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Nitrogen budget at sediment–water interface altered by sediment dredging and settling particles: Benefits and drawbacks in managing eutrophication

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ABSTRACT

Internal nitrogen (N) loading of lakes is commonly controlled by sediment dredging, although its comprehensive effect on internal N loading remains unclear. Herein, we examined the long-term effects of sediment dredging on internal N loading from a new perspective on the N budget at the sediment—water interface (SWI) through a simulation of field dredging performed by incubating intact sediment cores from a shallow eutrophic lake (Lake Taihu). We further evaluated the role of settling particles (SP) in the recovery of N cycle processes after dredging and its potential impact on the N budget. Our results demonstrated that dredging could help reduce organic matter and total N in sediments; improve the redox environment of the SWI; slow down N mineralization, N fixation, denitrification, and anaerobic ammonia oxidation (anammox); and alter the N budget at the SWI and the contribution of various N cycle processes. However, the input of SP enriched in fresh organic matter and N could accelerate the recovery of N cycle processes at the SWI, reducing the variation in the N budget and the contribution of each N cycle process caused by dredging. Dredging significantly reduced the N flux at the SWI, which was evident from the reduction of inorganic N release flux and N removal through denitrification and anammox. Therefore, sediment dredging has its advantages and disadvantages in managing internal N loading in lakes. To maintain a long-term control on the release of internal N through sediment dredging, measures should be taken based on the in-lake and watershed to inhibit the inflow and settlement of particulate matter.

1. Introduction

Nitrogen (N) is an important nutrient limiting factor for maintaining primary productivity and the food chain structure in lake ecosystems (Vitousek et al., 1997; Seitzinger and Kroeze, 1998). However, over the past decades, high-intensity human activities have resulted in the discharge of excessive active N into lakes, causing eutrophication and a series of environmental problems, such as algae outbreaks, hypoxia, and ecosystem disasters (Carpenter et al., 1998; Elser et al., 2007; Paerl et al., 2014). In addition to external N loading, increasing evidence suggests that internal N loading, i.e., N release from sediment, contributes considerably to the N content in overlying water (Sugimoto et al., 2014; Wu et al., 2017). Even if the external N loading of a lake is effectively reduced, internal N loading alone could sustain eutrophication, thereby delaying its restoration process (Sugimoto et al., 2014; Wu et al., 2017). Therefore, to control such eutrophication, managing the

internal N loading needs significant attention.

Environmental dredging is the most commonly used measure to control internal loading. Over the past decades, sediment dredging has been widely used in controlling internal loading globally (Ryding, 1982; Does et al., 1992; Lürling and Faassen, 2012; Waajen et al., 2016). As the effectiveness of sediment dredging on internal loading is affected by various factors, e.g., external input, dredging residue, and site-specific conditions (Bridges et al., 2010; Palermo and Hays, 2014), it is difficult to understand its effectiveness based on the variations in water quality and relevant indicators in dredged waterbodies, bringing considerable challenges in accurately evaluating its long-term efficacy in managing pollutants (NRC, 2007; Gustavson et al., 2008). As sediment dredging is a costly restoration measure, more pilot tests based on pollutant flux at the sediment—water interface (SWI) need to be conducted to understand the long-term efficacy and primary factors influencing sediment dredging in controlling internal loading, to improve the

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decision-making process for remediating contaminated sediments.

Environmental dredging aims to mainly reduce the risk of nutrients being released from the sediment to the overlying water. Therefore, most of the previous studies focused on the short-term/long-term efficacy of sediment dredging on internal N release (Cornwell and Owens, 2011; Yu et al., 2016a; Liu et al., 2017), whereas only a few focused on denitrification and other N cycle processes (Zhong et al., 2010a; Jing et al., 2013; Yu et al., 2016b). In fact, the N budget at the SWI depends on multiple N transformation processes. In the sediment, certain N cycle processes act as a source (e.g., N fixation), others represent a sink (e.g., denitrification), and some represent either a source or sink according to the environmental conditions (e.g., N mineralization and inorganic N diffusion at the SWI). Therefore, to comprehensively evaluate the efficacy of sediment dredging on the internal N loading, quantifying the N budget at the SWI is necessary.

In actual lake dredging, deep buried sediment is exposed and interacts with the overlying water to form a new SWI after dredging. This new SWI inevitably receives and accumulates settling particles (SP) from the water column (Murakami, 1984; Fan et al., 2004). SP could be either allochthonous i.e., they could originate from external sources, such as riverine input or atmospheric deposition, or autochthonous, meaning they stem from within a water body with the inclusion of surface sediments (due to advection and resuspension), phytoplankton debris, among others (Pettersson, 2001; Bukaveckas et al., 2019). SP are considered to be the primary transport media for nutrients, heavy metals and organic pollutants; they play an important role in transporting bioavailable nutrients and pollutants at the SWI (Weyhenmeyer et al., 1997; Bukaveckas et al., 2019). In shallow lakes, SP are continuously resuspended and resettled due to wind waves and bioturbation (Wevhenmeyer et al., 1997); this continuous deposition could be a key factor affecting the N budget at the SWI after dredging.

Herein, we examined the effects of dredging on sediment N mineralization, N fixation, denitrification and anammox through a one-year field dredging simulation. Based on this simulation, we quantified the N budget at the SWI, and evaluated the impact of continuous deposition of SP on the recovery of N cycle processes after dredging. We hypothesized that the dredging will reduce the SWI flux of internal N via directly influencing the sediment N transformation, whereas the SP sedimentation will promote it. We further aimed to unravel the joint effects of these two main drivers underlying the internal N loading. To the best of our knowledge, this is the first study that comprehensively evaluates the effect of sediment dredging on internal N loading based on the N budget at the SWI through the long-term incubation of intact sediment cores. The results could help provide a basis for decision-making in managing the internal N loading of shallow lakes.

2. Materials and methods

2.1. Experimental design

The sampling site is located to the north of Taihu Lake in the inner bay of Meiliang Bay (Fig. S1), which is the most eutrophic part of the lake, with an area of 129 km² and an average depth of 1.9 m (Chen et al., 2003). The site-specific environment is described in our previous study (Wen et al., 2020). In January 2018, 240 intact sediment cores were collected by a sediment core sampler (length: 70 cm, diameter: 9 cm), and the length of each core was approximately 60 cm. To ensure homogeneity, sediment cores were collected within a 50 m² area and then prepared in the field. For the dredged core samples, 120 intact sediment cores were randomly selected; a 30 cm sediment layer was removed from the surface of each core, and the remaining 25 cm was transferred to a fresh plexiglass tube without headspace and used as dredged cores. We considered a simulated dredging-depth of 30 cm based on the actual dredging-depth of the Meiliang Bay dredging project (Yu et al., 2016a). For the non-dredged sediment cores (control), 25 cm of the sediment layers from the surfaces of the remaining 120 cores were transferred to

fresh plexiglass tubes with the bottom plugged with rubber-plugs. To explore the influence of SP on the N transformation processes after dredging, we randomly took 55 dredged and control sediment cores and covered the top of the tubes with an 800 mesh nylon cloth (18- μ m mesh-size) to prevent SP from entering the culture tubes. To ensure that the overlying water could enter the tubes during incubation, we replaced the nylon cloth every month to prevent SP or a biofilm from blocking the mesh.

