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Variation in sources of inorganic nitrogen under different hydrological conditions in a floodplain lake: a case study of Bang Lake (Poyang Lake, Jiangxi Province, China)

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ABSTRACT

Bang Lake, a shallow floodplain lake in Jiangxi Province, China, is a complex environment with water levels and sources of pollutants that vary considerably on an annual scale. We collected rain and water samples from the lake and around connecting rivers through lentic, flowing, flooding, and lowering level periods. We distinguished sources of dissolved inorganic nitrogen (DIN as ammonium NH_4^+ and nitrate NO_3^-) in the lake from $\delta^{15}\text{N}$. Stable isotopes of hydrogen (δD), oxygen ($\delta^{18}\text{O}$), and $\delta^{15}\text{N}$ ($\delta^{15}\text{NH}_4^+$, $\delta^{15}\text{NO}_3^-$) in water samples were used to describe lake–river water exchanges and DIN sources at different water levels in the floodplain lake. The sources of DIN varied through the different hydrological stages in the lake. In the lentic periods, $\delta^{18}\text{O}$ and δD values were more positive and $\delta^{15}\text{N}$ ($\delta^{15}\text{NH}_4^+$, $\delta^{15}\text{NO}_3^-$) values were more negative in lake water than in river water and rainwater, indicating lake–river separation. During the flowing period, $\delta^{18}\text{O}$, δD , and $\delta^{15}\text{N}$ values in river and lake water were similar, and DIN mainly derived from agriculture and livestock wastewater. During the flooding period, $\delta^{15}\text{N}$ values in the lake and rivers differed even though they were connected at the highest water levels. When the water levels were declining, considerable variation was measured for $\delta^{15}\text{NH}_4^+$ or $\delta^{15}\text{NO}_3^-$ signatures between the lake and river water as the lake and river water gradually separated. The significant increase of $\delta^{15}\text{NH}_4^+$ and decrease of $\delta^{15}\text{NO}_3^-$ indicated strong ammonification and nitrification in the lake.

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floodplain lake; hydrology; inorganic nitrogen; Poyang Lake; source; stable isotope

Introduction

Floodplain lake dynamics can be driven by hydrologic processes under the influence of seasonal precipitation, including hydrologic connectivity during floods and separation at low flow with adjacent or inflowing rivers. That is, the floodplain lake is close to rivers receiving fluvial water during flood periods via direct connections and may temporarily exhibit some lotic characteristics during the flowing period. By contrast, longitudinal flow may cease during dry seasons (Hu et al. 1997). Floodplain lakes are therefore different from temperate lakes in various ways, such as nutrient sources, water residence time, and water quality state, mainly because of differences in the water exchange mechanisms and dynamics (De Emiliani 1997, Harrison et al. 2009). The sources and transformation processes of N in the floodplain lake vary in response to the hydrological processes at different water levels. This study will help in managing nutrient input and controlling pollution in the floodplain lake.

Excessive N inputs increase the trophic status of lakes; for example, about 75% of lakes (defined as having an area $>1 \text{ km}^2$) studied in China are either mesotrophic or hypereutrophic (Zhao et al. 2010). Many of these are floodplain lakes in the middle and downstream reaches of the Yangtze River, such as Taihu, Dongting, and Poyang lakes. Floodplain lakes have many N input sources, the most important of which is atmospheric wet deposition; for example, increasing N concentrations have been reported in precipitation in south China (Zhang et al. 2011). Mineralization is also an important N source in internal lakes, the main form of which is ammonium nitrogen (NH_4^+). In addition, N that derives from urban storm runoff, agricultural fertilizers, livestock and poultry farming, and rural residues, all of which are widespread in the south of China because of rapid agricultural development and rapid urban population growth (Le et al. 2010), is discharged into lakes from inflowing rivers.

