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Evaluation of *in situ* simulated dredging to reduce internal nitrogen flux across the sediment-water interface in Lake Taihu, China[☆]



Juhua Yu^a, Chengxin Fan^a, Jicheng Zhong^{a,*}, Yinlong Zhang^b, Changhui Wang^a, Lei Zhang^a

^a 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, PR China

^b Key Laboratory of Forestry Ecological Engineering of Jiangsu Province, Nanjing Forestry University, Nanjing 210037, PR China

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ABSTRACT

Sediment dredging is considered an effective restoration method to reduce internal loading of nitrogen (N) and phosphorus (P) in eutrophic lakes. However, the effect of dredging on N release from sediments to overlying water is not well understood. In this study, N exchange and regeneration across the sediment-water interface (SWI) were investigated based on a one-year simulated dredging study in Lake Taihu, China. The results showed low concentrations of inorganic N in pore water with low mobilization from the sediments after dredging. The calculated fluxes of NO_3^- -N from post-dredged sediments to overlying water significantly increased by 58% ($p < 0.01$), while those of NH_4^+ -N dramatically decreased by 78.2% after dredging ($p < 0.01$). N fractionation tests demonstrated that the contents and lability of N generally declined in post-dredged sediments. Further high-throughput sequencing analysis indicated that relative abundance of the bacterial communities decreased, notably by 30% (compared with undredged sediments). The estimated abundance of *Nitrospira* enhanced, although the relative abundance of *Thiobacillus*, *Sterolibacterium*, *Denitratisoma*, *Hyphomicrobium*, *Anaeromyxobacter* and *Caldithrix* generally declined after dredging. Therefore, dredging reduced N mobilization from the sediments, which primarily due to decreases in N mobility, in organic matter (OM) mineralization potential and in the bacterial abundance of post-dredged sediments. Overall, to minimize internal N pollution, dredging is capable of effectively reducing N release from sediments. In addition, the negative side effect of dredging on removal of NO_3^- -N and NO_2^- -N from aquatic ecosystems should be paid much more attention in future.

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1. Introduction

Eutrophication has become a serious environmental problem worldwide which has caused a series of threats to aquatic

Abbreviations: N, nitrogen; TN, Total nitrogen; IEF-N, ion exchangeable nitrogen; CF-N, carbonate nitrogen; IMOF-N, iron-manganese oxides nitrogen; OSF-N, organic matter-sulfide nitrogen; SWI, sediment-water interface; OM, organic matter; AVS, acid volatile sulfide; Anammox, anaerobic ammonium oxidation; DNRA, dissimilatory nitrate nitrogen reduction to ammonium; Rhizon SMS, rhizon soil moisture samples.

^{*} This paper has been recommended for acceptance by Min Liu.

^{*} Corresponding author.

E-mail addresses: juhuayu1984@gmail.com (J. Yu), cxfan@niglas.ac.cn (C. Fan), jjczhong@niglas.ac.cn (J. Zhong), ecoenvylz@163.com (Y. Zhang), chwang@niglas.ac.cn (C. Wang), leizhang@niglas.ac.cn (L. Zhang).

ecosystems including frequently harmful algal bloom, hypoxia and fish kills, taste and odor problem as well as degradation of drinking water supplies (Qin and Zhu, 2006; Jin, 2003; Smith and Schindler, 2009). In addition to phosphorus in Lake Taihu (the third largest lake in Chinese), N which is a key growth-limiting elements for toxic *Microcystis* blooms during the summer, can limit the lake's primary productivity and deteriorate lake water quality (Smith and Schindler, 2009; Xu et al., 2010; Abell et al., 2010). Sediment is generally regarded a major N source for overlying water, which is able to keep lake in a eutrophic state for several decades (Jiang and Shen, 2006; Azevedo et al., 2015). To minimize internal pollution load, many approaches have been applied to improve lake water quality (Pan et al., 2012; Yin et al., 2013; Zhang et al., 2015). Sediment dredging serves as an effective restoration method and is used to permanently remove surface contaminated sediments from aquatic ecosystems (Gustavson et al., 2008). However, the

effectiveness of dredging is still debatable because N loading reversion of lake water was usually observed after dredging, for reasons that are not clearly concluded in the literature (Gustavson et al., 2008; Fan et al., 2004; Pu et al., 2000).

To date, literature regarding the effects of dredging on nutrient removal are rather scarce compared with the pollutants of persistent organic pollutants (POPs) and heavy metals (Zhong et al., 2008; Yenilmez and Aksoy, 2013; Recknagel et al., 1995). Unfortunately, numerous studies reported that dredging was able to effectively control internal phosphorus release (Kleeberg and Kohl, 1999; Reddy et al., 2007). Whereas the effects of dredging on internal N release from published studies were usually equivocal, and most investigations just focused on ammonium release (Fan et al., 2004). In such case, some field investigations (Wang and Feng, 2007) and laboratory simulation (Spencer et al., 2006; Zhong et al., 2009; Yu et al., 2012) have been conducted by monitoring N concentrations in pore water and overlying water to evaluate temporary and long-term effects of dredging on N removal via flux calculations or instantaneous measurements alone (Wang and Feng, 2007). Most previous studies generally showed that dredging could lead to ammonium nitrogen ($\text{NH}_4^+\text{-N}$) release to overlying water temporarily, while some effectively reduced its release (Fan et al., 2004). Furthermore, few studies demonstrated long-term release risk was low and reported slight effects based on the fluxes of $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ (Yu et al., 2012; Lohrer and Wetz, 2003; Jing et al., 2013).

In principle, regeneration and transfer processes of N across the SWI are crucial to N release from sediments to water column, more specifically for the $\text{NH}_4^+\text{-N}$ release (Wu et al., 2001). Theoretically, two basic processes by which N can be released are the diffusion process of pore water N based on concentration gradients between pore water and overlying water, and the regeneration process from sediments (Wu et al., 2001; Cornwell and Owens, 2011; Ding et al., 2015). In shallow eutrophic lakes, the main determining factors can alter the exchange and regeneration of N across the SWI including the contents of OM (Yang et al., 2010), dissolved oxygen (DO), redox potential (Jing et al., 2013), pH (Medina et al., 2003), temperature, salinity, light (Wu et al., 2012), mineral clay lattices (Kemp and Mudrochova, 1972), cation exchange (Gardner et al., 1991), sediment grain size (Zhong et al., 2009), sediment accumulation rates (Cornwell and Owens, 2011), bioturbation/bioadvection and microbial nitrogen transformation (Graca et al., 2004; Yu et al., 2016). However, it is so far a formidable challenge to distinguish the effectiveness of dredging activities from the complicated environmental effects of natural processes on how much internal loading can be controlled by dredging (Gustavson et al., 2008). This investigation further provided a better way to quantitatively evaluate the contribution of dredging through simulating dredging in lake field conditions closely. Secondly, published studies regarding dredging mainly focused on the diffusion process of N, but little attention has been paid to the regeneration process (Wang and Feng, 2007; Jing et al., 2013; Cabrita, 2014). In particular, most previous studies just focused on the flux of $\text{NH}_4^+\text{-N}$, whereas the exchange of $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ across the SWI are also of significant components to assess the efficacy of dredging in reducing internal N release. In present study, combined the diffusion flux of various inorganic N, stability of different N fractions in sediments and bacterial community structure to investigate the probable mechanism of dredging on controlling N release from sediments. Thirdly, a great multitude of studies involving internal N control just focused on the amounts of TN removal from lake ecosystems (Qin et al., 2006), which was necessary for studying the background value of N in sediments and for large scale of watershed management, but it can not provide valuable information concerning N transfer and mobilization processes across the SWI induced by dredging, neither for N cycling in aquatic ecosystems (Wang et al.,

2008). Taken together, it is vital to study the effects of dredging on N mobility based on N fractionation and microbial transformation processes of N, to further understand the mechanisms responsible for the N migration and regeneration near the sediment-water interface.