Herein, four treatments were carried out: (i) ND, non-dredged sediment cores; (ii) D, dredged sediment cores; (iii) NDS, non-dredged sediment cores with SP deposition; and (iv) DS, dredged sediment cores with SP deposition. There were 65 cores for ND and D treatments and 55 cores for NDS and DS treatments. Long-term in-situ incubation was carried out in the lake using the field observation platform of Taihu Lake Ecosystem Research Station, located on the shore of Meiliang Bay (Fig. S1). The sediment cores were randomly placed into the stainless-steel frames, each having six sediment cores. The stainless-steel frames were fixed at one end of a nylon rope and then lowered into the lake-bottom (~2 m deep) for in-situ incubation. The other end of the nylon rope was attached to a fixture on the field observation platform. A flowchart of the experimental design is included in the supplementary materials (Fig. S2).

During incubation, nine sediment cores were taken from each treatment at the sampling interval to analyze the N cycle rate and physicochemical properties: three for collecting pore water samples and determining the inorganic N flux at the SWI as well as the sediment properties; three for determining the denitrification and anammox rates; and three for analyzing the N fixation rate. Simultaneously, the samples of in-situ overlying water at the lake bottom were collected to determine N cycle rates. At the same time, a polyethylene pipe with a height of 30 cm and a diameter of 9 cm was used to collect SP via the sedimentation method. All collected samples were quickly transported to the lab for subsequent analyses.

2.2. Diffusive flux of inorganic N across the SWI and analysis of pore

The dissolved inorganic N (DIN) flux across the SWI was evaluated through intact sediment core incubation. All sediment cores brought to the lab from the lake were incubated in a water tank in the dark at the insitu water temperature ($\pm~2~^\circ\text{C}$). The filtered in-situ water was then carefully poured along the sediment column walls to avoid disturbance, and the depth of the overlying water was maintained at 20 cm. Water samples (50 mL) were taken from the incubated cores at designated intervals (0, 12, 24, 36, 48, 60, and 72 h) at 5 cm above the SWI and were filtered through 0.45 μm GF/C filters (Whatman, UK) before being analyzed for DIN. Thereafter, the same volume of the original filtered water was immediately added to each core to maintain the water quantity. The detailed processes of this method are described in our previous work (Wen et al., 2020).

The average DIN flux across the SWI was calculated according to Eq. (1). This principle is based on the change in DIN concentration in the overlying water of incubated cores per unit time and unit area (Fan et al., 2002). Over the three-day incubation period, sampling was done 7 times at 12-h intervals. After each sample was obtained, the filtered lake water was used to compensate the water in incubated cores. As shown by Eq. (1), we also calculated the change in DIN concentration caused by the sampling and compensating water samples. The DIN flux obtained was the average values of DIN flux every 12 h.

$$F = \left[V(C_n - C_0) + \sum_{j=1}^n V_{j-1} (C_{j-1} - C_a) \right] / (s \times t)$$
 (1)

where F is the diffusive flux across the SWI (mg m $^{-2}$ d $^{-1}$); V is the volume of water (L) in the sediment core; C_n , C_0 , and $C_{j\cdot 1}$ are the nutrient concentrations of the water at time n, initial time (0), and time "j -1" in

mg L^{-1} , respectively; C_a is the nutrient concentration (mg/L) of the compensating sampled water; $V_{j\cdot 1}$ is the volume of water (L) sampled from the sediment core; s is the area (m²) across the SWI in the sediment core; and t is the incubation time (d). F represents the average exchange flux over three days.

At the end of the static release incubation experiment, a high-resolution equilibration dialysis device (HR-Peeper, provided by Easy-sensor Ltd. www.easysensor.net) was used to sample the pore water. This peeper had a total length of 20 cm with 30 chambers, and a 4 mm pore water resolution was obtained. The peeper probes were inserted vertically into the sediment cores and retrieved after 48 h. Then, the pore water samples were transferred to a 96-well plate, and the concentration of NH $^{\perp}_4$ -N was analyzed immediately. The peeper and the sampling methodology are described in detail in Johnston et al. (2009) and Xu et al. (2012). The photographs of the HR-Peeper in the process of collecting pore water are presented in Fig. S3.

2.3. Determination of denitrification, anammox and N fixation rates

Denitrification and anammox rates were determined simultaneously based on the method described by Nishio et al. (1982) and Usui et al. (2001). The entire sediment cores were tightly covered with resin glass to prevent the upper-part from coming in contact with air. The cores were then wrapped in aluminum foil and cultured in a water bath at in-situ water temperature, which ranged from 6 to 28 °C throughout the experiment. A continuous flow cultivation was adopted and the influent water was sampled from in-situ lake water, the concentration of which was adjusted to 100 μmol L⁻¹ using ¹⁵NO₃-N (the final percentage of $^{15}{
m N}$ was \sim 90–99%, depending on the background nitrate concentration) (Yu et al., 2016b). The influent flow rate was controlled at 1.0 mL min⁻¹ by adjusting the peristaltic pump. In the sediment cores, the inlet pipe was 1 cm away from the SWI and the outlet pipe at the top of the water column, which is conducive to the mixing of water in the column. After 24 h, the influent and effluent samples from each treatment were collected at 12-h intervals using gas-tight bottles (12 mL Exetainer, Labco, High Wycombe, UK) with 250 µL of 50% ZnCl₂ solution. The concentration of dissolved N2 in these samples was determined by membrane inlet mass spectrometry (MIMS) within 6 h. Detailed experimental methods are described in Yu et al. (2016b). To calculate the denitrification and anammox rates, we adopted the methods used by Yin et al. (2015), Nielsen (1992), and Thamdrup and Dalsgaard (2002).

The N fixation rate at the SWI was determined by the acetylene—ethylene assay (Hardy et al., 1968). The incubation tubes contained a 10-cm sediment sample, a 10 cm high water column, and a 2 cm high headspace; the water column contained 10% acetylene saturated water. Incubation was carried out for 4 h; at the end of culture, 20 mL of headspace gas was extracted into a pre-vacuumized bottle, and the ethylene concentration was analyzed by gas chromatography with a flame ionization detector (7890B GC System; Agilent Technologies, Santa Clare, CA, USA). The total amount of ethylene generated in the water and gas phase was calculated according to Henry's law and the solubility of ethylene based on temperature. The N fixation rate at the SWI was calculated quantitatively based on the theoretical conversion coefficient of ethylene generation and fixed N₂ (3:1). The calculation methodology is described in Gettel et al. (2013) and Newell et al. (2016).

