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Significance of dredging on sediment denitrification in Meiliang Bay, China: A year long simulation study

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Abstract

An experiment for studying the effects of sediment dredging on denitrification in sediments was carried out through a one-year incubation of undredged (control) and dredged cores in laboratory. Dredging the upper 30 cm of sediment can significantly affect physico-chemical characteristics of sediments. Less degradation of organic matter in the dredged sediments was found during the experiment. Denitrification rates in the sediments were estimated by the acetylene blockage technique, and ranged from 21.6 to 102.7 nmol N_2/(g dry weight (dw)·hr) for the undredged sediment and from 6.9 to 26.9 nmol N_2/(g dw·hr) for dredged sediments. The denitrification rates in the undredged sediments were markedly higher ($p < 0.05$) than those in the dredged sediments throughout the incubation, with the exception of February 2006. The importance of various environmental factors on denitrification was assessed, which indicated that denitrification was regulated by temperature. Nitrate was probably the key factor limiting denitrification in both undredged and dredged sediments. Organic carbon played some role in determining the denitrification rates in the dredged sediments, but not in the undredged sediments. Sediment dredging influenced the mineralization of organic matter and denitrification in the sediment; and therefore changed the pattern of inherent cycling of nitrogen.

Key words: sediment dredging; denitrification; Taihu Lake

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Introduction

Sediment pollution is a major problem in aquatic ecosystem management and restoration. Nutrients, toxic chemicals and toxin-forming microbes are found in much higher contents in sediments than in the overlying water column. Improvements in the quality of the overlying water and associated components of the aquatic ecosystem often cannot be achieved without some form of sediment remediation (Murphy et al., 1999).

Sediment dredging is a lake restoration technique that removes the surface sediment layer rich in pollutants thus decreases their release from the sediments to the water column. Dredging is currently the most commonly selected option for remedying contaminated sediments (Gustavson et al., 2008). However, there is an ongoing debate about the negative effects (e.g., resuspension and transport of contaminants) and positive benefits (e.g., permanently removing contaminants from an aquatic system) of dredging contaminated sediments (Je et al., 2007; Mackie et al., 2007). Sediment dredging can greatly modify the physical, chemical and biological conditions of the surface sediments (Robinson et al., 2005), including the inherent cycling of the biologically important elements such as nitrogen and phosphorus (Graca et al., 2004).

Denitrification is a microbially mediated process where NO_3^- is reduced to N_2O or N_2, and heterotrophic bacteria, i.e., denitrifiers, are the dominant organisms responsible for this process (Payne, 1973). Denitrification can play an important role in the nitrogen budgets of lakes (Cavari and Phelips, 1977). Sediment denitrification is particularly important in the context of N export because it represents a permanent removal of N from the aquatic ecosystem (Seitzinger, 1988; Sjodin et al., 1997). Denitrification within sediments can be limited by concentrations of NO_3^- and organic carbon, temperature, and redox conditions (García-Ruiz et al., 1998a; Pinay et al., 2003; Baeseman et al., 2006).

Taihu Lake is the third largest freshwater lake in China with a surface area of 2338 km^2 and an average depth of about 2 m (Chen et al., 2003). Meiliang Bay is one of the most eutrophied bays in the northern part of Taihu Lake. Blooms of blue-green algae have been frequently occurring in warmer seasons (Pu et al., 1998). In recent years, the external nutrient loading has been reduced

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considerably, but the trophic state of the lake has not been improved. Unfortunately, the net internal nutrient loading in eutrophic Taihu Lake is delaying the effect of reduction of the external nutrient loading (Qin et al., 2004; Fan et al., 2006). Sediment dredging has been carried out in Wuli Lake, a sublake of Lake Taihu (Cao et al., 2007). Dredging in Meiliang Bay will be carried out in the coming years. It is thus necessary to assess the environmental effects of sediment dredging on Meiliang Bay beforehand.