After dredging, we hypothesized that the $\text{NH}_4^+\text{-N}$ regeneration ability after dredging would substantially decline, and the instantaneous release of $\text{NH}_4^+\text{-N}$ probably regulated by the flux of pre-existing pore water, adsorbed concentrations and/or flocculent sediments resuspension (Cabrita, 2014). In addition, dredging would lead to a condition that enhanced $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ formation under more oxic condition, while probably weaken them removal from lake ecosystems due to lower denitrification rates. Therefore, the primary objective of this study was to quantitatively evaluate the effectiveness of dredging in reducing internal N pollution loading based on a one-year field simulation study in Lake Taihu, China. Additionally, N fractions and bacterial community structure was identified to further understand the real mechanisms responsible for N transfer and mobilization in sediments after dredging.

2. Materials and methods

2.1. Site description and sampling

Lake Taihu is the third largest freshwater lake in China, with an area of 2338 km² and a mean water depth of 1.9 m. This lake is a major source of drinking water for a population of much more than 20 million people residing in nearby city (Qin and Zhu, 2006). Since the 1990s, black bloom and microcystis algal blooms have periodically occurred with an increasing frequency and severity in sampling sites during the late spring and summer seasons, and it has been spreading to the central and south parts of the lake (Lu and Ma, 2009). Currently, to control and prevent the black bloom occurrence, dredging served as a significant restoration method was applied to this area, and previous laboratory and field results showed that dredging improved the water quality of Lake Taihu (Shen et al., 2011). The authors of this paper have conducted dredging projects since 2009, and have many more dredging operation planned for the researched area. The sampling site (31°24'35.8" N, 120°6'4.6"E) is situated in the northern near shore area of Yueliang Bay (Fig. 1).

A total of 12 intact sediment cores were collected with a large-caliber core sampler (Φ 110 mm, length 500 mm, Rigo Co., Japan) in August of 2013. The cores collected were generally more than 400 mm long, and they were covered with the near-bottom water from the same location. The cores were closed with rubber stoppers in order to avoid sediment oxidation during the sampling and transportation. With this sampling procedure, sediment cores surface 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 *in situ* location using 25 L polyethylene bottles.

2.2. Experimental setup

In the laboratory, twelve sediment cores were treated as follows: the overlying water of all sediment cores were siphoned. Subsequently the top sediment layer (ca. 12 cm) of six cores were removed and carefully transplanted into the Plexiglas cylinders (11 cm inner diameter, 17 cm height) to serve as the undredged treatments. As for dredged treatment, the uppermost 25 cm sediment were discarded according to previous studies in Lake

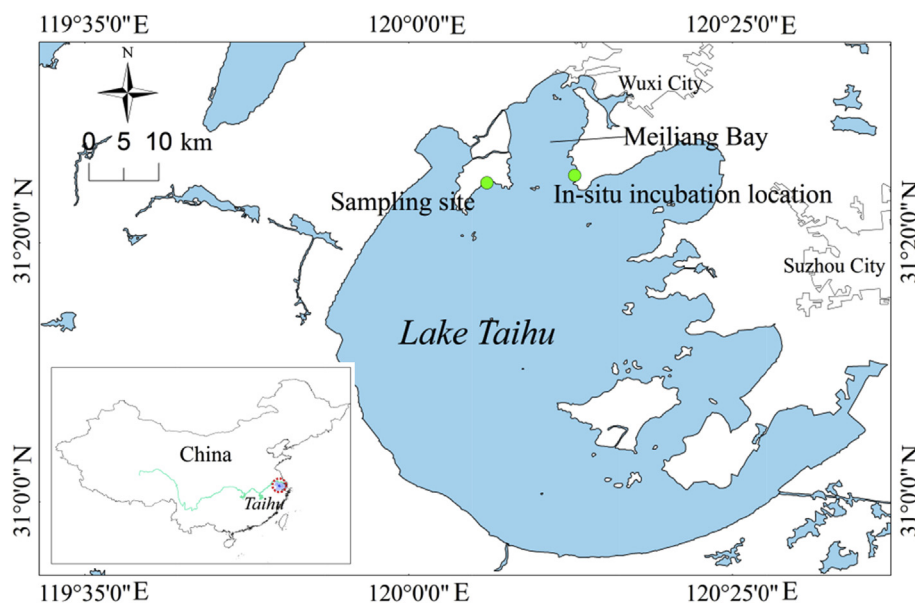


Fig. 1. Sampling site and in-situ incubation location.

Taihu (Fan et al., 2004; Zhong et al., 2008), and the underlying sediment (ca. 12 cm) were filled into the tubes undisturbed as the dredged treatments with six replicates. The overlying water was carefully dropped onto the sediment cores immediately. In order to better mimic the environmental conditions in the lake field more closely, non-dredged cores and post-dredged cores were incubated on the lake bottom, situated at the observation platform of the Taihu Laboratory for Lake Ecosystem Research (TLER) (31°25′10.78″ N, 120°12′50.7″ E). All prepared cores were put into a stainless steel shelving unit with six units for each shelf. The shelving unit was then fastened to the bridge of the observation platform using a nylon line, and was put down to the sediment surface of the lake (ca. 1.5 m). The experimental period was from August 3, 2013 to August 7, 2014. Pore water samples were sampled monthly to measure pore water chemistry from each core (3, 30, 60, 90, 120, 150, 180, 210, 240, 270, 300, 330, 360 d after dredging, respectively). At the end of the experiments, owing to the fact that reductive compounds in the sediments were sensitive to the redox potential and oxygen condition, a total of 12 sediment cores were used to allow analysis of their sediment characteristics and were placed in an anaerobic glove box (Plas Lab Model 818-GB) which had been purged with N₂ gas to obtain the vertical subsamples of sediment cores at 1 cm interval. Subsequently, the sediment subsamples were stored in gas-tight polyethylene bottles and then were freeze-dried and sieved through 0.15 mm pore-size mesh waiting for further analysis. Exceptionally, a total of six subsamples were stored under – 80 °C to identify the bacterial community structure using a high-throughput sequencing technique.

2.3. Collection of pore water sample

Pore water samples were collected monthly by using a non-destructive method to portray the nutrient profiles in interstitial water at different times after dredging. Rhizon soil moisture samplers (Rhizon SMS: Rhizosphere Research Products, The Netherlands) were used to extract successive pore water samples from the intact sediment 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 had been sealed in advance with hydrophobic tape. At each site, an

aliquot of 5 ml pore water was obtained through suction using above samplers. Then, the collected samples were immediately transported to the laboratory within the ice boxes, and the concentrations of NH₄⁺-N, NO₃⁻-N and NO₂⁻-N were measured using a Skalar flow-injection analyzer (Skalar Sanplus, The Netherlands).