2.4. N budget at the SWI

N loss in sediment occurs mainly due to the release of inorganic N from pore water and microbial nitrogen removal associated with denitrification and anammox processes. Simultaneously, microbial N fixation helps increase N in the sediment. Therefore, combining the above four processes, we can roughly estimate the N budget at the SWI based on the following equation in accordance to previous studies (Small et al., 2014; Yu et al., 2016b):

$$M = (R_1 + R_2 + R_3 - R_4) \times S \times T \tag{2}$$

where M is the total amount of N loss from sediments, R_1 is the denitrification rate, R_2 is the anammox rate, R_3 is the diffusion flux of DIN, R_4 is the N fixation rate in the sediment, S is the area of Meiliang Bay, and T is the time (365 d). A positive value of M represents the removal of N from the sediment, and a negative value represents its introduction into the sediment.

2.5. Analysis methodology

Dissolved oxygen (DO) concentration and oxygen penetration depth (OPD) of the sediment cores were measured using the Unisense Microprofling System with a vertical resolution of 0.05 mm (Unisense A/S, Aarhus, Denmark) immediately after they were transported back to the lab. The water content (WC) of the sediment and the SP was determined by drying the fresh samples to a constant weight at 105 °C. The organic matter (OM) content was measured as the loss on ignition (LOI) through the calcination of sediment and SP at 550 °C for 6 h, and total nitrogen (TN) was measured by the alkaline potassium persulphate oxidation method (Chinese EPA, 2002). Labile N (NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N) in the sediments or the SP was extracted using 2 mol L⁻¹ KCl. The DIN in the extracting solution and the other water samples was quantified by a flow-injection analyzer (Skalar Analytical B.V., Breda, The Netherlands) according to the methods described by Grasshoff et al. (1983). The NH₄⁺-N content in the pore water was analyzed using Nessler's reagent method (Chinese EPA, 2002). A microtiter plate reader (Biotek Epoch, Winooski, VT, USA) with a 96-well microtiter plate was used for the photometric detection of NH₄⁺-N.

2.6. Statistical analysis

The samples were analyzed in triplicate, and the results were presented as mean values (\pm SD). Significant differences among the treatments were identified through analysis of variance (ANOVA) followed by Tukey's test. The Pearson correlation analysis was used to test the correlation of the variables. To account for the seasonal variations, we further performed a generalized linear mixed model (GLMM) analysis to better distinguish the effects of dredging and SP deposition on nutrient concentrations, nitrogen transformation processes, and the SWI flux. GLMMs were performed with the R packages lme4 and lmerTest (Bates et al., 2015; Kuznetsova et al., 2017). The graphics were generated with OriginPro 2017C software (OriginLab, Northampton, MA, USA) and ggplot2. Excel 2013 and SPSS (IBM SPSS Statistics 22.0 for Windows) were used for the other quantitative and statistical analyses.

3. Results and discussion

3.1. Basic characteristics of surface sediment (SS) and SP

SS and SP samples were collected periodically from the outset of the experiment and Fig. 1 shows the basic physicochemical properties. Under natural deposition, the WC of SP was ranging 56.0–76.8%, with an average of 67.7%, whereas that of SS was 61.1%. The LOI of SP was significantly higher than that of SS (P < 0.01), with average values of 8.81% and 6.70%, respectively. The TN contents in SP and SS were 2985 and 2047 mg/kg, respectively. The higher LOI and TN content in SP compared to that in SS indicated that SP could be considered as external loading, which significantly influenced the N loading of sediment. For labile N, the NH₄⁺-N content in the SP was significantly higher than that in the SS (P < 0.01), with evident seasonal variation. Compared with NH₄⁺-N, the NO₃⁻-N and NO₂⁻-N contents in SP and SS were relatively low. Cyanobacteria blooms occur annually in Meiliang Bay from spring to autumn (Zhang et al., 2007; Xu et al., 2017). At the incubation time in spring (135 d), the growth and deposition of cyanobacteria-dominated

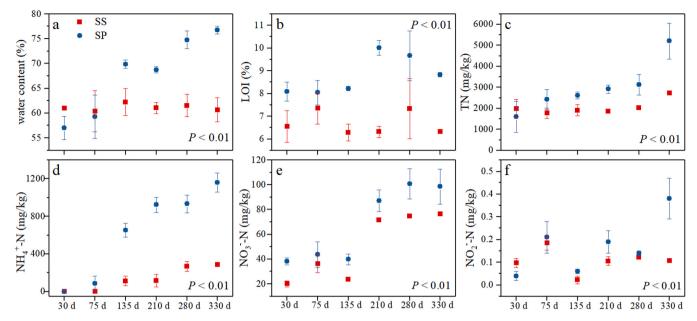


Fig. 1. Water content (a), LOI (b), TN (c), NH_4^+-N (d), NO_3^--N (e) and NO_2^--N (f) of settling particles (SP) and surface sediment (SS, 0–2cm of treatment ND). *P* value in the figure was the result of generalized linear mixed models for the environmental factor. Surface sediment (SS) and settling particles (SP) were considered as fixed factor, and sampling days as random factor.

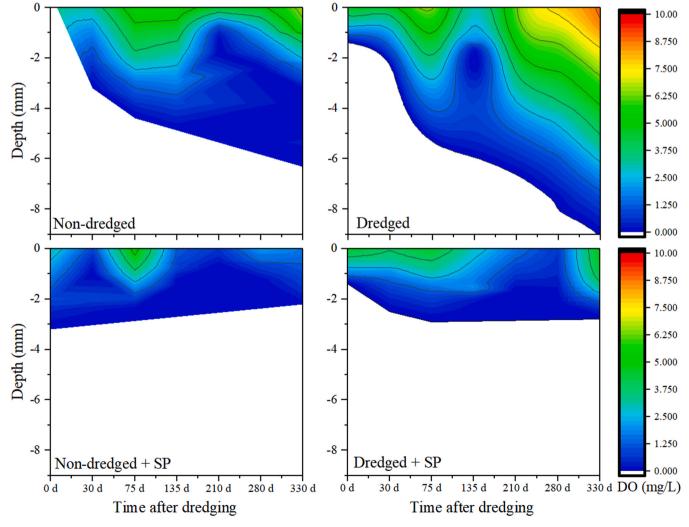


Fig. 2. Dissolved oxygen concentration and penetration depth at the SWI.

phytoplankton caused a significant increase in the WC, LOI, and N content of the SP. In addition, due to the mineralization and decomposition of OM in the SP, the NH $_{4}^{+}$ -N content of the SP increased rapidly after 135 d (Fig. 1d). Although there were evident temporal variations in the physicochemical properties, the statistical results showed that there were significant differences (P < 0.01) in WC, TN, LOI, NH $_{4}^{+}$ -N, NO $_{3}^{-}$ -N, and NO $_{2}^{-}$ -N between the SS and the SP after accounting for seasonal influences (Table S1).

