Environ Geol (2008) 55:1–8
DOI 10.1007/s00254-007-0957-6

ORIGINAL ARTICLE

Nitrogen dynamics in large shallow eutrophic Lake Chaohu, China

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Received: 24 March 2007 / Accepted: 11 July 2007 / Published online: 8 August 2007

Abstract Temporal and spatial dynamics of nitrogen in lake and interstitial water were studied monthly in a large shallow, eutrophic lake in subtropical China from October 2002 to September 2003. The distribution of nitrogen was consistent with the idea that high nitrogen concentrations in the western part of the lake resulted from high levels of the nutrients from the surrounding cities through sewage–drainage systems. Nitrate was the predominant form of nitrogen in the overlying water, while ammonium was predominant in the interstitial water, indicating that strong oxidative nutrient regeneration occurred near the sediment–water interface. Nitrate could be an important dissolved inorganic matter source for phytoplankton, which in turn influenced the seasonal variations of nitrate concentrations in lake water. Significant positive correlation between ammonium fluxes and water temperature was observed and could probably be attributed to the intensified ammonification and nitrate reduction with increased temperature. Positive correlation between ammonium fluxes and algae biomass and Chl *a* concentrations may indicate that phytoplankton was an important factor driving ammonium fluxes in our study lake, and vice versa that higher fluxes of ammonium supported a higher biomass of the phytoplankton.

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Keywords Nitrogen · Eutrophic · Flux · Sediment–water interface · Lake Chaohu

Introduction

Many freshwater lakes undergo eutrophication because of increased input of nutrients, e.g. nitrogen and phosphorus from anthropogenic sources. The dynamics of macronutrients such as nitrogen and phosphate have been vigorously studied because their availability is one of the most important controlling factors for primary production (Ryther and Dunstan 1971; Schindler 1977; Carpenter et al. 1998). Although phosphate is usually thought of as the major limiting nutrient of primary production in lake environments (Schindler 1977), it has been suggested that at least in eutrophic lakes, nitrogen could be a more important factor (Elser et al. 1990; Downing and McCauley 1992). Therefore, understanding the natural cycling of nitrogen (N) in eutrophic lakes is of key importance. Besides external input and regeneration in water columns, inorganic nitrogen released from sediments is an important nitrogen source for primary production (Gardner et al. 1987), especially in shallow lakes (Harrison 1980). The sediment–water interface plays a major role in N cycling of aquatic ecosystems through mineralization of particulate or dissolved organic N including ammonification, nitrification, denitrification (Blackburn and Sorensen 1988). Such processes result in exchange of DIN (dissolved inorganic nitrogen) at the sediment–water interface and accumulation of ammonium in the interstitial water (Denis et al. 2001, Hasegawa and Okino 2004). The immobilization of nutrients in sediments and the rate of regeneration are related to a series of interacting physical, chemical and/or metabolic processes. Factors that can alter the exchange balance are, the capacity of sediments to retain nutrients, the conditions of the overlying water (e.g. dissolved oxygen, redox potential and pH), and biotic composition (Redshaw et al. 1990; Staundinger et al. 1990).

With regard to nitrogen, sediments generally represent an important source of ammonium and a sink for nitrate (Seitzinger 1988; Denis et al. 2001; Liikanen et al. 2002; Liikanen and Martikainen 2003; Hasegawa and Okino 2004). Denitrification is known to remove nitrogen by bacterial reduction of nitrate to nitrogen gas via nitrite in anoxic sediment environments, and may be based on both nitrate diffusing from the water column into the sediments and on nitrate produced by nitrification (Seitzinger 1988; Howarth et al. 1988). Ammonium in interstitial water usually originates from two important biological processes: bacterial deamination of organic matter and excretion by benthic organisms. In aerobic conditions ammonium can be oxidized to nitrate during nitrification, assimilated by microphytobenthos and/or bacteria, and involved in exchange reactions between the adsorbed and interstitial-water ammonium pool (Kemp et al. 1990; Van Luijn et al. 1999). Ammonium inputs from the sediment can be important seasonally (Sornin et al. 1990; Souchu et al. 1998) and serve as a main source of N for phytoplankton (Gardner et al. 1987; Collos et al. 2003). Planktonic algae are capable of using both ammonium and nitrate as N sources. For example, DIN exchange at the sediment–water interface may provide a large part of the nitrogen requirements for phytoplankton, up to 30–80% in temperate estuaries (Blackburn and Henriksen 1983).