In this study, we used sediment-water microcosms to assess the effect of dredging on sediment denitrificaiton, and the effects of environmental factors (i.e., temperature, organic carbon and nitrate) on sediment denitrification rates.

1 Materials and methods

1.1 Site description

The sampling site (31°31′33.9″N, 120°12′35.2″E) is located in a key state tourism area in the Meiliang Bay (Fig. 1). There is a water intake and purification plant for the city of Wuxi (about 2 km near the sampling site). The sampling site is a proposed dredging region in the associated remedying plan for Taihu Lake.

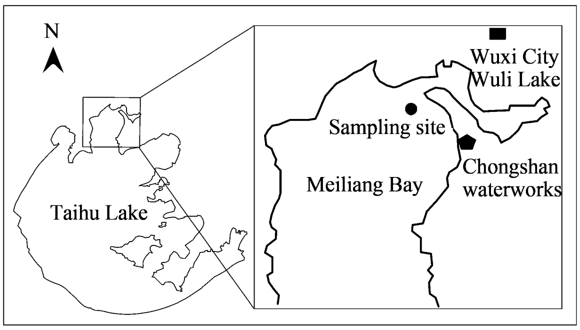

Fig. 1 Location of the sampling site in Meiliang Bay, Taihu Lake.

1.2 Sampling and microcosm experiment

Seventy-eight intact sediment cores (11 cm in diameter and 60–70 cm long) were collected with a core sampler at the sampling site on December 19, 2005, along with water from the overlying water column. The bottom of the cores was sealed with an additional rubber stopper. Care was taken to preserve sediment structure during sampling and transport.

In the laboratory, the overlying water was siphoned from each core. To simulate dredging, the uppermost 30 cm sediment layer of 39 undisturbed cores was skimmed by a suction pump. The water drained off was immediately replaced by filtered site water. The other 39 undisturbed cores were not treated and maintained as control cores. Undisturbed cores (control) and dredged cores (20 cm-long sediment and 20 cm-thick overlying water) were incubated in a water bath maintained with *in-situ* water temperature ± 2°C, and left open to the atmosphere. The water bath was placed in the incubation room, and all cores were incubated under either light or dark (wrapped in double aluminium foil) conditions according to *in-situ* photoperiod and light intensity. Metal halide lamps (400 W) were used as the light source.

During the incubation, the water overlying the undisturbed and dredged cores was replaced by site water biweekly. In order to simulate *in-situ* environmental conditions in the laboratory experiments, the suspending material captured by the traps (ten glass jars, 5 cm in diameter and 18 cm in length) at the sampling site was added biweekly to the incubated cores according to the *in-situ* sedimentary rate (0.25 g/(cm^2·yr), Qin et al., 2004).

Undisturbed (control) and dredged cores (in triplicate) were sacrificed for analysis monthly during the incubation period. The overlying water was siphoned from each core. The sediments were extruded and the upper 2 cm layer sediments were collected from the cores, homogenized and stored in a sterilized plastic bag. The subsamples were centrifuged at 5000 r/min and 4°C for 20 min to acquire pore water. The pore water was filtered through 0.45 μm membrane filters and stored at 4°C until analysis. Sediment subsamples were freeze-dried and ground, then sieved with a 100-mesh sieve. Wet and dry sediment samples were collected for analysis. This simulated dredging experiment was carried out according to the above protocol for one year.

1.3 Analytical methods

Concentrations of ammonium and nitrate plus nitrite in the pore water were measured with a flow-injection autoanalyzer (Skalar Sanplus, the Netherlands).