Just after dredging, OPD decreased from the original 3.2-2.4 mm (Fig. 2); the OPD of the dredged cores then gradually increased to 9 mm. The DO at the SWI of treatment D was greater than that of treatment ND, with mean values of 6.20 and 5.39 mg L⁻¹, respectively. This suggests that dredging could improve the redox environment at the SWI, which is consistent with the study of Jing et al. (2013). Under SP deposition, DO at the SWI and OPD of both treatments NDS and DS decreased significantly; the mean DO concentrations at the SWI of treatments NDS and DS were 2.13 and 3.30 mg L⁻¹, and their maximum OPDs were 3.2 and 2.9 mm, respectively. Due to a high content of fresh OM in the SP (Fig. 1b), DO is required for its decomposition as the SP settled to the surface sediment (Kristensen, 2000; Liu et al., 2017; Wen et al., 2020),

causing a significant reduction in DO in treatments NDS and DS (Fig. 2). Generally, the DO at the SWI of each group was lower in summer and autumn, and higher in spring and winter. This phenomenon could be attributed to the increased metabolism of microorganisms and greater DO consumption for the mineralization and decomposition of OM in the seasons with high temperatures (Malecki et al., 2004; Kreling et al., 2016).

3.2. N mineralization in sediment and NH₄⁺-N profile in pore water

Fig. 3 shows the changes in TN and labile N in sediments under the different treatments. Without SP deposition, TN in treatments ND and D was lower in summer and higher in winter; TN in D was 1147.3–2495.5 mg/kg, which was ~15% lower than that of ND. With the deposition of SP, TN in treatments NDS and DS increased with time, and was lower in DS than in NDS, but higher than that in ND and D on 330 d. At the beginning, the NH $_4^+$ -N content in the sediment was very low, and it remained at this level until day 75; with the arrival of summer, the deposition of SP significantly increased the NH $_4^+$ -N content in the sediment, making the NH $_4^+$ -N content in NDS and ND significantly

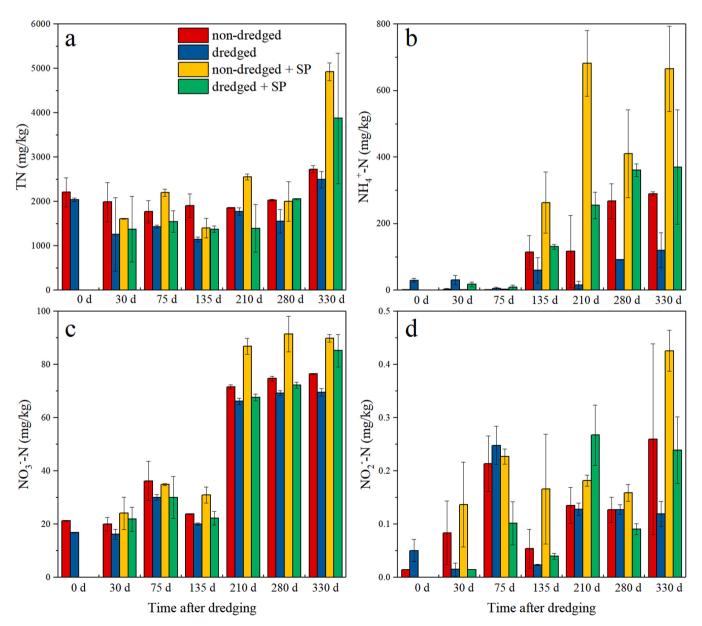


Fig. 3. TN (a), NH₄⁺-N, (b) NO₃⁻-N (c) and NO₂⁻-N (d) content in the surface sediment.

higher than that in DS and D, respectively, with that in NDS being higher than that in ND (P < 0.05). The NO $_3$ -N content in the sediment was higher in summer and autumn. NH $_4$ -N and NO $_3$ -N were the primary forms of inorganic N in the sediment, whereas the proportion of NO $_2$ -N was less than 1% (Table S2). NO $_3$ -N was dominant in winter and spring (0–75 d), and NH $_4$ -N in summer and autumn. During our experimental period, dredging and SP generally significantly affected TN, NH $_4$ -N, NO $_3$ -N and NO $_2$ -N content in the sediments, when the influence of the seasons were not considered (Table S3).

A pore water profile helps in better understanding N mineralization in sediments (Golterman, 2004; Johnston et al., 2009; Xu et al., 2012). As the concentration of NO₃-N and NO₂-N in pore water is significantly lower than that of NH₄⁺-N, we only examined the NH₄⁺-N distribution in pore water during the incubation period (Fig. 4) (Zhang et al., 2013). The NH₄-N concentration in the pore water at the initial stage after dredging was higher than that in the non-dredged group, and this was the same under SP deposition. At day 75, NH₄-N concentration in the pore water of the treatments ND and NDS was higher than that of treatments D and DS, and its concentration increased with depth. With the onset of summer (135–210 d), the concentrations of NH₄⁺-N in the deep layers increased rapidly, gradually spreading to the surface layer. The rate of increase of NH₄⁺-N in treatment NDS was higher than that in treatment ND. There was a delay in the increase of NH₄⁺-N in the pore water after dredging. At this time, the concentration of NH₄⁺-N in the pore water of treatments D and DS was still significantly lower than that in treatments ND and NDS (P < 0.05), whereas there was no significant difference between that in treatment ND and treatment NDS; and treatments D and DS (P > 0.05), respectively. However, at days 280 and 330, there was no significant difference in NH₄-N concentration in the pore water between ND and D (P > 0.05), whereas that of treatment NDS was significantly higher than that of treatment DS (P < 0.05).

The concentration of NH $_4^+$ -N in the sediment and pore water in treatment D was higher than that in treatment ND just after dredging (P < 0.05) (Figs. 3b and 4), which could be attributed to the exposure of deep anaerobic sediment after dredging; anaerobic sediment helps to generate and store NH $_4^+$ -N (Golterman, 2004). However, after 75 d, the NH $_4^+$ -N concentration in the sediment and pore water of treatment D

remained low, indicating that the N mineralization in the sediment was inhibited after dredging. Previous studies demonstrated that the structure of microbial communities in sediment changed after dredging (Yu et al., 2016a; Liu et al., 2017), and the total microbial activity and functional diversity of microbes decreased significantly (Zhong et al., 2010b). Herein, the LOI content in the dredged sediment was lower than that in the non-dredged sediment (Table S4). Kleeberg and Kohl (1999) showed that due to the early diagenesis of the sediment, the bioavailability of OM in the dredged sediment was lower than that of non-dredged sediment. These results together could lead to the inhibition of N mineralization in dredged sediment.

