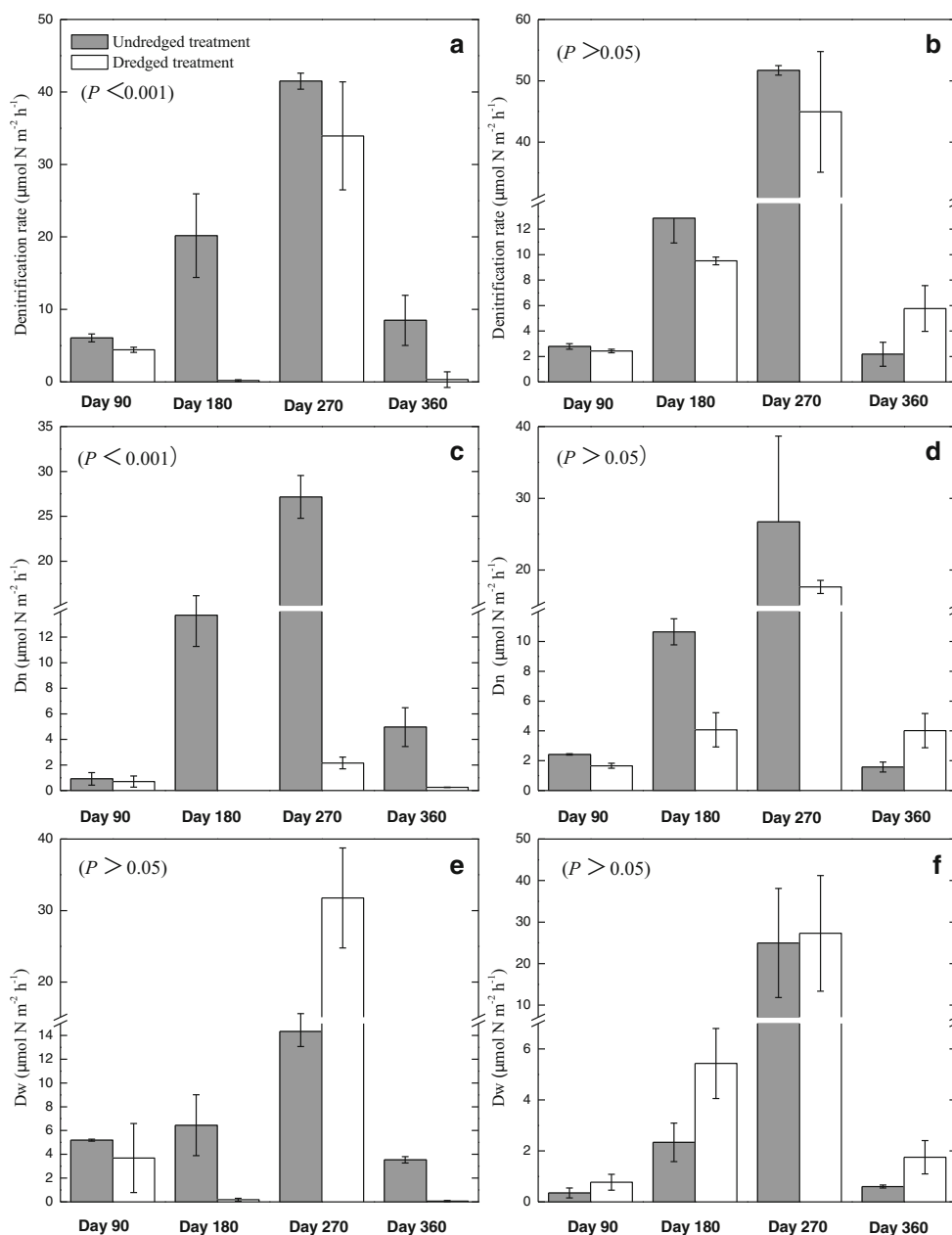


Fig. 4 Nitrification potential rates in the undredged and dredged sediments of the inner bay (a) and the outer bay (b) after different dredging times. All results are expressed as the mean±SD of three replicates. The *P* value shown in figures indicate the significant

difference between control and dredged sediments during the entire experiment using one-way repeated analysis of variance (RM-ANVOA) with time as the repeated factor

Fig. 5 Denitrification rates and denitrification rates of nitrate from coupled nitrification (Dn) and from the overlying water (Dw) across the sediment-water interface in the undredged and dredged sediments of the inner bay (a, c, e) and the outer bay (b, d, f) after different dredging times. All results are expressed as the mean \pm SD of three replicates. The *P* value showed in figures indicate the significant difference between control and dredged sediments during the entire experiment using one-way repeated analysis of variance (RM-ANVOA) with time as the repeated factor



maximum values of both control and dredged treatments appeared on day 270, but much lower values occurred on day 90 and day 360 at both sites (Fig. 6).