Stable isotopes provide an effective way to trace sources of pollutants and hydrological cycling at the catchment scale (Ichiyanagi et al. 2003, Chen et al. 2006, Zhao et al. 2011). Water stable isotopes of oxygen and hydrogen ($\delta^{18}\text{O}$ and δD , respectively) are ideal conservative tracers of water movement and provide an integrated view of sources because they are part of the water molecule (Coops et al. 2003, Yin et al. 2011). Information about lake hydrological processes, as indicated by $\delta^{18}\text{O}$ and δD , is essential for understanding lake biogeochemical cycling and biological conditions in lakes (Sokal et al. 2008, Brooks et al. 2014). Stable isotopes of dissolved inorganic nitrogen (DIN as $\delta^{15}\text{NH}_4^+$ and $\delta^{15}\text{NO}_3^-$) are used mainly to account for flow responses and to trace natural or anthropogenic sources and transformations of N in aquatic ecosystems (Ogrinc et al. 2008, Gu 2012, Yoshikawa et al. 2016). Stable isotopes have also been widely used to investigate water level fluctuations caused by seasonally available precipitation or flood events in aquatic environments (Schemel et al. 2004, White et al. 2008). At different water levels in rivers or lakes, runoff in channels and complex overland flow patterns produce various isotopic signatures (Wantzen et al. 2008), such as $\delta^{15}\text{N}$ values ranging from 6‰ to 22‰ in sewage (Dillon et al. 2007) and depleted ^{15}N ranging from -27 ‰ to 10‰ in rainwater (Gobel et al. 2013, Shrestha et al. 2013), and these different signatures can reflect different hydrological processes (Wolfe et al. 2007). In addition, biogeochemical cycling and transformations of N in internal lakes, including assimilation, nitrification, and denitrification (Gardner et al. 2006), are closely associated with variations in isotope signatures (Kendall et al. 2007). ^{14}N participates in a reaction prior to ^{15}N formation, enriching ^{15}N in the source or substrate but depleting ^{15}N in the product if the conversion is incomplete (Kendall 1998, York et al. 2007, 2010), but N fractions will be lower in sedimentation and biologically fixed and mineralized N (Xiao and Liu 2010).

Bang Lake, a shallow floodplain lake, is located at the edge of Poyang Lake, China, connected to 2 rivers during flood or high flow periods but separated from them during lentic periods. Between the wet and dry seasons, the water level fluctuates by about 6 m. Previous studies reported that Bang Lake is either mesotrophic or eutrophic (Hu et al. 2013, Liang et al. 2015) depending on the water level; however, the sources of pollutant loading have not been described in the context of the hydrological conditions of the river–lake system. To improve understanding of DIN from natural and anthropogenic sources at different water levels of the floodplain lake, we used stable isotopes (δD , $\delta^{18}\text{O}$, $\delta^{15}\text{N}$) in Bang Lake and connected waters, different pollutant origins, and rainfall to (1) investigate hydrological connectivity

between the river and lake water levels; (2) characterize DIN sources, including domestic sewage, cropland runoff, livestock effluent, urban rainfall runoff, and rainfall from river catchments; and (3) trace sources and transformations of DIN in the lake under different hydrological conditions.

Methods

Study area

Poyang Lake, located in the middle and lower reaches of the Yangtze River ($28^{\circ}22'–29^{\circ}45'\text{N}$, $115^{\circ}47'–116^{\circ}45'\text{E}$; Fig. 1), is the largest freshwater lake in China (Hu et al. 2007). Its water levels change dramatically over the course of a year (Zhang et al. 2011). In the dry season, Poyang Lake has a water area of only 216.62 km² and a water depth of 9 m with many small lakes at its edge. By contrast, because of water from receiving rivers in the wet season, Poyang Lake quickly expands to link with small lakes at its edge, reaching a water area of 3218.29 km² and a water depth of 20 m. Pollutants often occur and accumulate in the small lakes at the edges of Poyang Lake because of slow water velocity.

Bang Lake, with an area of 80 km², is one of several lakes at the edge of Poyang Lake with water level fluctuations similar to those in Poyang Lake. The Gan and Xiu rivers are the 2 main inflows to the Poyang Lake basin. Their tributaries also flow into Bang Lake, and the rivers are separated from the lake in the dry season by a natural dam (Fig. 1). The rainy season is in spring and summer (Apr–Aug), during which >56% of the annual rainfall occurs. Fall and winter (Sep–Feb) are dry and arid seasons, respectively. The water temperature in the catchments generally ranges from 2 °C in January to 32 °C in August. Approximately 80% of Gan and Xiu catchments are covered by agricultural land. The river catchments comprise some cities, county towns, and village residential areas. Soils in the catchments are fertile and support pastoral and other types of agriculture and forestry.