Sediment water content (wet weight (wt.)%) was determined by drying sediment samples at 105°C until reaching a constant mass. Porosity and bulk density were measured using a cutting ring. Particle size of sediments was determined using a Mastersizer 2000 Laser Size Analyzer (Malvern Co., UK). Loss on ignition (LOI%, *m/m*) was determined by calculating the weight loss after heating dry sediment samples to 550°C for 6 hr. Dried aliquots of sediment were homogenized and analysed for content of organic carbon (OC) and total nitrogen (TN) with a CHN elemental analyzer (CE-440, EAI, USA). For total phosphorus (TP) determination, 0.2 g finely ground dry sediments were combusted at 450°C in a muffle furnace for 3 hr, followed by 3.5 mol/L HCl extraction (Ruban et al., 1999). Soluble reactive phosphorus (SRP) concentrations in the extract were analyzed using the molybdenum blue method (Murphy and Riley, 1962).

1.4 Measurements of potential denitrification

Potential denitrification was determined using the acetylene inhibition technique for slurries according to Sørensen (1978) and Magalhães et al. (2005). Briefly, the slurries were prepared by adding 10 mL of incubation water to 50 mL serum bottles containing homogenized and weighed sediments (3 g). Serum bottles were hermetically sealed with butyl stoppers and aluminum crimp seals. Each serum bottle, with the sample and incubation water, was purged

Table 1 Physico-chemical characteristics of surface layer (0–2 cm) of control and dredged sediments

Treatment	Water content (%, *m/m*)	Porosity (%, *V/V*)	Bulk density (g/cm^3)	LOI (%, *m/m*)	OC (%, *m/m*)	TN (%, *m/m*)	TP (mg/kg)	Clay and silt (%, *V/V*)	Fine sand (%, *V/V*)	Coarse sand (%, *V/V*)
Control	63	93	1.46	4.50	1.50	0.35	912	70.9	27.8	1.3
Dredged	50	76	1.54	4.01	1.27	0.41	646	64.6	25.8	9.7

The original cores from December, 2005 were used for simulated dredging experiment. Data are mean of three replicates.
LOI: loss on ignition, OC: organic carbon, TN: total nitrogen, TP: total phosphorus.

with N_2 to remove O_2; triplicate samples with and without acetylene (20%, *V/V*) were treated and a separate set of time zero samples was sacrificed immediately after acetylene addition. All samples were incubated in the dark for 4 hr at 20°C with stirring (70 r/min). At time zero and 4 hr, 18 mL of gas were collected (after headspace equilibration via vigorous shaking) from each serum bottle and stored in an evacuated serum vial (18 mL) for analysis of N_2O. The gas samples were collected from each serum bottle by simultaneously adding 18 mL of a 3 mol/L NaCl solution to compensate the gas space (Joye et al., 1996). N_2 produced via denitrification was calculated as the difference between the N_2O produced with and without acetylene. N_2O was quantified using a gas chromatograph (GC-14B, Shimadzu, Japan) equipped with an electron-capture detector.

To compare the potential denitrification of the undredged and dredged sediments, the same artificial water was used for denitrication rate measurements during the experiment. The artificial water was enriched with 15 mg/L NO_3^--N, 5 mg/L NH_4^+-N, 0.7 mg/L PO_4^{3-}-P, 2.5 mg/L acetate-C, 2.5 mg/L glucose-C. Essential nutrients were added in excess to the incubations to assure that potential denitrification was not limited by the amount of nutrients available. Incubation temperature was maintained at 20°C. Thus, any possible temperature effect on denitrification was eliminated.

1.5 Influence of environmental factors on denitrification

1.5.1 Temperature

The effect of temperature on sediment denitrification was investigated on both undredged and dredged cores incubated for nine months in the laboratory microcosm. Sediment samples were collected and slurries made as above. The samples were incubated under three different temperatures (10, 20, 30°C) with the same artificial water as described in Section 1.4.

1.5.2 Organic carbon (glucose) and nitrate

The effects of glucose, nitrate and glucose plus nitrate on sediment denitrification were investigated for both undredged and dredged cores incubated for nine months in the laboratory microcosm. Sediment samples were collected and slurries made as above. The incubation water was the following solutions: (1) deionized water only (DW); (2) deionized water enriched with 30 mg/L glucose-C (DW + C); (3) deionized water enriched with 5 mg/L NO_3^--N (DW + N); (4) deionized water enriched with 30 mg/L glucose-C and 5 mg/L NO_3^--N (DW + C + N). Incubation temperature was maintained at 20°C.