The continuous deposition of SP significantly increased the concentration of NH₄⁺-N in the surface sediments (0–2 cm; Fig. 3b; Table S3) and in the pore water of the DS and NDS treatments (Fig. 4). After 75 d, the NH₄⁺-N content in the SP increased rapidly due to the accumulation and decomposition of fresh OM (Fig. 1). In addition, LOI in the SP was considerably higher than that in surface sediments (Fig. 1b), as the SP would have brought fresh OM to the surface sediment. In seasons with higher temperatures, the mineralization and decomposition of fresh OM could also produce a large amount of NH₄-N, releasing it into pore water (Golterman, 2004). After 75 d, with the continuous sedimentation of SP, the DO concentration at the SWI of the DS and NDS treatments gradually decreased ($< 2 \text{ mg L}^{-1}$), and the OPD also began to decrease (< 3 mm) (Fig. 2). Such anaerobic environments can inhibit the nitrification and promote remineralization of organic N, which is conducive to NH₄⁺-N accumulation in the surface sediments and pore water. In addition to the N remineralization, dissimilatory nitrate reduction to ammonium (DNRA) can compete with denitrification for nitrate to produce bioavailable NH₄⁺-N in the waterbody (Nizzoli et al., 2010). Although we did not measure the DNRA rate in this study, according to previous study (McCarthy et al., 2007), the DNRA rate in Meiliang Bay of Taihu Lake is relatively low compared with the denitrification rate and thus is not the main nitrate removal pathway. Notably, that the role of DNRA on the formation and accumulation of NH₄+N in sediments and pore water should not be ignored during our experimental period (i.e., 135-280 d) because the high algal biomass and low nitrate level can promote DNRA and inhibit N₂ production under serious cyanobacterial blooms (Jiang

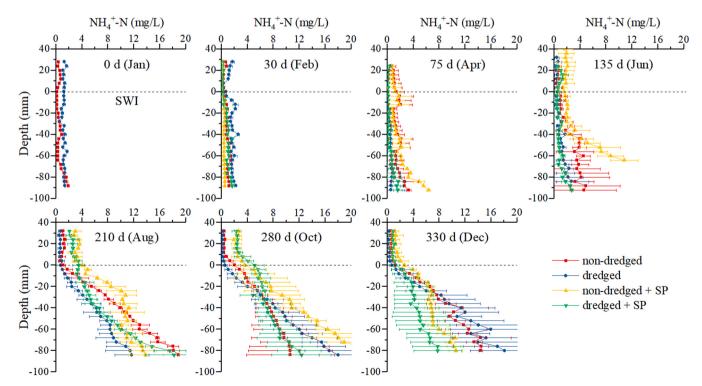


Fig. 4. Vertical profile of NH₄⁺-N concentration across the sediment–water interface.

et al., 2020).

3.3. Response of inorganic N flux across the SWI to dredging and SP

Overall, when the whole experimental period was considered, we found that the effects of dredging and SP were generally significant regarding the flux of NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N after the seasonal variations were considered (Table S5). Immediately after dredging, the NH₄-N from treatment D was released from the sediment to the overlying water (Fig. 5a), caused by the exposure of deep sediment that is high in NH₄-N to the surface layer. At that time, the sediment of treatment ND acted as a sink for NH₄-N, following which NH₄-N was released from the sediment to the overlying water, and its diffusion flux gradually increased. The NH₄⁺-N flux in treatment D remained low after dredging, and its annual average value was 27.2 mg m⁻² d⁻¹, which was lower than that in treatment ND. Under the influence of SP, the diffusion flux of NH₄⁺-N at the SWI increased rapidly. The NH₄⁺-N flux in treatment NDS reached a maximum value of 268.9 mg m⁻² d⁻¹ on day 135, and that in treatment DS reached a maximum value of $221.4 \text{ mg m}^{-2} \text{ d}^{-1}$ on day 280. The NH₄⁺-N flux in treatments NDS and DS was significantly higher than that in treatments ND and D, respectively. The difference in the NH₄⁺-N flux between treatments NDS and DS gradually decreased with time. The NH₄⁺-N flux at the dredged SWI with the deposition of SP was higher than that at the dredged SWI without SP deposition, and even higher than that at the non-dredged SWI with time, suggesting that the deposition of external SP could promote NH₄-N flux at the dredged SWI.

The NO $_3$ -N flux at the SWI was characterized by a release from the sediment to the overlying water in summer and autumn (135–280 d), and the diffusion from overlying water to the sediment in spring (30–75 d) and winter (0 d, 330 d) (Fig. 5b). The diffusion flux of NO $_3$ -N in treatment ND ranged from -24.9–15.1 mg m $^{-2}$ d $^{-1}$. A source–sink transformation occurred, with NO $_3$ -N diffusion from spring to summer, and dredging delayed this process until summer. Compared with the flux of NH $_4$ -N and NO $_3$ -N, that of NO $_2$ -N was relatively small (Fig. 5c), and its flux in treatments ND, D, NDS, and DS ranged from -0.8–1.3, -1.8–0.7, -0.6–5.0, and -0.2–6.4 mg m $^{-2}$ d $^{-1}$, respectively. The sediment acted as an NO $_2$ -N sink in winter and spring and as a source in summer and autumn. However, SP deposition changed the NO $_2$ -N flux on day 330, which was significantly (P<0.05) higher than the average NO $_2$ -N flux in treatments ND or D and had a different direction of diffusion.

The release of DIN from sediments can continuously supplement bioavailable N for the overlying water, and this plays an important role in maintaining primary productivity and the food chain structure of water bodies. The DIN flux at the SWI primarily results from the

interaction of multiple N transformation processes. In this study, the NH₄⁺-N flux was the dominant DIN flux at the SWI, and the main phenomenon was the upward flux, that is, the sediment was released to the overlying water. The average NH₄⁺-N flux in this study was similar to that obtained in Meiliang Bay in our previous study (Zhong et al., 2018), but slightly higher than that reported in other lakes (Gardner et al., 2001; Liu et al., 2018; Petranich et al., 2018). This result is related to the eutrophication degree and sampling time. Compared with the NH₄-N flux, the NO3-N and NO2-N fluxes were relatively small, and they showed a negative flux many times during the test, i.e., the overlying water diffused to the sediment. The NH₄⁺-N flux of the other treatments, except for treatment D, had noticeable seasonal variations; it was higher in summer and autumn. The seasonal variation pattern of the NH₄-N flux was consistent with the NH₄⁺-N concentration in the surface sediment and pore water (Figs. 3b, 4, and 5a). Our previous studies have shown that there was a significant positive correlation between the NH₄⁺-N flux and temperature in the sediment, which indicates that the seasonal variation of the NH₄⁺-N flux was mainly driven by temperature (Liu et al., 2017; Zhong et al., 2018). In addition, we found that the

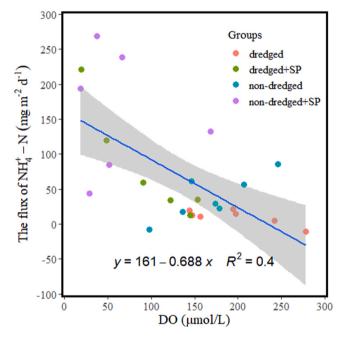


Fig. 6. Relationship between NH₄⁺-N flux and DO at the sediment—water interface.