The river–lake connectivity results from seasonal precipitation. The water level in Bang Lake changes by about 6 m between the dry and wet seasons. The river is separated from the lake when the water depth in Bang Lake is <0.8 m in the lentic period (Dec–Apr). The river and lake are possibly connected during the flowing period in May, when the water depth is between 1 and 4.5 m, as well as during the flooding period from June to August, when the water depth is between 4.5 and 6.5 m. The river and lake may be separated when the water levels are lowering

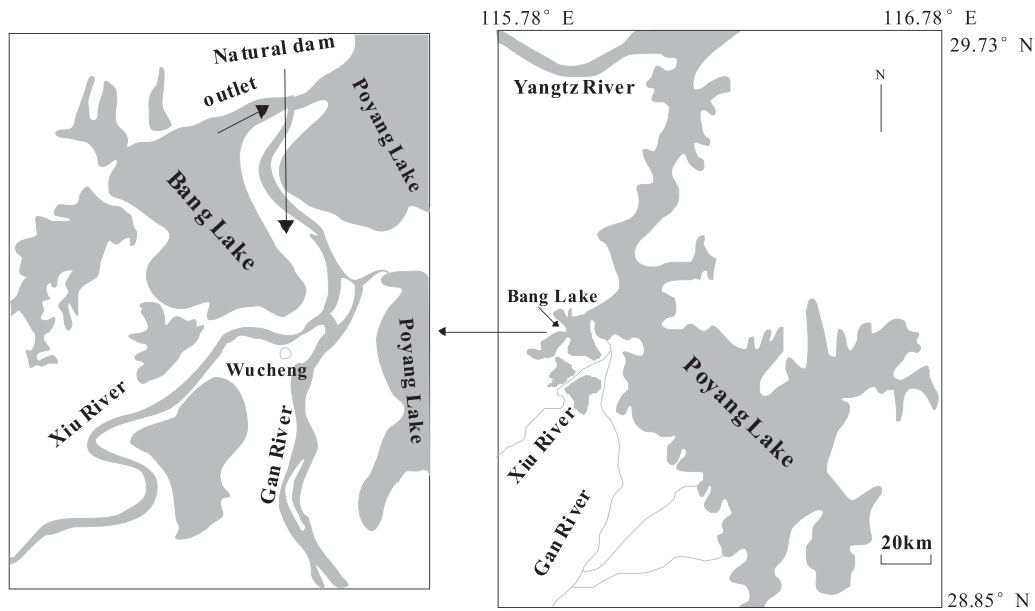


Figure 1. Location of Bang Lake and its surrounding watersheds.

in September and October and the water depth is <3 m. Therefore, the sources of pollutants vary at different water levels in Bang Lake. As a microcosm of Poyang Lake, studies of Bang Lake can be used to represent Poyang Lake.

Sample collection

During June 2012 to May 2013, 8 sampling sessions were conducted in Bang Lake and the adjacent rivers (the north branch of Gan River and the downstream area of Xiu River; Fig. 2). The sites were selected according to the unique geographical location of Bang Lake and variation of water level caused by the seasonal precipitation and different water depths during flooding (5–6 m), lowering (2–3 m), lentic (0.5 m), and flowing (3–4 m) periods. The same sites were not sampled every time

because of large changes in water level; for example, in the dry season, Bang Lake was actually a wetland, so samples were collected from small lakes. About 15 to 20 samples were collected each session. Water temperature, pH, and dissolved oxygen (DO) were measured *in situ* with a meter (HACH Co., Loveland, CO, USA). On collection, a small amount of each sample was immediately filtered through a 0.45 μm acetate membrane into a clean brown plastic bottle to determine the concentrations of N forms within 12 h. Additional aliquots were filtered through precombusted 0.7 μm filters (Whatman GF/F) and stored in plastic bottles, preserved with HgCl_2 , and stored at 4 $^{\circ}\text{C}$ for isotopic analysis.