Previous studies in aquatic systems have suggested that ammonium fluxes may be regulated by several factors including type of sediment, oxidation of organic matter, temperature and dissolved oxygen concentrations. For example, Dorota and Halina (2001) have reported that diffusive ammonium fluxes were significantly different in relation to sediment type ($p < 0.05$). Their values were higher in sediment with the largest silty-clay fraction than with the largest sand fraction. Massive input of organic matter settling on surface sediment increased DIN fluxes at the sediment–water interface (Cowan et al. 1996; Denis et al. 2001). However, little is known about the effect of planktonic primary producers on ammonium flux in sediment–water interface in freshwater ecosystems.

Lake Chaohu is one of the five largest freshwater lakes in China and has undergone serious eutrophication for decades. Based on a series of investigations, phosphate was usually thought of as one of the major limiting nutrients of primary production in lake environments (Xu et al. 2005; Zhang et al. 2006). However, at least in eutrophic lakes, it has been suggested that nitrogen could be an important limiting factor (Elser et al. 1990; Downing and McCauley 1992). In Lake Chaohu, the dynamic patterns of nitrogen species and the role of the sediment in nitrogen cycling are still not well understood, although its shallowness implies a strong interaction between its sediment and water column. Therefore, this investigation was conducted from October 2002 to September 2003 in Lake Chaohu and the objectives were to estimate seasonal, spatial and vertical variations of nitrogen species in lake water and interstitial water, and describe how sediment–water diffusive fluxes of ammonium vary seasonally and spatially, and discuss the possible controlling factors in this shallow eutrophic lake.

Materials and methods

Description of study lake

Lake Chaohu is located in the delta of the Yangtze River in southeastern China, between 117°16′E and 117°5′E longitude and between 31°25′N and 31°43′N latitude. It has a mean surface area 770 km^2, a mean depth of 2.7 m and a storage capability of 2.1 billion m^3 (Fig. 1). Prior to the 1950s, it was well known for its scenic beauty and richness of aquatic life. Since that time, however, the lake has undergone serious eutrophication. Increasing pressures from the population and economic development in the drainage area are responsible for the current state. A great amount of industrial wastewater and domestic sewage were discharged into the lake (Shang and Shang 2005). The results have created negative ecological, health, social, and economic effects on the lake and its use (Shang and Shang 2005; Xu et al. 2003, 2007). In the warm seasons of each year, the lake is eutrophic with dense cyanobacterial blooms (mainly composed of *Microcystis* and *Anabaena*) (Xu et al. 2007).

Sampling and assay

Two water column and three sediment samples were collected at six locations (Fig. 1) at monthly intervals from October 2002 to September 2003. Sites I, II and III were located in the western part of this lake, and Sites IV, V and VI in the eastern part. Water samples were collected at the surface (0.5 m) and overlying water. Sediment cores were obtained using a hand-driven stainless steel corer (100 cm long with an internal diameter of 3.9 cm). For depth profiles, sediment columns were sliced into three layers: 0–10 cm, 10–20 cm and 20–30 cm. The interstitial water was separated from the sediment particles by centrifugation at 3,000 rpm for 30 min (Degobbis et al. 1986) immediately. The supernatants and water samples were then filtered through a 0.45 μm membrane filter.

Ammonium was determined by the Nessler method, nitrate by ultraviolet spectrophotometric screening method, and nitrite by the α-naphthylamine method (Xu et al. 2005).