Discussion

The effects of dredging on nitrogen removal across the SWI

The fluxes of ammonium demonstrated that the sediment was the main source of the overlying water, but not for NO_3^- -N and NO_2^- -N (Table 1). The ammonium fluxes induced by

dredging were effectively reduced in the inner bay, which were identical to previous study by Zhong et al. (2009). In the inner bay, this result may be attributed to the following four reasons: (1) Ammonium release was due to the basic processes of ammonification and DNRA; similarly, dredging resulted in substantially lower redox potentials and oxygen depletion rates in sediment, which enhanced the DNRA and ammonification processes (Yin et al. 2002; Fig. 3). (2) The porosities of dredged sediment were lower than those of the control treatment (Fig. 2). Consequently, the diffusion of ammonium into the overlying water is hampered by the compact sediment of dredged cores (Zhong et al. 2009). (3) The presence of major factors blocking ammonium migration in

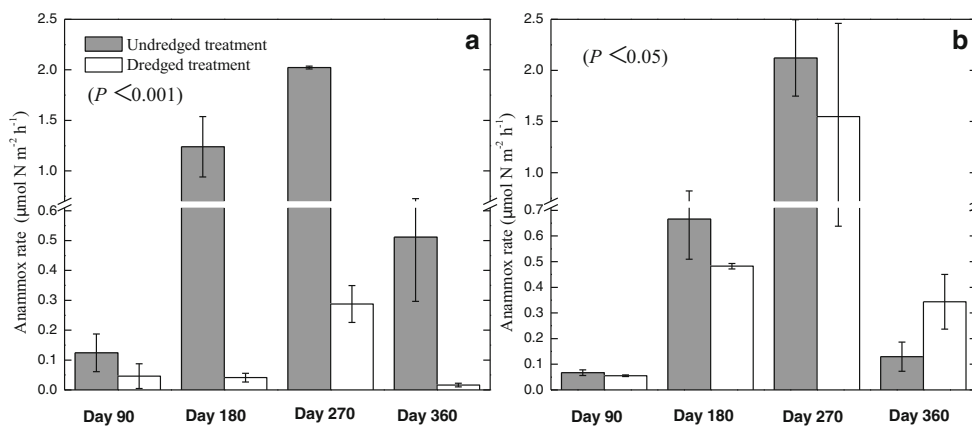


Fig. 6 Anammox rates across the sediment-water interface in the undredged and dredged sediments of the inner bay (a) and the outer bay (b) after different dredging times. All results are expressed as the mean±SD of three replicates. The *P* value showed in figures indicate

the significant difference between control and dredged sediments during the entire experiment using one-way repeated analysis of variance (RM-ANVOA) with time as the repeated factor

sediment that was adsorbed by soil colloids such as Fe/Mn oxides and organic matter, and precipitated with fine-grained sediment particles. Fan et al. (2004) showed that the average pore water $\text{NH}_4^+\text{-N}$ concentration showed a downward increasing trend from SWI to bottom sediment in Meiliang Bay, resulting from the lower redox potential and dissolved oxygen in subsurface sediment (Fan et al. 2000; Fig. 3). Therefore, the $\text{NH}_4^+\text{-N}$ -rich sediment layer was overturned to the surface. (4) The result may be driven by ammonium concentrations in preexisting pore water and adsorbed $\text{NH}_4^+\text{-N}$ concentrations plus any ongoing N regeneration (Morin and Morse 1999; Reddy et al. 2007).

Dredging had minor effect on the fluxes of $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$, which demonstrated that the concentrations of $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ were similar between pore water and overlying water (Table 1; Fig. S1) despite the potential nitrification rates of undredged sediments that were much greater than those of dredged sediments, and it was true for the denitrification rates (Fig. 5). Therefore, the reasons for these results were mostly due to nitrification undergoing anammox or denitrification in the sediment; furthermore, it could also be assimilated by the phytoplankton.