From 2012 to 2013, water samples from the catchments were also collected from potential origins of N pollution, including fish ponds, urban sewage outfalls,

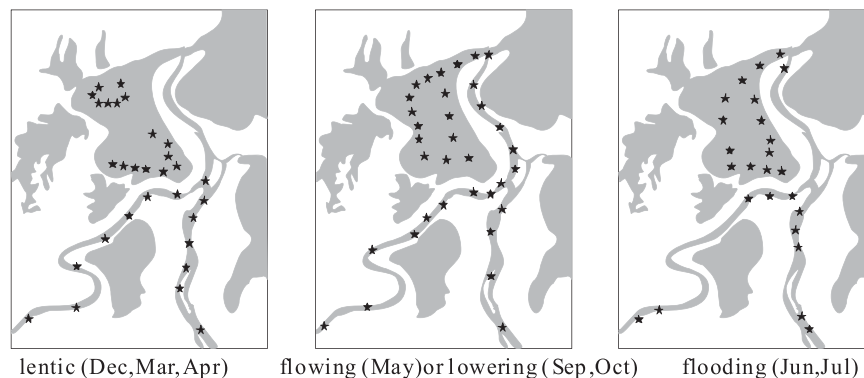


Figure 2. Sampling sites at different water levels in Bang Lake.

vegetable runoff from fields, runoff from rice fields, and pig farm outfalls. From April to June 2103, 25 rainwater samples were collected from a city in the north branch of the Gan River, 80 km from Bang Lake. All water samples were subjected to the same pretreatment before chemical concentrations, and isotopic compositions were determined.

Laboratory analysis

Samples were treated with Nessler's reagent before ammonium ($\text{NH}_4\text{-N}$) concentrations were measured by spectrophotometry, and nitrate (NO_3^-) concentrations were measured by ion chromatography (Dionex DX 500), both with a detection limit of 0.1 mg/L. Standard reference materials were provided by the Chinese National Research Center for Geo-Analysis.

NO_3^- and NH_4^+ in water samples were separated by continuously siphoning through ion exchange columns without adding air, with the cation exchange column (Dowex 50W-X8, 50-mesh, H^+) placed before the anion exchange column (Dowex 1-X8, 200-mesh, OH^-). NO_3^- and NH_4^+ from the resins were eluted with 30 mL of 2M KCl solution (Xiao and Liu, 2002). NO_3^- or NH_4^+ was collected from the eluate in a trap after diffusion with 2 mL of 0.1M KHSO_4 . Devarda's alloy was added to the NO_3^- eluate to transform NO_3^- to NH_4^+ . The resin sorption efficiencies of NO_3^- and NH_4^+ were about 99.9% and 98%, respectively. Elution recoveries of both ion resins were all >95% with 2M KCl solution. The diffusion procedure did not cause significant fractionation of N isotopes.

Stable isotopes were determined in the State Key Laboratory of Environmental Geochemistry at the Institute of Geochemistry of the Chinese Academy of Sciences in Guiyang, China. Once cooled and dried after diffusion for $\delta^{15}\text{N}$ analyses, NH_4^+ was loaded into tin capsules and combusted in an elemental analyzer (C/N/S) interfaced with a continuous-flow isotope ratio mass spectrometer (EA-IsoPrime, Euro3000). An $(\text{NH}_4)_2\text{SO}_4$ ($\delta^{15}\text{N}_1 = -30.4\text{‰}$, $\delta^{15}\text{N}_2 = 0.4\text{‰}$, $\delta^{15}\text{N}_3 = 20.3\text{‰}$) standard from the IAEA was used for $\delta^{15}\text{N}$, and the error was $\pm 0.2\text{‰}$.

In addition, the filtered water samples were collected in 20 mL high-density polyethylene bottles for $\delta^{18}\text{O}$ and δD analysis. Values of $\delta^{18}\text{O}$ were measured using $\text{CO}_2\text{-H}_2\text{O}$ equilibration on an isotopic mass spectrometer (Finnigan MAT253; Epstein and Mayeda 1953). Values of δD were determined by the zinc oxidation method on a mass spectrometer (Finnigan Delta E; Kendall and Coplen 1985). We used IAEA SLAP and GISP standards with analytical errors of $\pm 0.2\text{‰}$ for $\delta^{18}\text{O}$ and $\pm 2.0\text{‰}$ for δD .