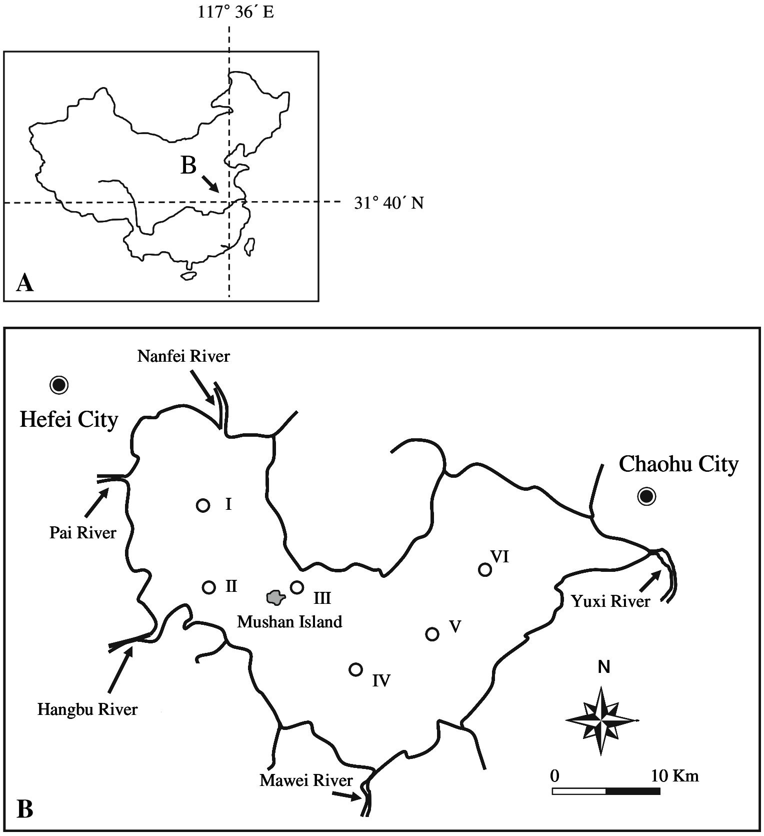


Fig. 1 Location of Lake Chaohu in China (**a**) and the sampling sites in this study (**b**)

Total nitrogen (TN) was measured by alkaline potassium persulfate digestion-UV spectrophotometric method (APHA 1995). Mean porosity and grain size in surface sediments were carried out according to Hua and Wang (1993). Sediments were air-dried and sieved through a 0.149-mm mesh. Total nitrogen in sediment was measured by BUCHI 339 Kjeldahl Analyzer Unit.

Diffusive flux calculation

Diffusive fluxes (F_{Diff}) were estimated according to Fick's first law by calculating a linear gradient between the surface interstitial water concentration and the bottom water concentration of solute (C) in the sediment:

$$F_{\mathrm{Diff}} = -\phi D_{\mathrm{S}}^{T}(\delta C/\delta z) \tag{1}$$

with $D_{\mathrm{S}}^{\mathrm{T}}$, the effective diffusion coefficient of the solute C in the sediment, ϕ, the sediment porosity, and z the depth in the sediment.

The effective diffusion coefficient of a solute in the sediment must take into account the convoluted path that molecules must follow around sediment particles (i.e. tortuosity θ). Following Berner (1980), we used Archie's law ($\theta^2 = \phi^{(1-\mathrm{m})}$), with the empirical coefficient $m = 3$ (Ullman and Aller 1982). The effective diffusion coefficient ($D_{\mathrm{S}}^{\mathrm{T}}$) of a solute in the sediment was then calculated as follows:

$$D_{\mathrm{S}}^{T} = D^{T}\theta^{-2} = D^{T}\theta^{2} \tag{2}$$

where D^T (cm^2 s^{-1}) is the infinite dilution diffusion coefficient at the ambient temperature T (°C). This value was calculated from the zero-degree coefficient $D°$ (cm^2 s^{-1}) according to Boudreau (1997):

$$D^{T} = D^{\circ} + a\,T \tag{3}$$

where a (cm^2 s^{-1} °C^{-1}) is an ion-specific coefficient.

In the calculation of diffusive fluxes, the diffusion coefficient D_S^T only includes molecular diffusion around sediment particles. The calculated diffusive fluxes must therefore be considered as minimal diffusive fluxes (only diffusion; Callender and Hammond 1982; Emerson et al. 1984).

Results

Sediment characteristics

The porosity, granulometric characteristics and total nitrogen in sediment of the six sampling sites were shown in Table 1. Mean porosity of the surface sediment was slightly higher in the western than in the eastern areas, ranging from 0.67 to 0.75. The sediments of all the six sites almost

Table 1 Average of physical and chemical properties of sediments in Lake Chaohu

	I	II	III	IV	V	VI
Mean porosity (mL cm^{-3})	0.75	0.74	0.72	0.69	0.72	0.67
Grain size (% dw)						
Sand	2.1	2.6	4.3	4.9	6.1	3.1
Silt	88.2	91.1	92.3	92.0	88.6	88.7
Clay	9.1	6.0	3.4	2.9	4.0	6.3
TN (mg g^{-1})	0.65	0.60	0.65	0.49	0.52	0.58