2.4. Calculation of N diffusion fluxes

The vertical profiles of NH₄⁺-N, NO₃⁻-N and NO₂⁻-N in pore water were used to estimate the diffusion fluxes across the SWI based on Fick's first law of diffusion (Ullman and Aller, 1982):

$$F = \phi D_s \left. \frac{\partial c}{\partial x} \right|_{x=0} \quad (1)$$

where ϕ is porosity, $(\partial c / \partial x)|_{x=0}$ is the pore water concentration gradient across the SWI. D_s is the sediment diffusion coefficient of NH₄⁺-N, NO₃⁻-N and NO₂⁻-N (with the values of 17.6×10^{-6} , 19.0×10^{-6} and 19.1×10^{-6} cm² s⁻¹, respectively) in pore water using $\phi \geq 0.7$ (Ullman and Aller, 1982).

2.5. Determination of sediment properties

To determine the sediment cores' micro-profiles, oxygen concentration, pH and redox potential profiles were measured using the oxygen, pH and Eh microelectrodes (Unisense, Aarhus, Denmark), respectively. Sediment porosity and bulk density were measured with the use of a cutting ring (Graca et al., 2004). Total Fe in sediments were measured using ICP-AES after fusing 0.05 g (DW) of sediment with 0.2 g LiBO₂, followed by dissolution with 4% HNO₃ (Xu et al., 2012). Active Fe in sediments were extracted using the ammonium oxalate-oxalic acid solution (pH = 3.0) (Danen-Louwerse et al., 1993) and were also determined by ICP-AES. Organic matter content in sediments was measured as loss on ignition (LOI) through calcination of the sediments at 550 °C for 6 h, and the pH value of sediments were determined using pH meter (Jin and Tu, 1990). Acid volatile sulfide (AVS) was measured using the Heih's cold diffusion method (Hsieh et al., 2002).

Genomic DNA was extracted from 0.4 g of fresh surface sediments (0–1 cm) using the bead-beating method and a FastDNA

Table 1
The physicochemical properties of undredged and post-dredged sediments.

Depth (cm)	pH	Porosity (%)	Buck density (g cm^{-3})	LOI (%)	Fe (g kg^{-1})	Fe ²⁺ (g kg^{-1})	AVS ($\mu\text{mol g}^{-1}$)	TN (mg kg^{-1})
Undredged sediments								
-1	5.20	1.003	1.308	9.89	37.4	24.2	40.92	3820
-2	5.51	0.952	1.317	9.91	36.2	20.7	41.53	3740
-3	5.79	0.936	1.385	9.39	35.8	20.6	13.03	3090
-4	6.12	0.862	1.423	7.00	30.2	17.2	6.52	2800
-5	6.39	0.838	1.567	5.81	27.6	16.3	2.20	2500
-6	6.49	0.769	1.613	4.95	26.0	14.8	6.52	2020
-7	6.52	0.753	1.687	4.44	24.4	14.7	7.79	1830
-8	6.53	0.746	1.689	4.30	23.6	14.2	7.61	1820
-9	6.51	0.737	1.723	4.29	23.3	13.8	7.30	1800
Post-dredged sediments								
-1	6.85	0.815	1.491	5.13	28.3	16.8	0.20	1760
-2	6.45	0.770	1.633	4.78	28.2	16.9	0.95	1670
-3	6.37	0.730	1.682	4.55	27.2	17.2	2.61	1580
-4	6.40	0.766	1.768	4.45	27.7	17.2	1.88	1810
-5	6.31	0.741	1.725	4.37	27.4	17.4	2.56	1870
-6	6.54	0.748	1.786	4.48	28.4	16.9	1.26	1710
-7	6.55	0.735	1.769	4.46	27.6	18.2	0.92	1720
-8	6.57	0.724	1.974	4.30	26.7	17.2	1.30	1650
-9	6.53	0.741	1.816	4.25	27.5	17.4	3.23	1730

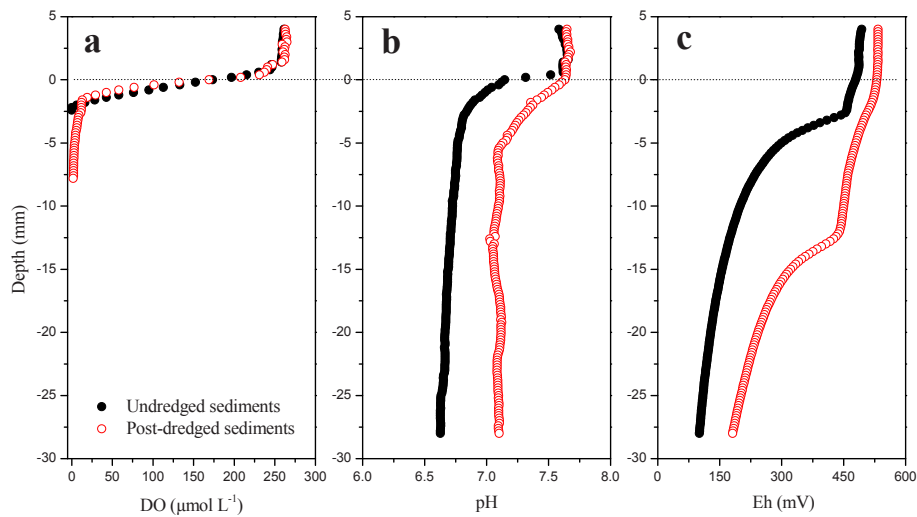


Fig. 2. The depth profiles of DO (a), pH (b) and Eh (c) in undredged and post-dredged sediments at the end of experiment. The location of the sediment-water interface is represented by zero.

SPIN kit for soil (BIO 101, Qbiogene Inc., Carlsbad, CA, USA) according to the manufacturer's instructions, and DNA extracts were then stored at -80°C waiting for analysis. The bacterial community structure was determined using high-throughput sequencing technique. Three subsamples of the upper 1 cm sediments for each treatment were used to determine the bacterial community structure. The DNA extracts were assayed by Personalbio company (Shanghai, China). According to the results provided by the company, we further identified and calculated the relative abundances of the bacterial communities on the basis of published literature. The contents of different N fractions were determined using the sequential extraction scheme (Wang et al., 2008; Ma et al., 2003), and the extracts including $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ forms were determined using a Skalar flow-injection analyzer. The N fractions in sediments were classified as an exchangeable form (IEF-N), carbonate form (CF-N), iron-manganese oxides form (IMOF-N) and organic matter-sulfide form (OSF-N). TN was measured by the alkaline potassium persulphate oxidation method (Jin and Tu, 1990), and the labile N in sediments was extracted using

2 mol L^{-1} KCl and quantified by Skalar flow-injection analyzer (Skalar Sanplus, Netherlands).