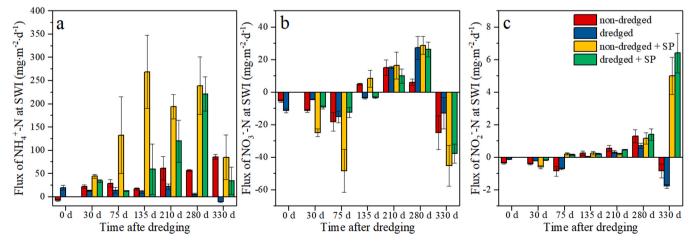


Fig. 5. Diffusion flux of NH₄⁺-N (a), NO₃⁻-N (b), and NO₂⁻-N (c) at the sediment–water interface.

NH₄⁺-N flux had a significant negative correlation with DO concentration at the SWI (Fig. 6). This implies that a rapid decomposition of OM caused an increase in DO consumption during the incubation period; such an anaerobic environment could cause NH₄⁺-N produced by OM decomposition to be more easily released to pore water, thereby increasing the NH₄⁺-N flux at the SWI (Malecki et al., 2004; Kreling et al., 2016; Liu et al., 2017).

The NH₄-N flux at the SWI increased immediately after dredging (Fig. 5a), which could be attributed to the high NH₄⁺-N content in the dredged sediment and pore water just after dredging (Figs. 3b and 4); this is also consistent with the results from our previous study (Zhong et al., 2018). However, over the course of the study, the NH₄⁺-N flux of treatment D remained significantly low, suggesting that dredging could help control the release of NH₄⁺-N in the long term (Yu et al., 2016a). Interestingly, in the presence of SP, the NH₄-N flux of treatment DS increased gradually in summer and autumn, suggesting that SP could significantly increase the NH₄+N flux at the SWI. Compared with dredging, SP were the main regulators of the NH₄⁺-N flux at the SWI after the seasonal variations were accounted for (Table S5). On the one hand, due to the high content of NH₄⁺-N in SP, the deposition of SP increases the NH₄⁺-N content in the sediments; on the other hand, the deposition of SP brings fresh OM to the sediments, and the decomposition of fresh OM also produces NH₄⁺-N, leading to an increase in the NH₄⁺-N concentration in surface sediments and pore water and a corresponding increase in the NH₄⁺-N flux at the SWI. Liu et al. (2019) also found that riverine suspended particles in the estuary of Chaohu Lake included finer particles than surface-sediment, enabling the suspended particles to absorb large amounts of water-soluble OM; the decomposition of the dissolved OM could cause a significant increase of NH₄-N in the sediment and pore-water, and subsequently in the NH₄⁺-N flux at the SWI.

3.4. Response of denitrification, anammox, and N fixation at the SWI to dredging and SP

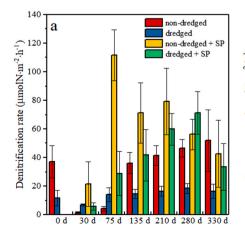
Fig. 7 shows the denitrification, anammox, and N fixation rates of the sediments. The annual average denitrification rate of treatment ND was $31.3\pm18.6~\mu mol~N~m^{-2}~h^{-1}.$ After dredging, the denitrification rate significantly decreased (Table S6) to an annual average of $14.2\pm3.5~\mu mol~N~m^{-2}~h^{-1};$ there was no fluctuation throughout the year (Fig. 7a). SP deposition caused a significant increase (Table S6) in the denitrification rates such as for treatments NDS and DS; the rates in treatments NDS and DS increased to a maximum on days 75 and 280, respectively, followed by a gradual decrease (Fig. 7a). The annual average denitrification rates for treatments NDS and DS were 63.8 ± 28.5 and $40.2\pm21.2~\mu mol~N~m^{-2}~h^{-1},$ respectively.

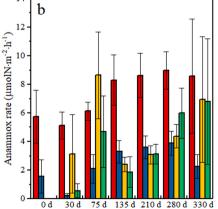
The annual average anammox rates in the four treatments ND, D, NDS, and DS were 7.4 ± 1.5 , 2.4 ± 1.2 , 4.8 ± 2.3 , and

 $3.8\pm2.2~\mu mol~N~m^{-2}~h^{-1}$, respectively. Dredging significantly reduced the anammox rate of the sediments (Table S6); however, SP deposition caused a gradual increase for treatment DS, and a reduction in the rate for treatment NDS, except for that on day 75 (Fig. 7b). Both dredging and SP deposition caused an increase in the proportion of denitrification in N removal, whereas the proportion of anammox decreased from 30.5% to approximately 10% (Table S7).

The N fixation rates of the sediment in the four treatments ND, D, NDS, and DS were 0.80 ± 0.10 , 0.10 ± 0.01 , 0.9 ± 0.02 , and 0.4 ± 0.10 µmol N m $^{-2}$ h $^{-1}$, respectively (Fig. 7c). The N fixation rate decreased significantly after dredging (Table S6), and there was no fluctuation throughout the year. SP deposition caused an increase in the rate after dredging, and there were significant joint effects of SP deposition and dredging on the N fixation rates (Table S6). Generally, the N fixation rate was relatively low compared to that of the other processes of denitrification or anammox and accounted for $\sim\!2\%$ of the N removal.