1.6 Statistical analysis

Statistical analysis was performed using SPSS 12.0. One-way ANOVA was performed to determine significant differences in denitrification among different months and between different environmental factor treatments. Linear regression analysis was further used to determine the relationship between denitrification rates and temperature. Additionally, a *t*-test was performed to determine significant difference between the denitrification rates of the undredged and dredged sediments. For all statistical methods used in this study, results were considered significant if probabilities were less than 0.05.

2 Results

2.1 Physico-chemical characteristics of sediments

Physico-chemical characteristics of the original control and dredged sediment (0–2 cm layer), are shown in Table 1. As compared with control sediment, water content and porosity of dredged sediments decreased markedly, and bulk density increased accordingly; organic matter content (as LOI% dw) and OC content decreased, but content of TN had minor increase; The contents of TP in dredged sediments decreased markedly; particle size distribution was also different in control and dredged sediments, and the amount of the clay, silt and find sand fractions decreased in the dredged sediments, and that of the coarse sand fraction increased.

2.2 Inorganic nitrogen in pore water

Inorganic nitrogen concentrations varied during the experiment in pore water of the undredged sediments (Fig. 2), with clear seasonal trend. Ammonium concentration was high in warmer months and low in other months. Nitrate plus nitrite concentration had a different trend compared to ammonium, with high concentration in colder months and low concentration in warmer months. Inorganic nitrogen had no clear seasonal trend in pore water of the dredged sediments (Fig. 2). Ammonium concentration was high at the beginning of the experiment and decreased with time thereafter. Nitrate plus nitrite concentration was low and increased with time, and decreased in warmer months and increased again in colder months during the experiment.

2.3 Denitrification rates

Denitrification rates varied during the experiment in the undredged sediments (Fig. 3), with the highest values in August and September (ANOVA, $p < 0.05$) and much lower in December 2005, January and February 2006 (ANOVA, $p < 0.05$). The rates were positively correlated

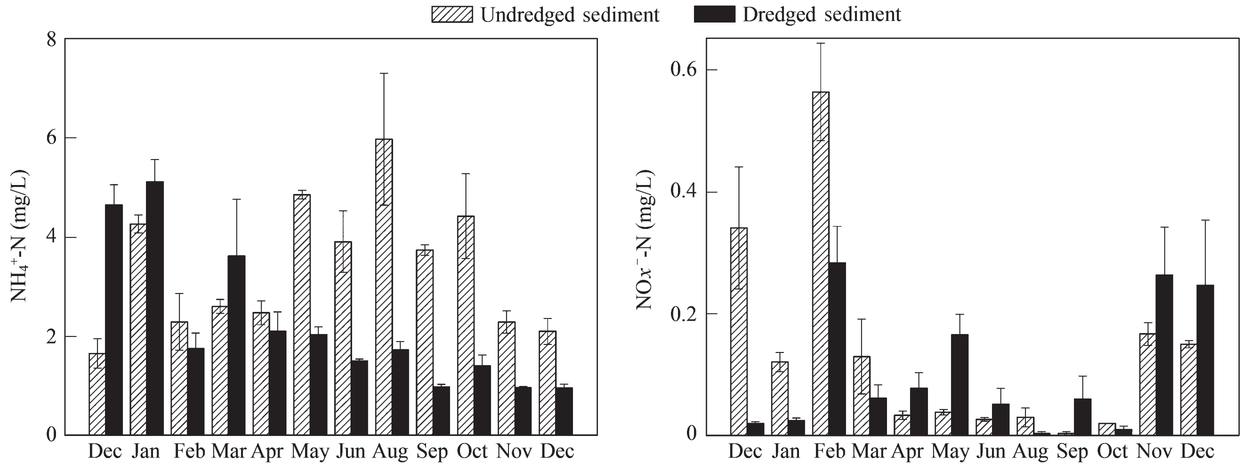

Fig. 2 Seasonal variations in the concentration of inorganic nitrogen in the pore water (0–2 cm levels) of the undredged and dredged cores collected monthly from the macrocosm experiment. Error bars represent standard error of the mean of three replicates, with no data in July 2006.