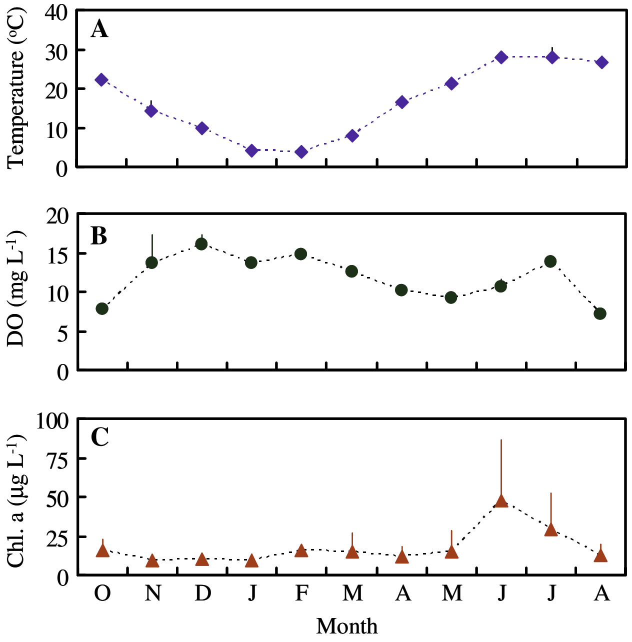


Fig. 2 Seasonal variations of temperature (**a**), dissolved oxygen (**b**) and Chl *a* (**c**) in the water column of Lake Chaohu. Mean values were calculated from data of all sampling sites. *Error bars* represent one standard deviation unless smaller than symbol

entirely comprised silt (>88.2%). The clay content was obviously higher at station I than at the other stations. The concentrations of total nitrogen were significantly higher in the western sites I, II, III than in the eastern sites IV, V, VI (*t* test, $p < 0.05$).

Lake and interstitial water

Water temperature ranged from 3.8°C in February to 28.0°C in July (Fig. 2a) with an annual average of 16.6°C. Dissolved oxygen concentrations varied between 16.0 and 7.2 mg L^{-1} (Fig. 2b), and Chl *a* concentration changed between 9.4 and 47.9 μg L^{-1} with peak values occurring in June (Fig. 2c).

Concentrations of nitrate, nitrite and TN in the lake water exhibited a consistent spatial pattern decreasing from station I to station VI (Fig. 3a, c, d), and were significantly higher in the western sites (sites I, II, III) than in the eastern sites (sites VI, V, VI; *t* test, $p < 0.01$). However, ammonium concentrations in the lake water showed inconspicuous spatial variation (Fig. 3b). In the interstitial water, both ammonium (Fig. 3b) and TN (Fig. 3d) concentrations were significantly higher in the western sites than in the eastern sites (*t* test, $p < 0.01$).

Vertical profiles of nitrate and ammonium in the lake and interstitial water were shown in Fig. 4. Nitrate was dominant in the lake water, while ammonium was dominant in the interstitial water. In both the western and eastern areas, there was no significant difference in mean nitrate concentrations between the surface and overlying