Denitrification is the primary N removal mechanisms in a lake ecosystem. In this process, nitrate or nitrite is used as electron acceptor, and organic carbon as electron donor to produce N₂, NO, or N₂O, which could effectively reduce its N content, thereby mitigating eutrophication and regulating primary productivity of the water body (Seitzinger, 1988). Our previous study has indicated that the denitrification rates in Meiliang Bay are co-regulated by water column temperature and nitrate concentration (Zhong et al., 2020). In this study, we used the same lake water for different treatments in determining the denitrification rate and kept the analysis conditions consistent. Therefore, the influence of dredging and SP on denitrification rate should be attributed to their influence on sediment properties. During the experiment, the denitrification rate in treatment D has been maintained at a relatively low level, indicating that the denitrification rate of dredged sediments has been inhibited, which is consistent with our previous studies (Zhong et al., 2010a; Yu et al., 2016b). We also found that the denitrification rate of non-dredged sediment is more sensitive to temperature changes than that of dredged sediment; it is limited by organic carbon content in dredged sediment (Zhong et al., 2010a). When the experiment lasted for 75 d, the continuous deposition of SP significantly increased the denitrification rate in treatment DS (Fig. 7a), and the statistical results of GLMM also showed that SP was the main regulator of the denitrification rate (Table S6). Compared with the surface sediments, SP composed of more fine particles and a higher content of OM, organic carbon, N and phosphorus (Eadie et al., 1984; Meyers et al., 1984; Liu et al., 2019). On the one hand, SP deposition can bring fresh OM to the dredged sediment, which can change the situation that the denitrification rate in dredged sediment is limited by organic carbon. On the other hand, SP deposition can accelerate the recovery of microbial community structure (Liu et al., 2017), which can significantly improve the denitrification rate of dredged sediment.





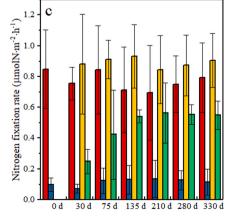


Fig. 7. Denitrification (a), anammox (b) and nitrogen fixation (c) rates at the sediment-water interface.

Anammox is another important N removal process in aquatic environment, and in this process, anammox bacteria oxidize ammonia by using nitrite as the electron acceptor to produce N2, it has recently been recognized as an important sink for fixed inorganic N in lacustrine ecosystem (Schubert et al., 2006). In the present study, the statistical results of GLMM showed that the anammox rate was significantly affected by dredging treatment, and the anammox rate in treatment D was significantly lower than that in treatment ND (Fig. 7b; Table S6), which could be attributed to a change in sediment properties after dredging. In this study, the water content and OM content of sediments decreased significantly (P < 0.05) after dredging and were not restored to the status recorded before dredging within our experimental period (Tables S3 and S4). Previous studies have shown that there is a decrease in the activity and functional diversity of microbes in dredged sediments and also a change in the abundance and diversity of N cycling microbes (Zhong et al., 2010b; Yu et al., 2016a). The interaction of physical, chemical and biological factors can regulate the N transformation in sediments (Gardner et al., 2001). However, SP deposition did not significantly increase the anammox rate (Fig. 7b; Table S6), and this result imply that the recovery of sediment properties, especially anammox bacteria, requires a long-term process.

N fixation involves transforming dinitrogen gas into bioavailable ammonium, which could possibly increase N content in a lake ecosystem (Howard et al., 1970; Scott and McCarthy, 2010; Gettel et al., 2013). N fixation is the only process that can directly offset the N loss due to denitrification and anammox (Minjeaud et al., 2009). Previous studies have indicated that several factors including nutrients and light availability, micronutrients and excessive turbulence could regulate biological N fixation (Paerl et al., 1987; Bell et al., 1999). In this study, dredging had a significant effect on the N fixation rate in sediment, resulting in the N fixation rate of treatment D being less than that in treatment ND (Fig. 7c; Table S6). This result can be attributed to the influence of sediment dredging on sediment properties. Our previous study have indicated that the sediment N fixation rate was significantly related to the water content, TN content and total phosphorus content of the sediments in Taihu Lake (Yao et al., 2018). Therefore, the differences in the properties of dredged and non-dredged sediments lead to significant differences in N fixation rates. The deposition of SP can significantly increase the nitrogen fixation rate of sediments (Fig. 7c; Table S6), which is attributed to the gradual restoration of sediment properties under the continuous sedimentation of SP (Fig. 3; Table S4).

The annual average rates of denitrification and anammox in treat- $63.8 \pm 28.5 \ \mu mol \ N \ m^{-2} \ h^{-1}$ were $4.8 \pm 2.3 \, \mu mol \, N \, m^{-2} \, h^{-1}$, respectively (Fig. 7), which were close to those reported by McCarthy et al. (2007) and Xu et al. (2009) for the SWI in Taihu Lake. In treatment NDS, denitrification and anammox contributed 92% and 8% to N removal, respectively (Table S7), suggesting that denitrification was the most important N removal process in Taihu Lake (Zhong et al., 2020; Zhu et al., 2020). The annual average rate of N fixation in treatment NDS was 0.89 \pm 0.18 $\,\mu mol\,$ N $\,m^{-2}\,$ $\,h^{-1}.$ This value is similar to the previous studies on lake and coastal systems (Eyre et al., 2011; Gettel et al., 2013). Herein, the N fixation accounted for only approximately 2% of the N removal. These results suggest that N fixation could not substantially influence the N budget at the SWI, which is consistent with the results of Yao et al. (2018), showing N fixation in Taihu Lake compensated for only approximately 1.8% of the N loss through denitrification.

3.5. Effect of dredging and SP on N budget at the SWI

Table 1 quantifies the preliminary N budget at the SWI. The annual N removal in Meiliang Bay was 1946.2 t; it decreased to 642.1 t after dredging. With SP deposition, N removal increased to 6906.9 and 3635.2 t in treatments NDS and DS, respectively. Fig. 8 shows the contribution of each N removal process toward N loss in sediments; dredging promoted denitrification and impeded DIN diffusion, whereas

Table 1

Preliminary N budget at the sediment–water interface (Dn: denitrification rate; An: anammox rate; DIN flux: diffusion flux of DIN across SWI; N fixation: N fixation rate; Sum: the sum of Dn, An, DIN flux, and N fixation; N budget: N budget at the SWI of Meiliang Bay throughout the year in tons (t); the area of Meiliang Bay was $129 \, \mathrm{km}^2$, and the number of days was 365. A positive value represents the removal of N from the sediment and a negative value represents the introduction of N into the sediment).

μmol N m ⁻² d ⁻¹	Non- dredged (ND)	Dredged (D)	Non- dredged+SP (NDS)	Dredged+SP (DS)
Dn	751.8	339.6	1530.0	963.7
An	176.6	58.0	114.3	92.1
DIN flux	2019.8	571.7	8774.4	4427.8
N fixation	18.5	2.8	21.4	11.5
Sum	2929.6	966.6	10,397.3	5472.2
N budget (t)	1946.2	642.1	6906.9	3635.2

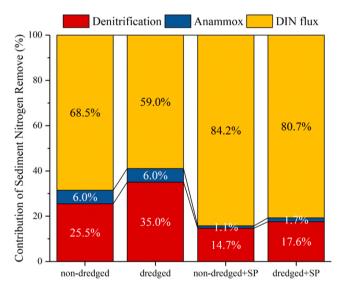


Fig. 8. Contribution of nitrogen removal processes toward N loss in sediment.