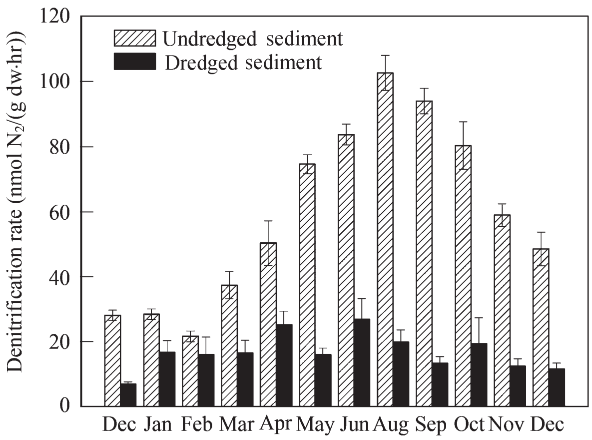

Fig. 3 Rates of potential denitrification in the sediments of the undredged and dredged cores collected monthly from the macrocosm experiment. Eerror bars represent standard error of the mean of three replicates, with no data in July 2006.

to the monthly mean temperature (r = 0.98, p < 0.001, n = 12; Fig. 4a) and showed clear seasonal periodicity. Denitrification rates in the dredged sediments showed a weak increase with temperature, but the values do not fluctuate markedly during the experiment. Also, they did not show significant correlation with the monthly mean temperature (Fig. 4b). Denitrification rates of the undredged sediments were generally greater than those of the dredged sediments. Differences in denitrification rates between the undredged and dredged sediments were evaluated by a t-test, and were statistically significant (p < 0.05) during the experiment with an exception in February, 2006.

2.4 Influence of environmental factors on denitrification

2.4.1 Temperature

The denitrificaiton rates in both undredged and dredged sediments increased within temperature range 10–30°C (Fig. 5). The increase for the undredged sediments was markedly higher than that for the dredged sediments.

2.4.2 Carbon and nitrate

The denitrification rates of the undredged and dredged sediments were low when the sediments incubated with deionized water (Fig. 6). No effect of glucose on the denitrification rates was observed for both undredged and dredged sediments in the absence of nitrate, but single nitrate addition significantly promoted the denitrification rates in both undredged and dredged sediments (p < 0.05). Denitrification rate in the undredged sediment incubated in

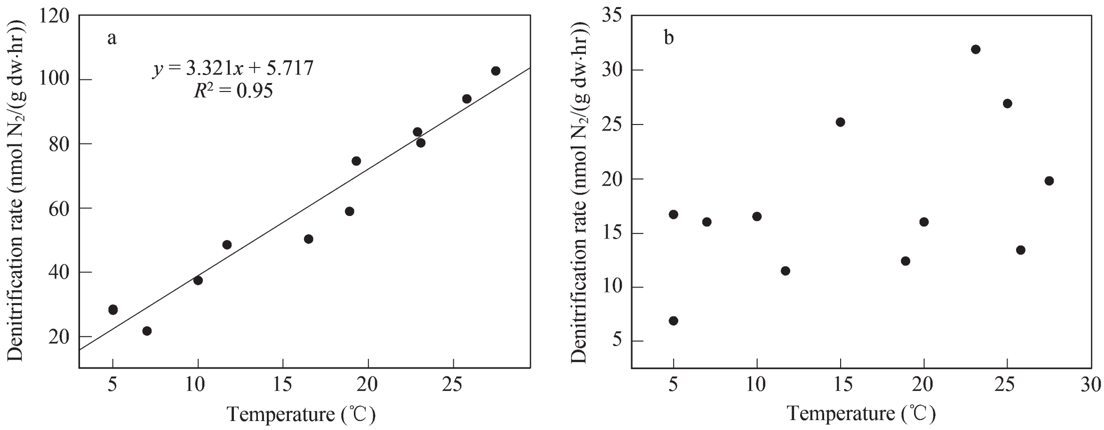

Fig. 4 Relationships established for monthly surveys between denitrification rates and monthly mean temperature for the undredged (a) and dredged (b) sediments.