N and O isotopic data are reported on the permil (‰) scale referenced to:

$$\delta^{15}\text{N} = ([^{15}\text{N}]/[^{14}\text{N}]_{\text{sample}}/[^{15}\text{N}]/[^{14}\text{N}]_{(\text{NH}_4)_2\text{SO}_4} - 1) \times 1000\text{‰}, \text{ and}$$

$$\delta^{18}\text{O} = ([^{18}\text{O}]/[^{16}\text{O}]_{\text{sample}}/[^{18}\text{O}]/[^{16}\text{O}]_{\text{IAEASLAP}} - 1) \times 1000\text{‰}.$$

SPSS 17.0 was used for statistical analyses. The monthly differences among $\delta^{15}\text{N}$ concentrations were statistically assessed using analysis of variance (ANOVA), and the results were plotted in SigmaPlot 11.0.

Results

Isotopic hydrology characteristic

The simple linear regression equation of the isotopic composition of rainwater is $\delta\text{D} = 6.8\delta^{18}\text{O} + 4.6$ in this study (Fig. 3), which differed from the Craig equation ($\delta\text{D} = 8\delta^{18}\text{O} + 10$) and from the equation of rainfall for the entire year ($\delta\text{D} = 8.9\delta^{18}\text{O} + 11.0$; Liu 2012) because of the distinct differences in the isotope masses at different altitudes (Craig 1961), temperatures, seasons, and precipitation amounts (Celle-Jeanton et al. 2004, Liu et al. 2014). $\delta^{18}\text{O}$ and δD values in rainwater spanned wide ranges of δD from -36.8‰ to 8.7‰ and $\delta^{18}\text{O}$ from -5.7‰ to 0.6‰ (Fig. 3). The wide ranges were the result of composite individual rain events in spring and early summer in the warmer wetter season (McDonnell et al. 1990, Liu et al. 2014).

The data from the 2 rivers (Gan and Xiu) clustered around depleted isotopes at one end of the current rainfall line, the annual rainfall line, and the Craig line

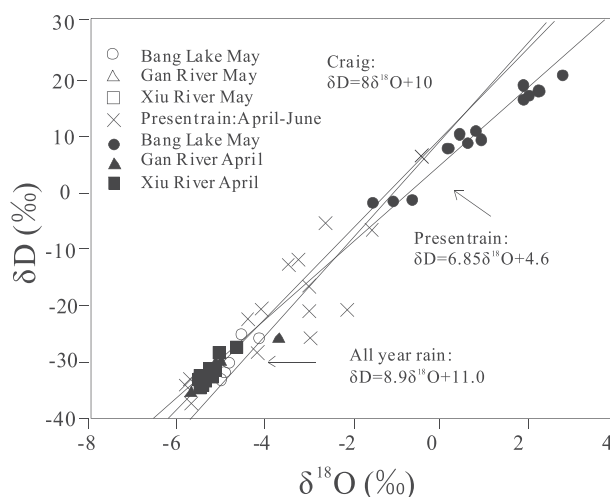


Figure 3. The isotopes distribution of water for Bang Lake and adjacent rivers and Present rain in lentic period (Apr) and in flowing period (May).

(Fig. 3). This result suggested that precipitation was the main source of water for the 2 rivers, and the differences between their isotopic compositions were closely related to meteoric water as well as with the amount of precipitation, the water retention time, and water–rock interactions (McGuire et al. 2005, McGuire and McDonnell. 2006).