2.6. Statistical analysis

The data shown in this study were represented as means and standard deviation of six replicates. The differences of inorganic N concentrations in non-dredged and post-dredged pore water at different depths were tested using two-way repeated analysis of variance (RM-ANOVA) with time as the repeated factor. The differences of inorganic N fluxes across the SWI between non-dredged and post-dredged sediments were tested by one-way repeated analysis of variance (RM-ANOVA) with time as the repeated factor. Similarly, Pearson correlations analysis was used to test the correlation between the N fractions and sediment properties. For the variables of the N forms, bacterial community structure and physicochemical characteristics were recorded for both non-dredged and post-dredged sediments using the Paired-samples T test. All statistical analyses in this study were performed by SPSS 13.0 (SPSS,

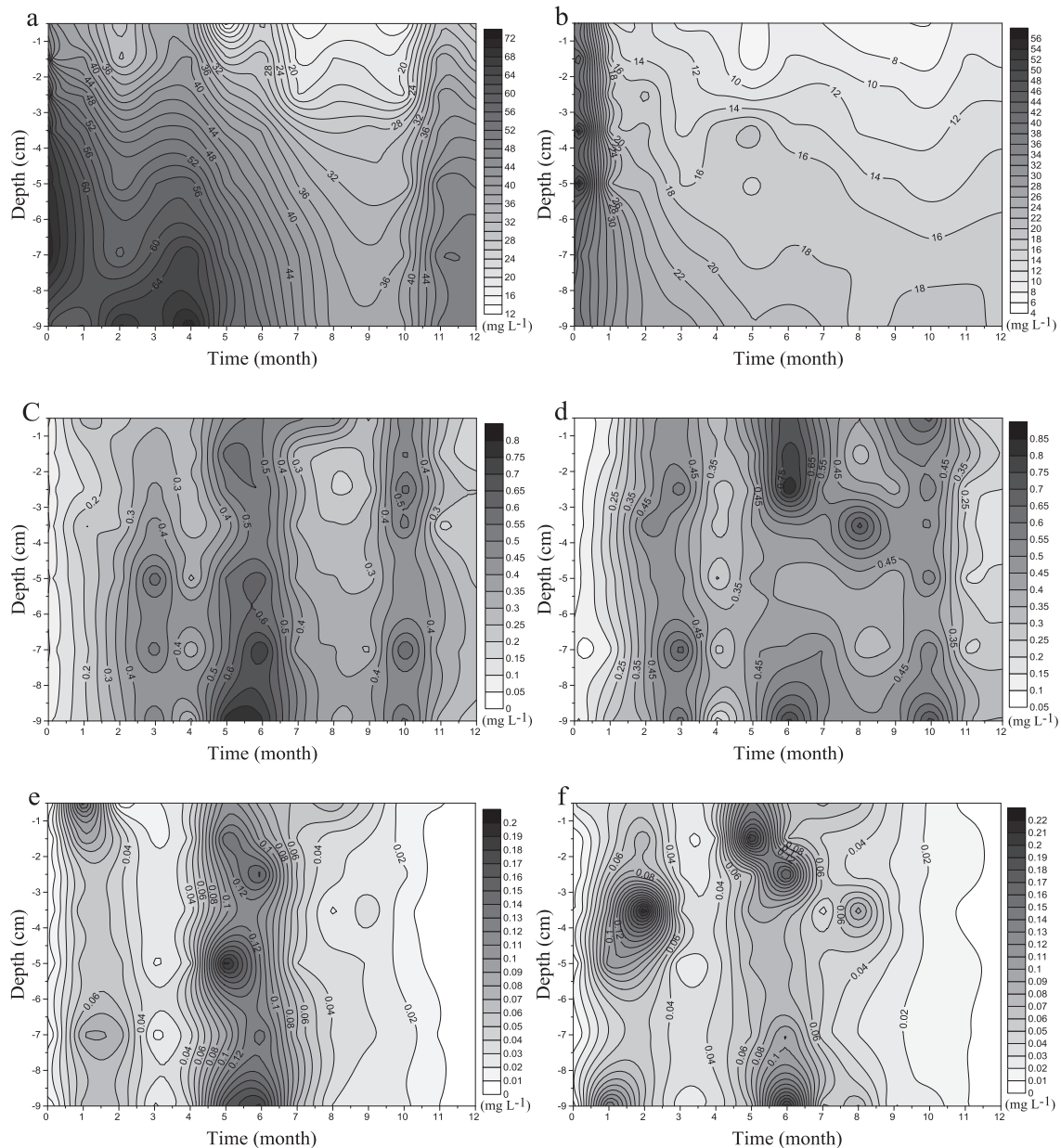


Fig. 3. Pore water profiles of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ in undredged (a, c and e) and post-dredged (b, d and f) after different dredging time, and values are expressed as means \pm SD of three replicates.

USA).

3. Results

3.1. Sediment properties

In general, the values of control sediments was much greater than those of post-dredged sediment, with values of 151%, 112%, 147%, 101%, 106% and 895% of TN, porosity, LOI, Fe, active Fe and AVS, respectively, as compared to those in post-dredged sediments (Table 1). While the bulk density of control sediments was 87.8% of post-dredged sediments. pH values in control sediments were lower than those of treatment sediments for all of the sediment horizons. DO, pH and Eh values in control were lower than those in post-dredged sediments. The oxygen penetration depths by dredging were deeper than those of control sediments, and the

penetration depths were approximately 2 mm and 8 mm for control and post-dredged sediments, respectively (Fig. 2).

3.2. Effects of dredging on pore water chemistry

Pore water $\text{NH}_4^+\text{-N}$ concentrations significantly declined after dredging during the course of experiment (two-way RM-ANOVA, treatment effect, $p = 0.000$), approximately by 222%. While $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ concentrations in post-dredged pore water were slightly greater than those of control, notably by 115 and 104% (Fig. 3, $p = 0.030$; $p = 0.612$). With regard to the vertical distributions of inorganic N, there were significant differences among depths for $\text{NH}_4^+\text{-N}$ (two-way RM-ANOVA, depth effect, $p = 0.029$), but the differences of $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ were not significant (Tukey's HSD test, $p > 0.05$). Moreover, pore water $\text{NH}_4^+\text{-N}$ concentrations showed a downward increasing trend from the SWI to