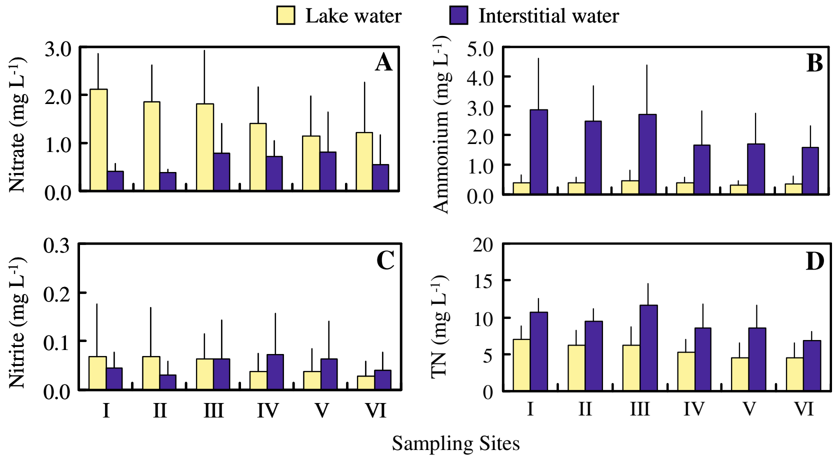
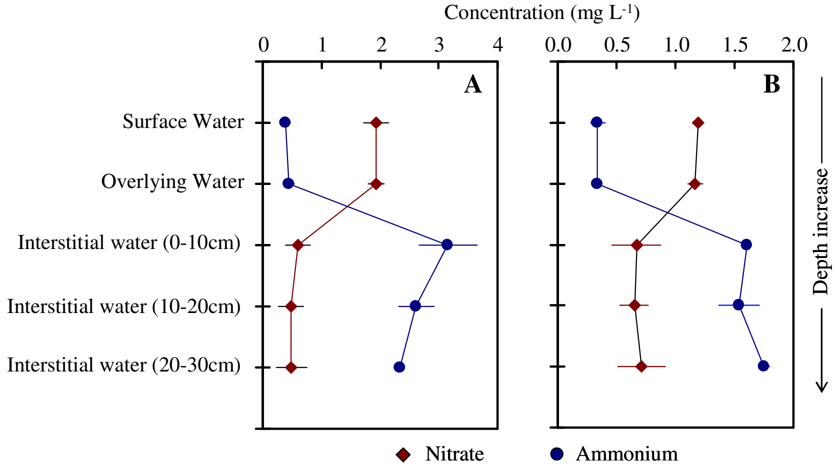


Fig. 3 Spatial changes in nitrate (**a**), ammonium (**b**), nitrite (**c**) and TN (**d**) concentrations of the lake and interstitial water from Lake Chaohu. Mean values were calculated from data of all sampling dates. *Error bars* represent one standard deviation unless smaller than symbol

Fig. 4 Vertical changes of nitrate and ammonium in the lake and interstitial water in the western (**a**), Sites I, II, III, and eastern (**b**), Sites IV, V, VI, areas of Lake Chaohu. Mean values were calculated from data of all dates. *Error bars* represent one standard deviation unless smaller than symbol



water. Vertical patterns of ammonium concentrations in the interstitial water were different spatially. In the western sites the maximum ammonium concentration occurred in the surface sediment with gradual declines towards deeper sediments (Fig. 4a), whereas in the eastern sites the maximum was observed in the deeper sediment layers (Fig. 4b). The vertical changes of nitrate concentrations in the interstitial water were relatively stable.

Temporal variations of total nitrogen, nitrate and ammonium concentrations in the lake and interstitial water were shown in Fig. 5. Nitrate concentrations in the lake water changed between 0.50 and 2.65 mg L^{-1} (Fig. 5a) with the minimum in summer. Ammonium was the predominant dissolved nitrogen species in the interstitial water with the maximum in summer (Fig. 5b). Comparatively, nitrate in the interstitial water and ammonium in the lake water (Fig. 5b) showed no apparent seasonal variations.

Diffusive ammonium fluxes

In most situations, ammonium fluxes were directed from the sediment to the water column. Ammonium fluxes were significantly higher in the western sites than in the eastern sites (Fig. 6). Ammonium fluxes increased significantly with water temperature (Fig. 7a), algae biomass (Fig. 7c) and Chl *a* (Fig. 7d) concentrations, and decreased significantly with dissolved oxygen in the lake water (Fig. 7b).

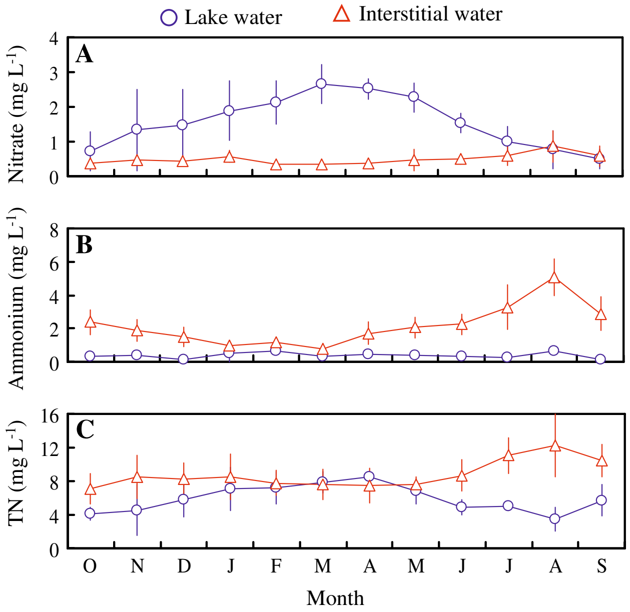


Fig. 5 Seasonal variations of nitrate (**a**), ammonium (**b**) and TN (**c**) in the lake and interstitial water of Lake Chaohu. Mean values were calculated from data of all sampling sites. *Error bars* represent one standard deviation unless smaller than symbol