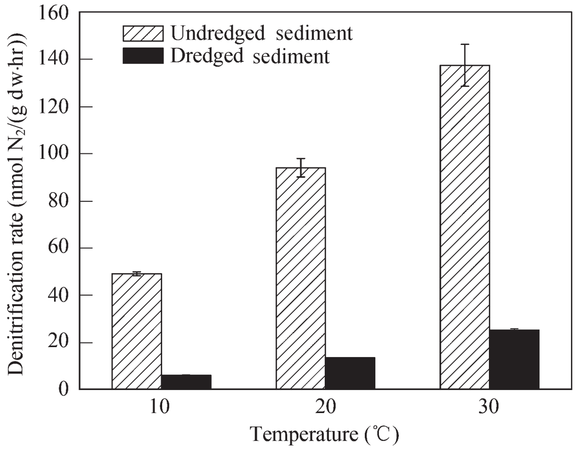

Fig. 5 Effect of temperature on denitrification rates in the undredged and dredged sediments collected from the microcosm experiment in September 2006. Error bars represent standard error of the mean of three replicates.

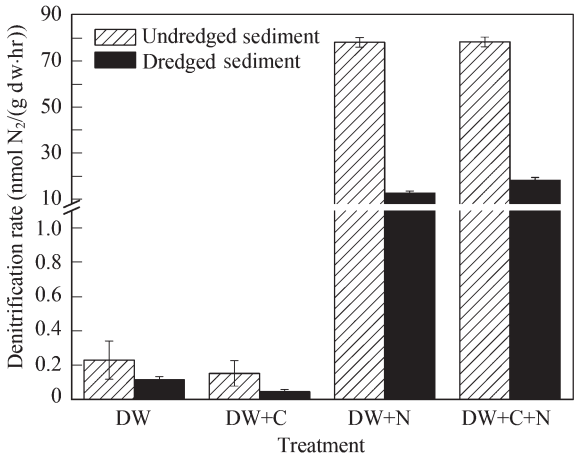

Fig. 6 Effects of glucose and nitrate on denitrification rates in the undredged and dredged sediments collected from the microcosm experiment in September 2006. Error bars represent standard error of the mean of three replicates. DW: deionized water only; DW+C: deionized water with 30 mg/L glucose-C; DW+N: deionized water with 5 mg/L NO_3^--N; DW+C+N: deionized water with 30 mg/L glucose-C and 5 mg/L NO_3^--N.

the presence of nitrate plus glucose was not significantly different ($p > 0.05$) compared to that with single nitrate treatment, but significantly higher ($p < 0.05$) in the dredged sediments (Fig. 6).

3 Discussion

3.1 Seasonal variation of inorganic nitrogen in pore water

In this study, dredging the upper 30 cm of sediment can markedly affect physico-chemical characteristics of sediments (Table 1), and the changes of the sediment characteristics would influence nitrogen transformation in the dredged sediments.

Concentration of inorganic nitrogen in pore water of the undredged and dredged sediments had different seasonal trends (Fig. 2). Ammonium released from sediments is resulted from decomposition of organic matter. Ammonium concentration in pore water of undredged sediments increased in the warmer months, due to intensified degradation of organic matter. High concentrations of ammonium were recorded in dredged sediments at the beginning of experiment because a considerable upward flux of ammonium was occurred at the sediment-water interface, which was also shown by the increase in ammonium concentrations with sediment depth as described by others (Berner, 1974; Christensen et al., 1988). The decrease in ammonium concentration in dredged sediment with time may be due to the less degradation of organic matter during the experiment.