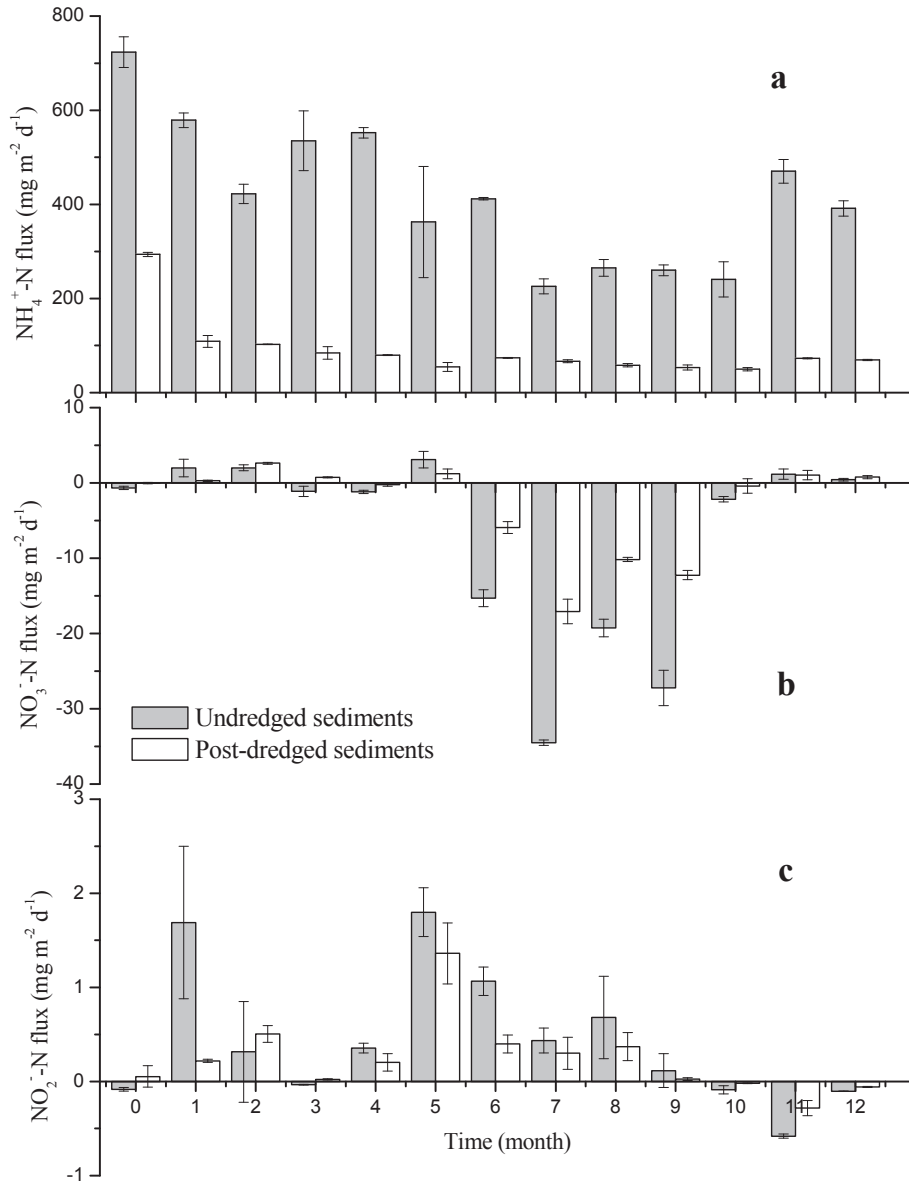


Fig. 4. Diffusive fluxes of inorganic nitrogen across the sediment-water interface according to Fick's first law. Positive fluxes are out of sediment, and values are expressed as the mean \pm SD of three replicates.

the 9 cm depth in both control and post-dredged sediments, and the NO₃⁻-N and NO₂⁻-N concentrations varied irregularly, with the greater values in upper post-dredged sediments and in the bottom of control sediments.

3.3. Effects of dredging on fluxes of inorganic nitrogen

During the course of the experiment, sediment was a major source of NH₄⁺-N for overlying water, whereas it also served as both the pool and source of NO₃⁻-N and NO₂⁻-N (Fig. 4). The diffusion fluxes of NH₄⁺-N, NO₃⁻-N and NO₂⁻-N in post-dredged sediments were significantly lower than those of control sediment (Tukey's HSD test, $p < 0.05$), notably by 458, 235 and 179% respectively, and dredging partly weakened NO₃⁻-N and NO₂⁻-N pools of sediments for overlying water. Fluxes of various inorganic nitrogen varied differently after dredging. The fluxes of NH₄⁺-N in post-dredged sediments gradually decreased, but in control sediment, the fluxes were changing in an irregular pattern. The patterns of NO₃⁻-N

and NO₂⁻-N fluxes also changed irregularly, and sediments was a sink for NO₃⁻-N, but not for the latter.

3.4. Effects of dredging on labile N of sediments

The content and depth distributions showed that NH₄⁺-N and NO₃⁻-N were the predominant forms of inorganic N in both control and post-dredged sediments (Fig. 5). Dredging significantly decreased the concentrations of NH₄⁺-N and NO₃⁻-N over the whole depths of sediments ($p < 0.01$) (with the decreases of 14.64 and 10.15 mg kg⁻¹, respectively). Conversely, dredging remarkably increased the contents of NO₂⁻-N in sediments ($p < 0.05$). The profiles of NH₄⁺-N changed inversely between control and post-dredged sediments, with downward decreasing trends in the control sediments and gradually increasing trends with depths in post-dredged sediments. In addition, the depth changing patterns of NO₂⁻-N and NH₄⁺-N in post-dredged sediments were opposite. However, both profiles of NH₄⁺-N and NO₃⁻-N in control sediments

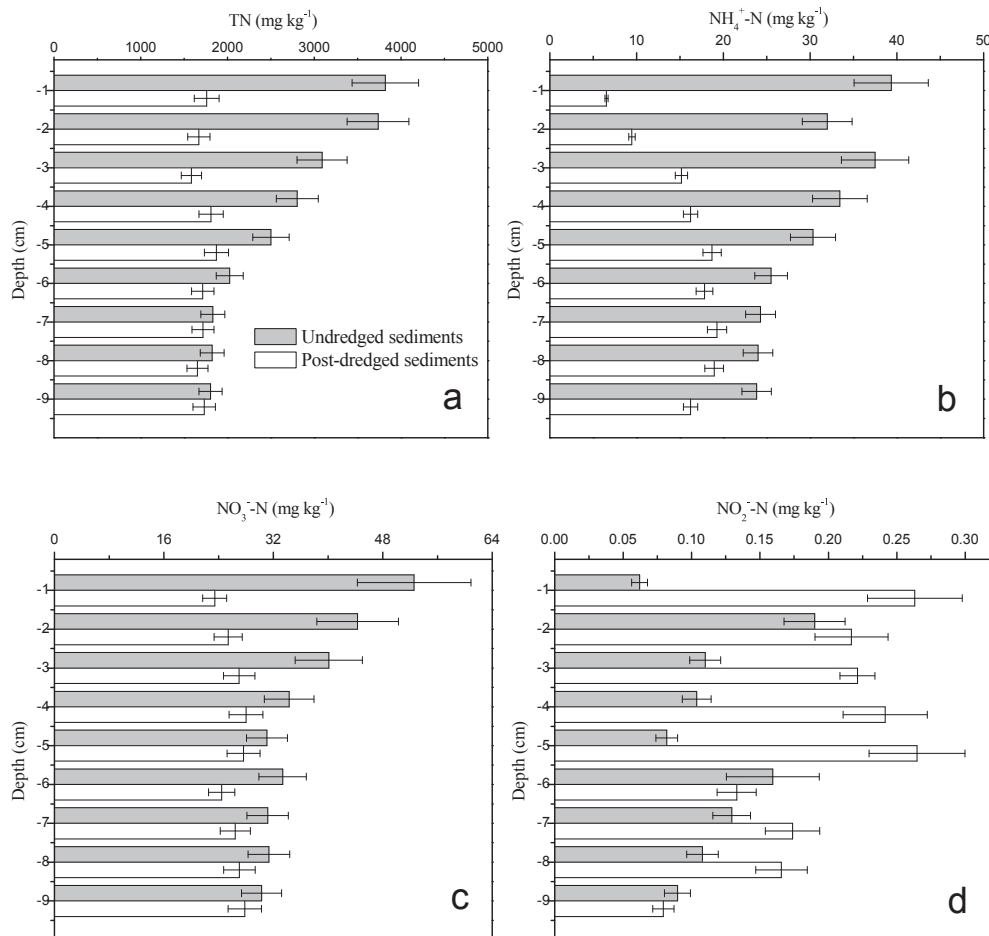


Fig. 5. Vertical profiles of TN (a), $\text{NH}_4^+\text{-N}$ (b), $\text{NO}_3^-\text{-N}$ (c) and $\text{NO}_2^-\text{-N}$ (d) in sediments of undredged and post-dredged after dredging one year, and values are expressed as means \pm SD of three replicates.

were decreasing downward, while $\text{NO}_2^-\text{-N}$ varied irregularly with depths.