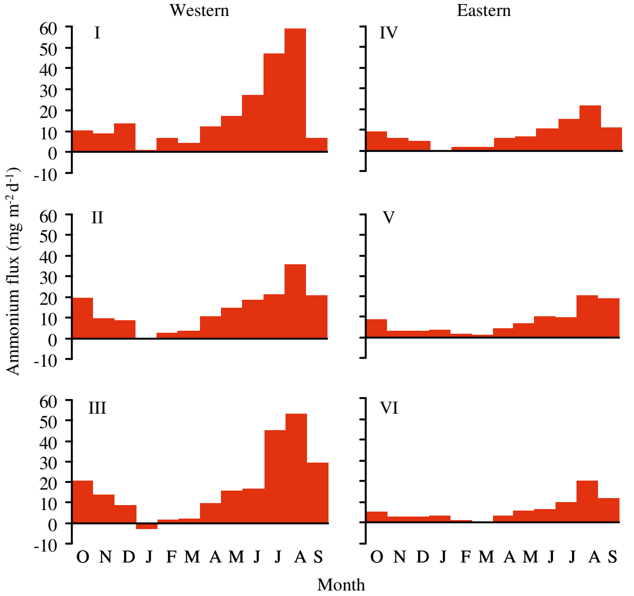


Fig. 6 Temporal and spatial changes of ammonium flux at the sediment–water interface in Lake Chaohu

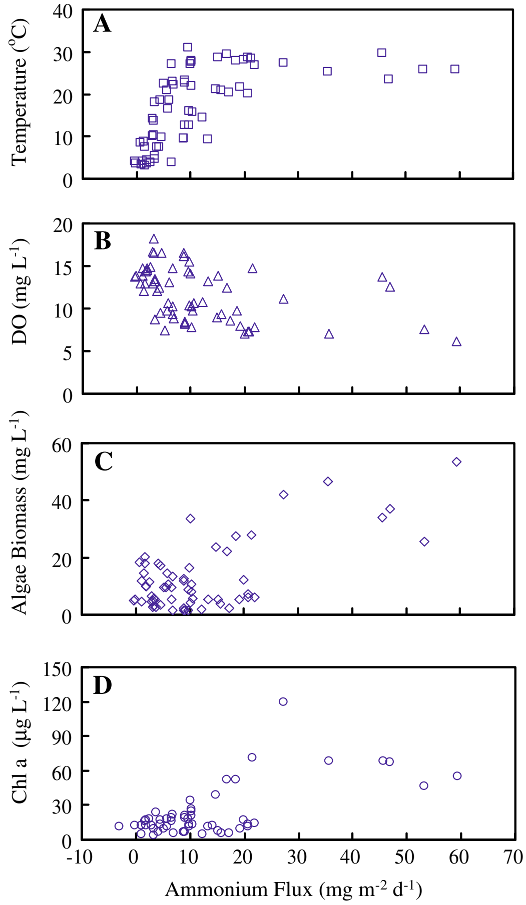


Fig. 7 Correlation of ammonium fluxes with temperature (**a**), dissolved oxygen (**b**), algae biomass (**c**) and Chl *a* concentrations in the lake water of Lake Chaohu. Significantly positive correlations were found between ammonium fluxes and temperature ($r = 0.60$, $p < 0.0001$, $n = 63$), algae biomass ($r = 0.67$, $p < 0.0001$, $n = 63$) and Chl *a* concentrations ($r = 0.65$, $p < 0.0001$, $n = 56$) of lake water. Significantly negtive correlation was found between ammonium fluxes and dissolved oxygen ($r = -0.42$, $p < 0.001$, $n = 63$)