SP deposition impeded microbial denitrification and anammox, and promoted DIN diffusion.

Inorganic N flux contributed the most to the N budget at the SWI, accounting for 60-84% of various treatments (Table 1), followed by denitrification and anammox. As stated earlier, N fixation showed minor contributions to N budget at the SWI. Dredging and SP can regulate the N budget at the SWI by significantly influencing the N transformation processes at the SWI. Dredging substantially reduced N loss at the SWI, caused mainly due to a decrease in inorganic N flux, followed by a decrease in the N removal. However, continuous SP deposition could strongly increase N loss in treatments NDS and DS. The results suggest that SP play a critical role in the N budget at the SWI. The contribution of various N cycle processes towards N loss at the SWI was influenced by dredging and SP deposition. In the absence of SP deposition, the contribution of inorganic N flux in treatment D was reduced by 9.5% as compared to that in treatment ND (Fig. 8); accordingly, the contribution of microbial N removal increased by 9.5%. However, in the presence of SP, the contribution of inorganic N flux for treatment NDS and treatment DS increased significantly, reducing the difference in the contributions of inorganic N flux between treatment ND and treatment D (Fig. 8). This suggests that SP helped in accelerating the recovery of N cycle processes after dredging; thereafter, the N budget at the SWI could gradually recover to the state before dredging.

3.6. Implications toward restoration of shallow eutrophic lakes

Geoengineering techniques such as sediment dredging and sediment capping has been extensively used in managing lake eutrophication (Lürling and Faassen, 2012; Spears et al., 2013; Waajen et al., 2016). Geoengineering techniques primarily aims to mitigate nutrient release from sediment to the overlying water, helping in reducing the concentration of bioavailable nutrients in the water column, thereby mitigating eutrophication, and maintaining normal ecological functions of a water body (Lürling and Faassen, 2012; Waajen et al., 2016). Sediment dredging has been widely used in internal loading management (Ryding, 1982; Does et al., 1992). Sediment capping is a promising alternative to sediment dredging because of its low cost. In-situ capping of sediment can create an active barrier between overlying water and contaminated sediment, thereby reducing the nutrient release at the SWI (Lürling et al., 2016). Sediment capping employs efficient adsorption materials to control the internal phosphorus loading. Aluminum- and/or lanthanum-modified compounds are the most effective phosphorus adsorption compounds (Lürling et al., 2016), whereas natural and modified zeolites can reduce the release of ammonium from sediments (Lin et al., 2011; Gu et al., 2019). In addition, sediment capping is mainly applied to small water bodies, such as urban lakes and ponds (Spears et al., 2016), whereas sediment dredging has been widely used in large water bodies, such as large shallow lakes (Chen et al., 2018; Jing et al., 2019; Li et al., 2020). The present study demonstrates that sediment dredging effectively reduces the release of inorganic N (mainly NH₄⁺-N) at the SWI, which could be advantageous. However, after dredging, N removal via denitrification and anammox was significantly hindered simultaneously, suggesting that more bioavailable N could be retained in the sediment; this could be disadvantageous for eutrophic lakes. Therefore, sediment dredging has benefits and drawbacks in managing internal N loading in eutrophic lakes.

Continuous SP deposition could accelerate the recovery of N cycle processes after dredging, thereby undermining the long-term efficacy of dredging on internal N loading, bringing huge challenges while managing internal N loading in turbid phytoplankton-dominated shallow lakes. Our previous study found that SP could also significantly increase phosphorus flux at the SWI, resulting in a rapid recovery of P loading after dredging, undermining the long-term efficacy of dredging on internal P loading (Liu et al., 2019; Wen et al., 2020). In practice, to maintain the effectiveness of sediment dredging on internal loading, measures based on in-lake and watershed should be undertaken to inhibit the deposition of SP during and after dredging. The general consensus is that managing external loading is the premise for controlling internal loading (Kleeberg and Kohl, 1999; Reddy et al., 2007; Hamilton et al., 2016). Therefore, it is necessary to reduce external input at the watershed scale; the input of external particles can be blocked by constructing a forebay, buffer-zone, or lakeside wetland, especially at the estuary/confluence of lakes. Additionally, when dredging is undertaken, sediment resuspension must be mitigated as much as possible (Bridges et al., 2010). On the one hand, sediment resuspension will cause nutrient release, and on the other hand, it will increase the sedimentation of particulate matter. In addition, the selection of dredging equipment for dredging engineering should minimize the amount of dredging residuals (Bridges et al., 2010; Patmont et al., 2018), as the residuals can influence the control effect of dredging on the internal loading in both short-term and long-term scales (Liu et al., 2019; Chen et al., 2020). After dredging, silt-curtains or enclosures could be installed to inhibit sediment resuspension and deposition of particles from the non-dredged area (Palermo and Hays, 2014). Moreover, the resuspension of sediment after dredging can be prevented by the restoration of macrophytes; during algal blooms, algal harvesting can reduce the deposition of algal debris. Generally, the findings of this study indicate that in shallow lakes with high productivity, particulate matter deposition will diminish the long-term effect of dredging on internal N loading, and therefore, frequent dredging could help in maintaining a sustained control on internal loading.

4. Conclusions

- (1) Dredging helps reduce OM and TN content in surface sediment; improves the redox environment at the SWI; slows down the rate of N mineralization, denitrification, anammox, and N fixation after dredging; and alters the N budget at the SWI and the contribution of various N cycle processes.
- (2) Fresh OM, TN and inorganic N were rich in SP; their continuous deposition could accelerate the recovery of N cycle processes at the SWI, thereby reducing the differences in the N budget and contributions of various N cycle processes caused by dredging.
- (3) Dredging has both advantages and disadvantages in controlling internal N loading in turbid shallow lakes, i.e., it helps reduce inorganic N flux (mainly NH⁺₄-N) at the SWI, but N removal through denitrification and anammox at the SWI is also hindered. To maintain a sustained control of internal N loading through sediment dredging, measures should be taken based on in-lake and watershed to inhibit the input and settlement of particulate matter.

CRediT authorship contribution statement

Jicheng Zhong: Conceptualization, Writing - original draft, Writing - review & editing, Validation, Methodology, Funding acquisition, Project administration. Shuailong Wen: Investigation, Methodology, Data curation, Formal analysis, Visualization, Writing - original draft. Lu Zhang: Resources, Methodology, Jianjun Wang: Formal analysis, Writing - review & editing. Cheng Liu: Resources, Methodology, Conceptualization. Juhua Yu: Conceptualization, Writing - review & editing. Lei Zhang: Resources, Methodology. Chengxin Fan: Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2020.124691.

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