3.5. Effects of dredging on N fractions of sediments

IMOF-N and OSF-N were the predominant forms in sediments (Figs. 5 and 6). The contents of TN, IEF-N, CF-N, IMOF-N and OSF-N were generally decreased after dredging, notably by 66, 74 and 50% for TN, IEF-N and OSF-N, respectively ($p < 0.01$), but not for CF-N and IMOF-N ($p > 0.05$). As a whole, the concentrations of N fractions in both sediments were in the order of IMOF-N > OSF-N > IEF-N > CF-N.

3.6. Effects of dredging on sediment bacteria community structure

The estimated abundances of bacterial phyla communities in sediments were presented in Fig. 7. *Proteobacteria* was the predominant phylum in both undredged and post-dredged surface sediments, approximately accounting for 56 and 37% respectively. After dredging, the *Nitrospira* phyla were obviously greater than that of those in the control treatment (around 244% compared with undredged sediments), and the phyla of *Chloroflexi*, *Acidobacteria* and *Firmicutes* also generally increased compared with non-dredged sediments. On the other hand, dredging induced decreases in bacteria abundances, but increased the biodiversity (Table S1). Dredging was beneficial to the enrichment of *Nitrospira* abundance, approximately 184% compared with control. However,

the abundances of *Nitrosomonas*, *Nitrospira* and *Nitrosococcus* generally decreased after dredging. Similarly, the abundance of *Thiobacillus*, *Sterolibacterium*, *Denitratisoma* and *Hyphomicrobium* generally declined after dredging, and the abundance of *Anaeromyxobacter* and *Caldithrix* also decreased. In the unclassified genus, the abundance of the *Nitrospiraceae* family also increased in post-dredged sediments (Fig. 8).

4. Discussion

The cycling of N in freshwater sediments is regulated by the rate and lability of organic matter deposition, sediment accumulation rates and bioturbation/bioadvection, and rates of microbially mediated N transformation (Cornwell and Owens, 2011). Regeneration and transfer of N from sediments to overlying water are affected by the chemical and biological stability of sediments (Wu et al., 2001). Dredging is a large-scale, anthropogenic disturbance associated with high costs, and it often destroys the balance of the SWI, 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; Cabrita, 2014). Specifically, dredging could notably increase the turbidity and suspended particulate matter (SPM) concentration in the water column (Cabrita, 2014). Moreover, dredging could result in lower sediment oxygen demand, aeration state (Eh) and pH values in dredging regions via removal of the upper sediment rich

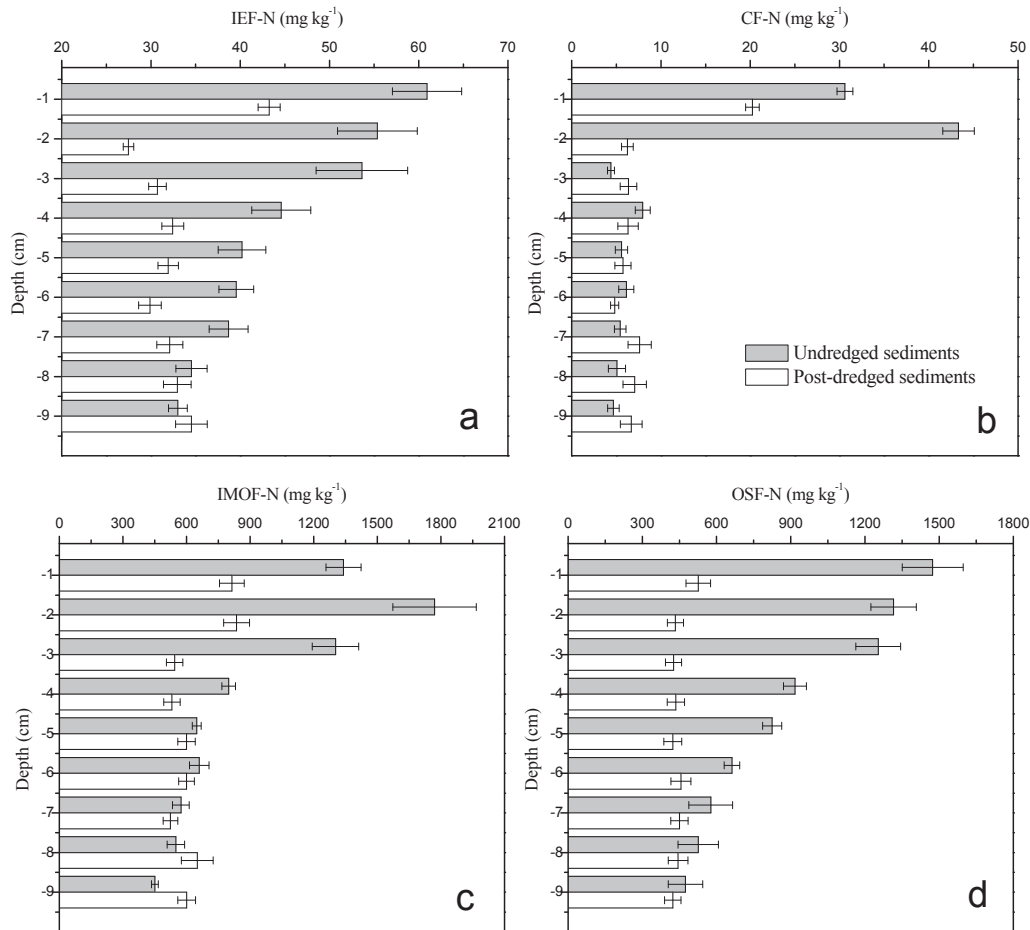


Fig. 6. Vertical profiles of different N fractions in undredged and post-dredged sediments after dredging one year, and values are expressed as means \pm SD of three replicates.

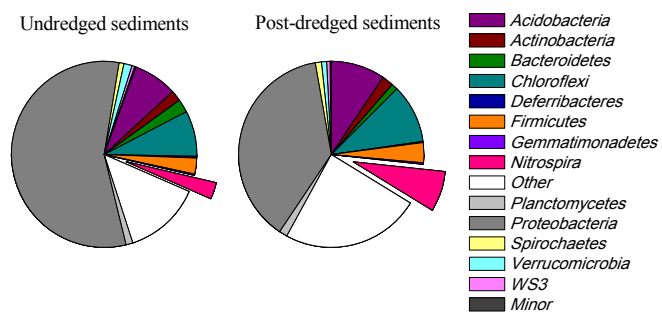


Fig. 7. Average abundances of different bacterial phyla in surface sediments (0–1 cm). The abundance is presented in terms of percentage in total effective bacterial sequences in a sample.

in TOC and/or TN (Table 1; Fig. 2). Thus, it is inevitably changed the inherent cycling and exchange of N across the SWI in aquatic ecosystem.