The difference in degradation of organic matter in the undredged and dredged sediments may be attributed to the different characteristics of sediments. Due to diagenesis, the amount of organic matter in the dredged sediments decreased (Table 1). Furthermore, there was more highly degraded refractory organic matter in dredged sediments (buried sediments), which is less susceptible to microbial degradation (Kleeberg and Kohl, 1999). In addition, the porosity of the dredged cores was low because of an increasing compaction and dehydration of sediment (Table 1), which did not facilitate the transport of oxidants into the deeper sediment. The total number of bacteria and the rate of hydrolytic activity decreased with sediment depth (Mermillod-Blondin et al., 2005). This supports our deduction that the biological activity in dredged sediments was weaker than that in undredged sediments. Consequently, lower mineralization rates of organic matter can be expected in dredged sediments.

At the sediment-water interface upward flux of nitrate plus nitrite hardly occurs (Golterman, 2004), therefore, the concentrations of nitrate plus nitrite in the pore water were low during the experiment (Fig. 2b). Lower concentrations of nitrate plus nitrite in the pore water of the undredged sediments in the warmer months (April–October) indicated the less important role of nitrification (Fig. 2b), probably due to the low oxygen concentration. It has been reported that a strong negative correlation between the temperature of near-bottom waters and the nitrification in surface sediments (Kemp et al., 1990). This is due to the general decrease in oxygen concentration, essential for the maintenance of nitrification, with increasing temperature. Low oxygen concentration will be favorable for denitrification process (Seitzinger, 1988). Under aerobic conditions, intense denitrification processes depleted nitrate plus nitrite in interstitial water in warmer months. The concentrations of nitrate plus nitrite increased in colder months, due to the weak denitrification and intense nitrification process. The same phenomenon was also found for dredged sediments.

3.2 Denitrificaton rates of sediments

In monthly assays, denitrification rates of the undredged sediments were generally greater than those of the dredged sediments (Fig. 3), This result was consistent with other studies that denitrification rate is the highest in the surface sediment layer and decreases with depth (García-Ruiz et al., 1998b; Livingstone et al., 2000). This phenomenon may be attributed to the differences in microbial activity, the content of readily biodegradable organic C and the concentration of nitrate (García-Ruiz et al., 1998b; Livingstone et al., 2000). Additionally, denitrifying bacteria are known to be a heterogeneous group of organisms (Knowles, 1982), thereby, differences in density and composition may be expected in the undredged and dredged sediments. Sediments physical properties such as water content and particle size distribution may also influence the potential denitrification in the dredged sediments (Groffman and Crawford, 2003; D'Haene et al., 2003).

In this study, denitrification in the undredged and dredged sediments were not limited by available nitrate and organic C because the artificial water was enriched with enough nitrate and organic C. Simultaneously, any possible temperature effect on denitrification was also eliminated during the analysis. Denitrification rates in the undredged sediments were positively correlated to the monthly mean temperatures (Fig. 4a). Since an increase in temperature leads to an enhancement of biological activity (Christensen et al., 1988), the biological activity should be considered as the key factor determining the potential denitrification in the sediments. No significant correlation was found between the temperature and the denitrification rate of the dredged sediment (Fig. 4b), suggesting that the biological activity in the dredged sediment was not sensitive to temperature. This result may be attributed to the difference in the number of bacteria and community composition in the undredged and dredged sediments.

3.3 Influence of environmental factors on denitrification

The denitrification rates in both undredged and dredged sediments increased with temperature under experimental condition (Fig. 5). The increase for the undredged sediments was markedly greater than that for the dredged sediments, suggesting that the response of the undredged sediments was more sensitive than that of the dredged sediments. The results were consistent with the observation for the relationships between the denitrification rates and monthly mean temperature during the one-year incubation. Previous studies using sediment slurries reported the exponential increase of denitrification rates within temperature range 14–35.5°C for lake sediments (Messer and Brezonik, 1983), 5–18°C for marine sediments (Seitzinger et al., 1984) and 5–25°C for river sediments (García-Ruiz et al., 1998a).