The isotopic compositions in Bang Lake were distinctly different as the water levels changed (Fig. 3). In the flowing period (May), the isotopes (δD : -34.9‰ to about -29.4‰ ; $\delta^{18}O$: -5.7‰ to about -4.1‰) were depleted and were close to those of the adjacent rivers. During the lentic period (early Apr), the isotopic compositions of the lake water were distinctly heavier (δD : -2.0‰ to about 20.6‰ ; $\delta^{18}O$: -1.5‰ to about 2.8‰) than those of the rivers. The $[d_{\text{excess}} = \delta D - 8\delta^{18}O = 3.9 < 10]$ indicated strong evaporation from the shallow lake water (Brooks et al. 2014, Gibson et al. 2016). The water depth in the lake is <0.8 m for 6 months through the dry season from November to early of April of the next year, during which the lake is a closed wetland and is separated from the rivers.

Comparison of $\delta^{15}N$ of DIN between the rivers and the lake

The $\delta^{15}NH_4^+$ or $\delta^{15}NO_3^-$ of the lake and the adjacent rivers at different water levels (Fig. 4a–b, Table 1) showed

that during the lentic period (Apr), the $\delta^{15}NH_4^+$ ($-3.1 \pm 4.1\text{‰}$) and $\delta^{15}NO_3^-$ ($-2.2 \pm 0.7\text{‰}$) values in the lake were obviously more negative than the values in the adjacent rivers ($\delta^{15}NH_4^+$: $7.9 \pm 1.9\text{‰}$, $\delta^{15}NO_3^-$: $2.9 \pm 0.6\text{‰}$). The flowing period (May) showed a smaller difference for $\delta^{15}NH_4^+$ and $\delta^{15}NO_3^-$ signatures between the rivers and the lake. In the flooding period (Jun–Jul), when the water level was highest, the values of $\delta^{15}NH_4^+$ or $\delta^{15}NO_3^-$ between the rivers and the lake were distinctly different. For instance, in June, $\delta^{15}NH_4^+$ in the lake was most negative ($-18.6 \pm 5.2\text{‰}$), whereas that of the river waters was relatively enriched with $^{15}NH_4^+$ ($-8.9 \pm 1.8\text{‰}$). In July, $\delta^{15}NO_3^-$ values in the lake ($4.3 \pm 1.3\text{‰}$) were more positive than those in the rivers ($-1.8 \pm 0.8\text{‰}$). In the lowering period, when the water level was decreasing (Sep and Oct), $\delta^{15}NH_4^+$ or $\delta^{15}NO_3^-$ was more variable between the rivers and the lake: $\delta^{15}NO_3^-$ values tended to be negative in the lake ($-1.3 \pm 4.1\text{‰}$) but more positive in the rivers ($5.9 \pm 0.5\text{‰}$).

The $\delta^{15}N$ values of DIN in lake water

The $\delta^{15}NH_4^+$ and $\delta^{15}NO_3^-$ values of Bang Lake differed significantly as the water levels varied (Fig. 5a–b). The $\delta^{15}NH_4^+$ was most negative (-27.1‰ to about -7.0‰) during the flooding period when the water level was highest. As the water level declined, $\delta^{15}NH_4^+$ gradually