4.1. Effect of dredging on N release across the sediments-water interface

During the course of the experiment, dredging could significantly decrease the diffusion fluxes of NH_4^+ -N from sediments to overlying water ($p < 0.001$), but slight effect on NO_3^- -N and NO_2^- -N (Figs. 3 and 4). The mechanisms of dredging for these phenomena

were due to the following reasons: (1) according to the vertical profiles of pore water N, the diffusive layer of the NH_4^+ -N, NO_3^- -N and NO_2^- -N in control sediments were obviously thicker than those in dredged treated sediments (Fig. 3); Moreover, the ionic diffusion potential of NH_4^+ -N and NO_2^- -N in control sediments were also greater than those in dredged sediment, which were in agreement with previous studies (Yu et al., 2012; Ding et al., 2015). (2) Fick's first law indicates that low sediment porosity can inhibit the N exchange across the SWI (Jing et al., 2013; Zhong et al., 2009). (3) ammonium migration blocked by the presence of major factors in sediment was adsorbed by soil colloids such as Fe/Mn oxides and organic matter and precipitated with fine grained sediment particles (Wang et al., 2008; Gardner et al., 1991). In this study, the contents of Fe and Fe (II) in dredged sediments were lower than those in the control sediment (Tables 1 and 2), which implied that the effectiveness of dredging in reducing NH_4^+ -N release were underestimated because NH_4^+ -N in control sediments were more inclined to adsorb by Fe hydroxides under anoxic condition (Fig. 2).

4.2. Mechanisms of dredging on N regeneration ability in sediments

With regard to N mobilization from sediments, NH_4^+ -N originated from the decomposition of organic nitrogen compounds could accumulate in the interstitial water and partly diffuse out of the sediments into the overlying water or become adsorbed into the sediment (Medina et al., 2003; Yang et al., 2010). In this present study, OM was the major reason for the results why the fluxes of NH_4^+ -N and pore water NH_4^+ -N concentrations induced by dredging

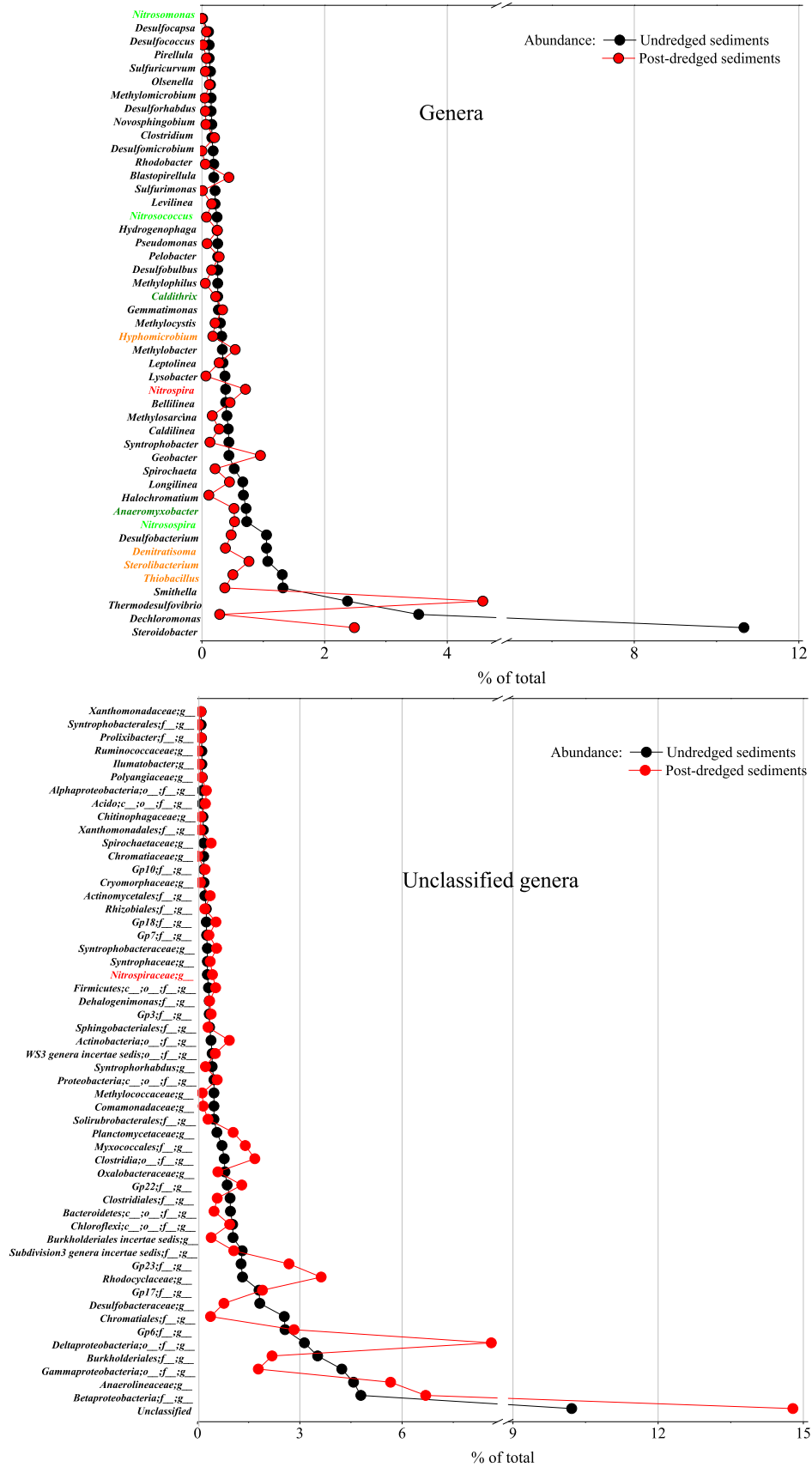


Fig. 8. Relative abundances of the top 47 classified bacterial genera and of the top 53 unclassified bacteria genera in surface sediments (0–1 cm). The blue represents ammonia-oxidizing bacterium; red represents nitrite-oxidizing bacterium; yellow represents denitrifiers and green represents nitrate-reducing bacterium. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2

Pearson correlations between nitrogen forms in sediments of undredged and post-dredged and sediment characteristics.

Fractions	pH		LOI		Fe		AVS	
	Undredged	Post-dredged	Undredged	Post-dredged	Undredged	Post-dredged	Undredged	Post-dredged
TN	−0.971**	−0.074	0.977**	−0.046	0.975**	0.223	0.840**	0.065
IEF-N	−0.974**	0.779*	0.974**	0.494	0.985**	0.124	0.829**	−0.288
CF-N	−0.804**	0.836**	0.728*	0.796*	0.704*	0.332	0.953**	−0.563
IMOF-N	−0.911**	0.512	0.951**	0.733*	0.935**	0.467	0.865**	−0.559
OSF-N	−0.970**	0.902**	0.989**	0.789*	0.995**	0.502	0.801**	−0.765*
NH ₄ ⁺ -N	−0.874**	−0.520	0.911**	−0.908*	0.929**	−0.581	0.574	0.464
NO ₃ ⁻ -N	−0.975**	−0.721	0.896**	−0.800**	0.905**	−0.738*	0.917**	0.784*
NO ₂ ⁻ -N	0.033	−0.097	0.066	0.555	0.042	0.104	0.189	−0.300

Note: The one and two superscript asterisks show the significant level at $p < 0.05$ and 0.01 , respectively.

were effectively reduced (Table 2, Figs. 3 and 4). Owing to the organic early diagenesis process, OM in dredged sediments would be resistant to decomposition by mineralization, which is partly imputed to a lower abundance of microbes (Zhong et al., 2009; Table S1). The results was in agreement with previous studies showed that heavily polluted sediment has a higher mineralization rate and net mineral-N and mineral-P than slightly polluted sediment. Thus, these results further confirmed this early diagenesis (Yang et al., 2010; Zhong et al., 2009). In addition, numerous studies showed a noticeable decrease in abundances of benthic animals and microbes in the dredged areas, which inevitably hindered the organic N decomposition processes and then reduced N regeneration (Zhong et al., 2009, Table 1; Table S1; Fig. S1).