The results of glucose and nitrate amendments indicated that nitrate concentration was probably one key factor limiting denitrification for both undredged and dredged sediments. Unamended or glucose amended sediments did not stimulate denitrification in the absence of nitrate (Fig. 6). The role of nitrate in limiting denitrification in the sediments of Meiliang Bay is marked after amendment of the sediments with nitrate. Studies have shown that nitrate is usually the most important factor in limiting denitrification in aquatic ecosystems (Esteves et al., 2001; Magalhães et al., 2005).

Low organic C contents of sediments may limit potential denitrification (Morris et al., 1988; Bradley et al., 1995). Additionally, the type of organic C available as an electron donor can also influence potential denitrification in sediments (Pfenning and McMahon, 1996; Baeseman et al., 2006). In this study, the addition of high level of C source (30 mg/L) alone did not lead to a marked increase in denitrification rate in both undredged and dredged sediments (Fig. 6). However, the situation was different for the nitrate plus glucose treatments. For the undredged sediments, denitrification rates had no difference between the treatments with the addition of nitrate and with the addition of nitrate plus glucose, suggesting that organic C did not influence denitrification in the undredged sediments. For the dredged sediments, denitrification rate of the sediments with nitrate plus glucose was significantly greater ($p < 0.05$) than that of the sediments with nitrate alone; suggesting that denitrification in the dredged sediments was limited by the availability of organic C.

Organic C did not affect the denitrification in undredged sediments, which is consistent with the relatively high content of organic C (Table 1). The denitrification rates were limited by organic carbon content in dredged sediments, which is in agreement with Pfenning and McMahon (1996), who have indicated that the availability of organic C generally limits the denitrification potential in buried sediments with less organic C content, but not in surficial sediments with relatively high organic C content. Due to the diagenesis of sediments, dredging the upper 30 cm layer sediments reduced the organic C content in the sediments (Table 1). In addition, the quality of organic matter in the dredged sediments may not be the same as that in the surficial sediment. Thus, additional organic C stimulated the denitrification rates in the dredged sediments due to the shortage of the readily available carbon source as an electron donor.

3.4 Significance of dredging on sediment denitrification

Basic processes to which nitrogen is subjected in the sediments are ammonification, denitrification and nitrification. All the three processes can result in nitrogen release from sediments (Graca et al., 2004). Nitrate release due to nitrification, and ammonium release due to ammonification have adverse effects for the eutrophic water body such as the Meiliang Bay, because both processes will intensify the primary production. Denitrified nitrogen is permanently removed from nitrogen-rich aquatic system, and in this way, denitrification counteracts excessive nitrogen loads (Seitzinger, 1988).

Dredging modified the inherent cycling of nitrogen in the sediments. In our accompanying article, the decomposition of organic matter in the dredged sediments was weaker compared with the undredged sediments, and the ammonium release due to mineralization of organic matters reduced effectively in the dredged sediments (Zhong et al., 2009). However, potential denitrification in the early dredged sediments can not reach as that high rate in undredged sediment, which should be a potential negative effect for nitrogen removal.

4 Conclusions

Dredging the upper 30 cm of sediment can significantly affect physico-chemical characteristics of sediments in the study area, and the changes of sediment characteristics would influence nitrogen transformation in the dredged sediments. The mineralization of organic matters in the dredged sediment was inhibited, which can lead to reduction in release of ammonium at the sediment-water interface. It is a positive role of dredge for pollution control of eutrophic lake. The dinitrification in the dredged sediments was also inhibited, and the denitrification rates of the dredged sediments were generally lower than those of the undredged sediments. Sediment dredging changed the pattern of inherent cycling of nitrogen in the sediments.

Acknowledgments

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