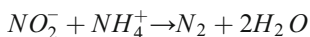
Basic microbial processes to which nitrogen was subjected in the sediment included denitrification, nitrification, ammonification, anammox, and DNRA processes, all of which may result in nitrogen removal from the sediment (Hanaki et al. 1990; Zhong et al. 2010a, b; Trimmer et al. 2003; Wang and Li, 2015). In the present study, the rates of denitrification and anammox induced by dredging were significantly reduced in the inner bay, but not for the denitrification rates of the outer bay (Figs. 5 and 6). These differences can be attributed to complicated environmental factors such as nitrate concentration (Liu et al. 2015; Venterrink et al. 2003; Zhong et al. 2010a, b; Seitzinger 1994), quality and quantity of the organic carbon (Bastviken et al. 2005), soil texture (D’Haene et al.

2003), oxygen concentration (Cavari and Phelps 1977), temperature (Mogge et al. 1999), light conditions, salinity (Toet et al. 2003), and bioturbation of benthic animals (Svensson and Leonardson 1996). There were five reasons for this given as follows. (1) The new surface sediments after dredging were buried in the deeper depth below the bottom for several decades, even for hundreds of years, and the labile organic matter content and bioavailability were limited in sediments after dredging (Pinay et al. 2003; Bastviken et al. 2005). (2) The microbial activity and functional diversity of the microbial community in dredged sediments were significantly lower than those of in undredged sediments (Zhong et al. 2010a, b; Wang and Li, 2015). Furthermore, previous study showed that adding glucose can increase the denitrification rate in the outer bay (Zhong et al. 2010a, b; Chen et al. 2015), which can also be confirmed by the lower TOC content in dredged sediments compared with undredged sediments (Fig. 2). (3) There were lower water contents, porosities, oxygen depletion rates, and oxygen penetration depths in dredged sediments than those of in the undredged treatment (Figs. 2 and 3). (4) Bioturbation stimulated the denitrification in the undredged sediments, whereas bioturbation in dredged sediments was weakened as benthic animals located in the surface sediment were removed by dredging (Svensson and Leonardson 1996). (5) Above all, nitrate from the water and sediment was considered to be the key factor in regulating the denitrification. In the present study, the denitrification rates of nitrate from overlying water in dredged sediments was much higher than that in undredged sediments during the experiment (Table 2). This result was accordant with results from Christensen et al. (1990) who also demonstrated that diffusion of nitrate from the overlying water into the sediment was the major nitrate source for denitrification in a Danish nitrate-rich stream.

Table 2 The relative contributions of Dn, Dw, and anammox processes to the microbial nitrogen removal of undredged and dredged sediments after different dredging times in the inner bay and the outer bay

	Percentage (%)	Time (day)	Inner bay		Outer bay	
			Undredged	Dredged	Undredged	Dredged
Dn		90	14.8	15.6	84.6	66.7
		180	64.1	0.1	78.7	40.8
		270	62.4	6.3	49.7	38.0
		360	55.1	75.0	68.3	65.7
Dw		90	83.9	82.5	12.2	31.0
		180	30.1	81.5	17.3	54.4
		270	32.9	92.9	46.4	58.7
		360	39.2	20.1	26.1	28.7
Anammox		90	2.0	1.0	2.3	2.2
		180	5.8	18.4	4.9	4.8
		270	4.6	0.8	3.9	3.3
		360	5.7	4.9	5.6	5.6

Note that the rates of anammox decrease was partly due to the denitrification process. It was unexpected that the temporal changing in denitrification rates was significantly correlated with the anammox rates during the entire simulation course ($P < 0.01$; Figs. 5 and 6). Previous work showed that anaerobic ammonium oxidation coupled the oxidation of NH_4^+ to the reduction of NO_2^- as in the following equation:



As in the above general reaction, the concentration of NO_2^- , which serves as an electron acceptor, rapidly increases by denitrification. This process can effectively enhance the anammox process in dredged sediment (Trimmer et al. 2003). The results also indicated that the denitrification rates significantly correlated with anammox rates in sediments of both the inner bay and the outer bay ($R^2 = 0.959$ for undredged sediments in the inner bay, $R^2 = 0.990$ for dredged sediments in the inner bay, $R^2 = 0.993$ for undredged sediments in the outer bay, and $R^2 = 0.933$ for dredged sediments in the outer bay), which was also verified by previous study (Rysgaard et al. 2004).