Generally, it is well known that N regeneration in sediments were significantly related with N stability in sediments (Wang et al., 2008). In terms of mobility of diverse N fractions, IEF-N that was the weakly sorbed N was significantly correlated with factors that affected N sorption process (Lv et al., 2005). The regeneration of CF-N in sediment was based on the contents of carbonate and organic matter with the concentrations of CF-N increasing with the content of carbonate and organic matter (Wang et al., 2008). IMOF-N represented a redox-sensitive N fraction, mainly bound to Fe and Mn oxides (Kozerski and Kleeberg, 1998). OSF-N represents the organic matter and sulfide form, which is considered as a relatively stable fraction that mainly consists of organic N (Wang et al., 2008; Ma et al., 2003). In this study, the stability of sediment N generally declined by dredging (Figs 3, 5 and 6). The likely reason was primarily attributed to removal of the upper sediment enriched with N loading. On the other hand, it could be argued that the stability and concentrations of sediment N forms significantly correlated with the differences in particle grain size composition. Numerous previous studies observed that dredging was able to enlarge the proportion of coarse grain size particle in sediment (Zhong et al., 2010; Yu et al., 2012). The contents of N forms in post-dredged sediments were generally refractory fractions and thereby resistant to decomposition by mineralization during early diagenesis process (Yang et al., 2010). The effects of sediment grain size on N release caused by dredging was not only differences in contents of N in sediments, but also influences of N release processes. Therefore, low flux of N forms in post-dredged sediments with low contents and lability of sediment N forms after dredging.

Above all, basic microbial processes to which nitrogen was subjected in the sediment included denitrification, nitrification, ammonification, anaerobic ammonium oxidation (anammox) and dissimilatory NO₃⁻-N reduction to ammonium (DNRA) processes, which may result in N transformation and N removal from aquatic ecosystem (Xia et al., 2009; Hanaki et al., 1990; Trimmer et al., 2003). The fluxes magnitude of NO₃⁻-N and NO₂⁻-N induced by dredging varied slightly compared with the NH₄⁺-N fluxes range, which presumably due to nitrification undergoing mineralization, anammox, denitrification or DNRA processes in sediments (Fig. 8).

The higher NH₄⁺-N concentrations in control sediments seemed more likely due to much higher ammonium release through ammonification which can proceed aerobically and anaerobically over a wide pH range. Furthermore, ammonium produced by decomposition of organic matter was mainly attributed to larger contents of bacterial degradable organic matter in control sediments (Kemp and Mudrochova, 1972; Zhong et al., 2009). On the second place, higher DNRA rates were observed with higher sulfide and NO₃⁻-N concentrations (An and Gardner, 2002). The higher abundances of nitrate-reducing bacterium provided the valid evidence needed for this presentation (Sanford et al., 2002, Fig. 8). However, with higher oxygen depletion, more negative values of redox potential, greater contents of organic matter and greater nitrate concentrations, coupled denitrification with anammox processes in control sediments would enhance and then converted NO₃⁻-N, NO₂⁻-N and NH₄⁺-N into dinitrogen (N₂) or nitrous oxide (N₂O) (Pinay et al., 2003; Bastviken et al., 2005; Venterrink et al., 2003; Trimmer et al., 2003), while nitrification would be inhibited under anoxic condition (Xia et al., 2009). This hypothesis can be further verified in this study showed that abundance of denitrificans (such as *Thiobacillus*, *Sterolibacterium*, *Denitratisoma* and *Hyphomicrobium*) generally decreased, while the nitrite-oxidizing bacterium abundance (*Nitrospira*) effectively increased by dredging (Kelly and Wood, 2000; Tarlera and Denner, 2003; Fahrbach et al., 2006; Ehrich et al., 1995; Jones and Morita, 1983, Figs. 7 and 8). In brief, the reason for this phenomenon could be attributed to weaker denitrification and DNRA rates, whereas enhanced nitrification after dredging (Jing et al., 2013; Sanford et al., 2002; Zhong et al., 2009, Table 1). Taken together, the study showed that dredging would slow down the N cycling and further weaken NO₃⁻-N and NO₂⁻-N removal from aquatic ecosystems, which should be paid much more attention when dredging is applied to lake restoration in the future (Graca et al., 2004; Zhong et al., 2010; Jing et al., 2013).

It was unexpect that many previous studies have showed that temporary NH₄⁺-N releases to overlying water would be augmented by dredging both in-field investigations and simulation studies, which were inconsistent with the results obtained in this study (Zhong et al., 2009; Jing et al., 2013; Wang et al., 2008; Fan et al., 2004). In this regard, as for field dredging practices, the reason for this phenomenon can be driven by NH₄⁺-N concentrations in pre-existing pore water and adsorbed NH₄⁺-N concentrations, plus any ongoing N regeneration and dissimilatory microbial processing of N (Morin and Morse, 1999; Reddy et al., 2007). Furthermore, the fresh deposited sediments onto the new surface in field dredging projects and suspended fine grain particles retained in water columns inevitably altered the properties of SWI and then caused the field instantaneous measurements to significantly deviate from their true values (Fan et al., 2004; Yu et al., 2012). In the second place, according to the Chinese environmental dredging common standard, when TN concentration in sediments is larger than

1000 mg kg⁻¹, the sediments are regarded as heavily polluted and should be dredged (Wang et al., 2008). In this study, TN concentrations in post-dredged sediments were still higher than the threshold value, approximately 50% to TN in undredged sediments, which further showed that dredging implemented in highly polluted sediments can significantly decrease the NH₄⁺-N release from sediments by permanently removing the surface sediments rich in TN and/or OM.

5. Conclusion

The results showed that dredging was able to reduce the NH₄⁺-N flux from sediments over the whole experiment, while had slight effects on NO₃⁻-N and NO₂⁻-N fluxes. Moreover, the diffusion layers of NH₄⁺-N were shallower and ionic diffusion potentials were also lower after dredging, but not for NO₃⁻-N and NO₂⁻-N. The N fractionation showed the contents of different N fractions in post-dredged sediments were significantly lower than those of control. Further analysis indicated that after dredging, the nitrite-oxidizing bacterium abundance enhanced, whereas the relative abundances of denitrifiers and nitrate-reducing bacterium decreased. Taken together, this study demonstrated that dredging applied to highly polluted sediments can effectively reduce N release, especially for NH₄⁺-N release, resulting from removal of the upper contaminated sediment enriched with N and/or OM. Additionally, dredging seems to weaken NO₃⁻-N and NO₂⁻-N removal from freshwater ecosystems.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2016.03.062>.

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