Discussion

Vertical and spatial variations of nitrogen

Higher total nitrogen concentrations in the western sites could be attributed to high levels of nutrient inputs from the surrounding cities because the previous studies showed that nitrogen inputs from the rivers in the western part of this lake accounted for 79% of whole-lake nitrogen discharging from the rivers (Tu et al. 1990). TN increased with increasing clay content with the exception of site II, which may reflect the greater sorptive capacity of clay minerals relative to silt and sand. Positive correlation between clay content and nitrogen content in the sediment was also reported in previous studies (Lu et al. 2005). Nitrate was found predominantly in the overlying water, while ammonium dominated the interstitial water, indicating a strong oxidative nutrient regeneration at sediment–water interface (Wu et al. 2001; Landing et al. 1991). Because of shallowness and vertical mixing of the lake water all the year, vertical variations of nitrate and ammonium concentrations in the water column were not observed in this lake. The decreases in nitrate concentrations in both the eastern and western portions of Lake Chaohu, probably indicated the microbial denitrification in the absence of adequate DO and under anaerobic conditions in this shallow eutrophic lake. Though related properties of sediments, e.g. absorptive capacity, DO, redox potential and pH, were not analyzed, previous studies carried out in the eutrophic freshwater lakes with increased input of nutrients, e.g. nitrogen and phosphorus, from anthropogenic sources demonstrated that the excess availability of nutrients not only stimulates primary production but also has significant effects on microbial processes (e.g. Liikanen et al. 2002). The elevated mineralization consumes oxygen (O_2) in the overlying water and interstitial water, leading to lake anoxia (the absence of adequate DO) (Smith et al. 1999), favoring anaerobic microbial processes, e.g. denitrification (Hasegawa and Okino 2004). The increase of ammonium in the interstitial water in the eastern sites with sediment depth might be mainly related to regeneration by anaerobic metabolism with the development of anoxia as generally reported in other studies (e.g. Liu et al. 2002). However, high ammonium in the interstitial water was observed in the surface sediment, which might be attributed to strong mineralization at the surface layers of the sediment because the organic matter content of surface sediments is higher in the western than in the eastern areas (Tu et al. 1990).

Seasonal variations of nitrogen and correlation with phytoplankton

Nitrate concentrations in the lake water showed obvious seasonal changes in this study, while ammonium did not. It has been documented that ammonium is often used preferentially over nitrate when both substrates are available (e.g. Berman et al. 1984; Gu et al. 1997), and therefore more obvious seasonal changes in ammonium than in nitrate can be expected. However, both obvious seasonal changes in nitrate concentrations in this study and a significantly negative correlation between nitrate concentration and algae biomass by Deng (2004) indicated the effective utilization of nitrate by phytoplankton in this eutrophic lake. This was supported by previous studies that distinct seasonal trends of nitrate concentrations in lake water indicate the winter stock of nitrate and the utilization of nitrate during the summer bloom (Temponeras et al. 2000).

Diffusive ammonium fluxes

Diffusive ammonium fluxes in Lake Chaohu always moved in the direction from the sediment into the water column, and the fluxes in the western sites were relatively high where there was higher organic matter input and hence intense mineralization processes at the sediment–water interface (Cowan et al. 1996; Liikanen and Martikainen 2003). Significant positive correlation between ammonium fluxes and water temperature could probably be attributed to the intensified ammonification and nitrate reduction with increased temperature (Dorota and Halina 2001; Liikanen and Martikainen 2003). There was a negative correlation between ammonium concentrations in the sediments and dissolved oxygen concentrations in Lake Chaohu. It seems that the elevated ammonium release may be caused not only by increased ammonification of organic matter in the sediments, but also by decreased nitrification due to low-oxygen concentrations (Dorota and Halina 2001; Liikanen and Martikainen 2003). In the present study, there were significantly positive correlations between ammonium fluxes and algae biomass and Chl *a* concentrations. These correlations may indicate that phytoplankton was an important factor driving ammonium fluxes in our study lake, because algae assumably utilized nitrogen in the lake water and created a physical diffusion gradient, which in turn triggered the release of nitrogen from the surface sediment. Meanwhile, these observations might also be explained by the fact that the higher fluxes of ammonium supported a higher biomass of the phytoplankton. However, other factors, e.g. desorption and ammonium uptake less due to crowding of phytoplankton, could not be excluded and would be incorporated in further studies.

Acknowledgments We are very grateful to H. Yang, S. Wu and Y. Xing for field sampling. Thanks to Dr. Wayne S. Gardner and the anonymous reviewers for the English corrections and their comments. This work was supported by a fund from National Natural Science Foundation of China (30530170) and a key project of CAS (Grant No. KSCX2-SW-129).

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