The implications of dredging on nitrogen cycling under different pollution statuses of sediments

Nitrogen cycling in sediments was commonly regulated by organic matter decomposition, sediment deposition rates, bioturbation/bioadvection, and rates of microbial nitrogen transformation (Joye and Anderson 2008). There were different water quality statuses and diverse ecological types across Lake Taihu (Duan et al. 2009; Ma et al. 2008). In such cases, some dredging plans were implemented or will be put into practice mainly in the northern bays to reduce internal nutrient loading of sediments. To date, more attention has been paid to

evaluate the environmental effectiveness of dredging projects, but little knowledge is available concerning the implications of dredging to nitrogen mass balance under different pollution statuses of sediments. On the other hand, the TOC, TP, and TN contents of the inner bay sediments were generally greater than those of the outer bay, and the TOC and TP in the inner bay were approximately twice as high as those of the outer bay (Fig. 2). Therefore, nitrate removal after dredging was inevitably influenced by pollution status (Fan et al. 2004). Taken together, it was vital to assess the implications of dredging to nitrogen cycling in lake ecosystem restoration.

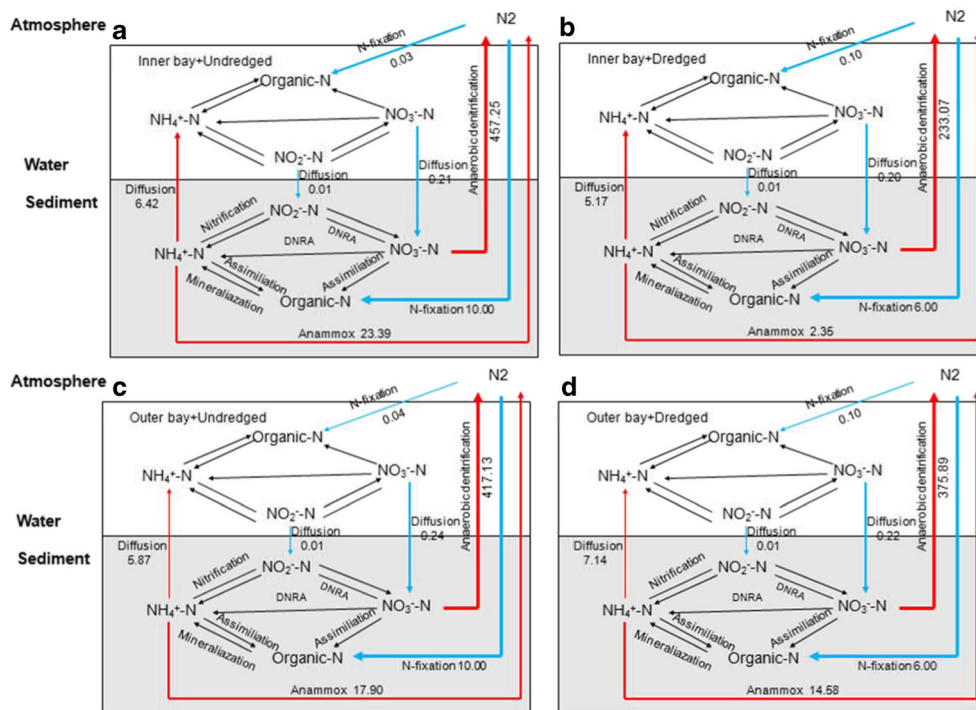
First, note that the patterns of change in NH_4^+ -N fluxes of the inner bay and the outer bay were opposite (Table 1). The opposite changing pattern for the inner bay and the outer bay may be attributed to several reasons as follows: (1) The NH_4^+ -N flux was regulated by the flux of preexisting pore water and adsorbed NH_4^+ -N concentrations plus any ongoing N regeneration, and this process was mainly driven by the decomposition of organic nitrogen (Reddy et al. 2007; Zhong et al. 2009). This result could be further validated by the TOC content of sediments in inner bay (Fig. 2). (2) The TOC and TN contents generally decreased with depth at both sites, and the NH_4^+ -N release of the outer bay may be driven by the preexisting pore water because the NH_4^+ -N-rich sediment layer was overturned to the surface (Fan et al. 2000; Fig. 2). However, the TOC and TN-rich sediment was removed by dredging, and the fresh surface sediment had a lower NH_4^+ -N gradient potential between the pore water and the overlying water, which resulted in a lower NH_4^+ -N flux (Yu et al. 2012). (3) The main basic microbial processes for NH_4^+ -N release usually include denitrification, nitrification, ammonification, anammox and DNRA processes, all of which may result in changes in NH_4^+ -N fluxes (Hanaki et al. 1990; Zhong et al. 2010a, b; Trimmer et al. 2003). The yearly average potential nitrification rates of dredged sediments for the inner bay were

greater than those of the outer bay. The same was true for the yearly average denitrification and anammox rates (Figs. 4, 5, and 6), which demonstrated the lower capacity of the outer bay to transform $\text{NH}_4^+\text{-N}$ to $\text{NO}_3^-\text{-N}$ compared with that of the inner bay, resulting from the lower abundance of nitrifying bacteria, the lower energy yield of nitrification, and the lower nitrifier biomass of freshly generated surface sediments in the outer bay (Cornwell and Owens 2011).

A second important issue was how to budget the associated denitrogen mass caused by dredging in the inner bay and the outer bay. Using the information from the fluxes of inorganic nitrogen and dissimilatory microbial processing of N, we can construct a rough budget for N induced by dredging in both the inner bay and the outer bay (Fig. 7). The total mass of nitrogen removal associated with denitrification and anammox processes was estimated to be 486.80 and 240.36 $\mu\text{mol m}^{-2} \text{day}^{-1}$ of undredged and dredged sediments, respectively; in the inner bay, 440.62 and 397.41 $\mu\text{mol m}^{-2} \text{day}^{-1}$ of undredged and dredged sediments in the outer bay (Fig. 7). More specifically, the rates of denitrification were 457.25 and 233.07 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for undredged and dredged sediments in the inner bay and 417.13 and 375.89 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for undredged and dredged sediments in the outer bay, respectively. The rates of anammox were 23.39 and 2.35 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for undredged and dredged sediments in the inner bay and 17.9 and 14.58 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for undredged and dredged sediments in the outer bay, respectively. In terms of nitrogen releases from sediment to overlying water, the sediment was a main source of ammonium based on Fick's first law, but not

for $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ (Table 1; Fig. S1). Additionally, taking into consideration the nitrogen fixation rates in the overlying water and sediment at the above two sites in Meiliang Bay, the rates of microbial nitrogen fixation in Meiliang Bay were 10.03 and 6.10 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for the dredged and undredged treatments in the inner bay and 10.04 and 6.10 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for the dredged and undredged treatments in the outer bay, respectively (Zhang 2012). Moreover, the final denitrogen mass was 476.77 and 234.26 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for the control and dredged treatments in the inner bay and 430.58 and 391.31 $\mu\text{mol m}^{-2} \text{day}^{-1}$ for the control and dredged treatments in the outer bay, respectively (Fig. 7). Based on the above analyses, the mass of denitrogen and diffusive fluxes induced by dredging in Meiliang Bay were decreased by 161.10 and 26.09 t N a^{-1} for the inner bay and the outer bay, respectively (with an area of 130 km^2 for Meiliang Bay), suggesting that dredging had a negative effect on decreasing nitrogen removal from sediments in Meiliang Bay, Lake Taihu. In addition, the nitrogen removal induced by dredging was mainly by denitrification process (Liu et al. 2014), accounting for 90.97 and 95.44 % for the inner bay and the outer bay, respectively. Therefore, the results demonstrated that dredging may be more applicable to heavily polluted sediments, and the reasons may be attributed to the fact that higher nutrient concentrations and organic matter contents in sediments will enhance water-sediment interface processes and nutrient regeneration (Fan et al. 2004). Dredging with certain depth can effectively reduce the nutrient loading in sediments but also can reduce the release of ammonium across the SWI.

Fig. 7 Rough budgets constructed for the nitrogen balance of undredged and dredged sediments at two sites in Meiliang Bay, Lake Taihu: **a** inner bay+undredged; **b** inner bay+dredged; **c** outer bay+undredged; **d** outer bay+dredged. Units are $\mu\text{mol m}^{-2} \text{day}^{-1}$ for all processes, which are denoted with superscript. Data for N fixation rates in the overlying water and sediments at the two sites in Meiliang Bay originate from Zhang's Master's degree thesis. The red line represents the N loss out of sediments, and the blue line represents the N transfer into sediment



Conclusions

The results showed that, with regard to the pollution status of sediments, simulated dredging implemented in TOC and/or TN-rich sediments can slightly reduce the NH_4^+ -N release rates from sediments, whereas the temporary NH_4^+ -N release (from day 3 to day 180 after dredging) induced by dredging in sediments with less TOC and/or TN would be enhanced. Moreover, dredging had minimal effects on the fluxes of NO_3^- -N and NO_2^- -N from sediments of both sites. Further analysis demonstrated that dredging would decrease the rates of nitrification, denitrification, and anaerobic ammonium oxidation in sediments, especially in TOC and/or TN-rich areas. After the sediment dredging, the nitrogen removal was reduced by 161.10 and 26.09 t N a⁻¹ for the inner bay and the outer bay, respectively. Taken together, this field study demonstrated that dredging may be more applicable to nutrients and TOC-rich sediments in minimizing the internal N pollution once the external N pollution was controlled.

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