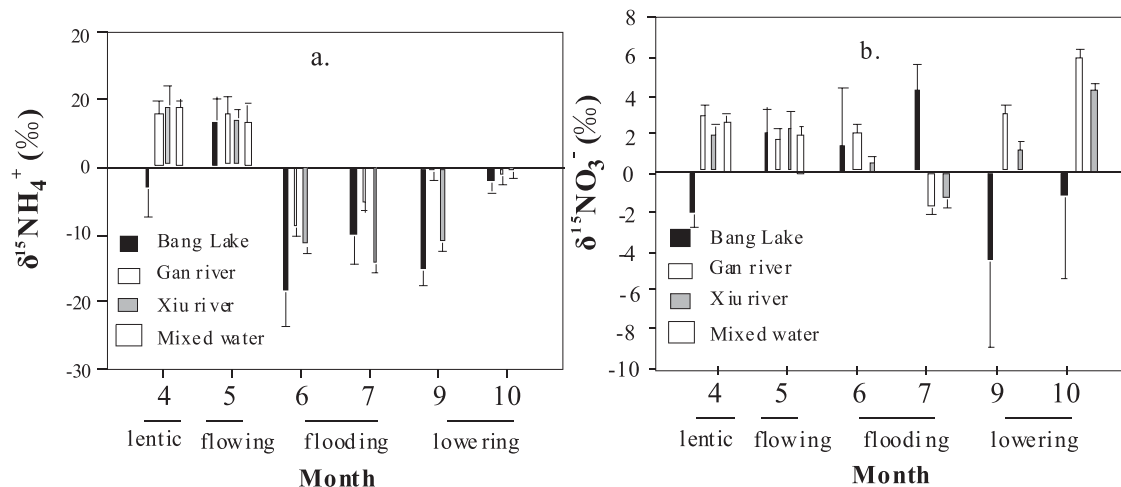


Figure 4. Monthly comparison for (a) $\delta^{15}NH_4^+$ or (b) $\delta^{15}NO_3^-$ values in Gan River, Xiu River, Bang Lake, and mixed water (meeting area between Gan River and Xiu River).

Table 1. $\delta^{15}NH_4^+$ and $\delta^{15}NO_3^-$ in Bang Lake and its adjacent rivers.

Water level	$\delta^{15}NH_4^+$ (‰)			$\delta^{15}NO_3^-$ (‰)		
	Bang Lake	North branch of Gan River	Downstream of Xiu River	Bang Lake	North branch of Gan River	Downstream of Xiu River
Apr (lentic)	-3.08 (4.14)	7.94 (1.92)	8.98 (3.20)	-2.17 (0.73)	2.92 (0.60)	1.95 (0.53)
May (flowing)	6.88 (3.38)	8.01 (2.59)	7.15 (0.69)	2.13 (1.14)	1.75 (0.52)	2.22 (1.03)
Jun (flooding)	-18.56 (5.17)	-8.88 (1.79)	-11.82 (1.61)	1.37 (3.05)	2.10 (0.40)	0.49 (0.40)
Jul (flooding)	-11.03 (3.76)	-5.39 (1.64)	-14.50 (1.93)	4.29 (1.28)	-1.75 (0.82)	-1.42 (0.32)
Oct (lowering)	-2.38 (1.50)	-1.44 (1.50)	-0.73 (1.20)	-1.29 (4.12)	5.89 (0.48)	4.35 (0.52)

increased from between -19.1‰ and -11.2‰ in September to between -5.3‰ and -0.9‰ in October. During the lentic period, when the water level was lowest, $\delta^{15}\text{NH}_4^+$ ranged from -7.8‰ to 5.2‰ , showed only relatively small changes over several months, and was significantly different from those at other water levels. During the flowing period, the water level began to rise, and $\delta^{15}\text{NH}_4^+$ values were the heaviest, ranging from 1.1‰ to 12.4‰ . The trends in $\delta^{15}\text{NO}_3^-$ were distinctly different from the trends of $\delta^{15}\text{NH}_4^+$ at different water levels. The $\delta^{15}\text{NO}_3^-$ was heaviest during the July flooding period, when it ranged from 0.1‰ to 5.9‰ . During the flowing period, $\delta^{15}\text{NO}_3^-$ ranged from 0.3‰ to 4.5‰ . $\delta^{15}\text{NO}_3^-$ was most depleted in September when the water level was declining, ranging from -12.7‰ to -0.1‰ . During the lentic period, $\delta^{15}\text{NO}_3^-$ values ranged from -7.5‰ to -0.9‰ and were higher than those in September of the declining water level period, with no significant difference among the values for several months. The changes in the $\delta^{15}\text{NH}_4^+$ and $\delta^{15}\text{NO}_3^-$ values indicated different sources of NH_4^+ and NO_3^- and transformations between different forms of NH_4^+ and NO_3^- at different water levels.

Discussion

River–lake connectivity based on $\delta^{18}\text{O}$ and δD of H_2O or $\delta^{15}\text{N}$ of DIN at different water levels

Based on $\delta^{18}\text{O}$ and δD of H_2O or $\delta^{15}\text{N}$ of DIN at different water levels, we estimated river–lake connectivity. During the lentic period (Apr), δD and $\delta^{18}\text{O}$ values of the lake water were distinctly heavier than those of the rivers, and $\delta^{15}\text{NH}_4^+$ and $\delta^{15}\text{NO}_3^-$ values in the lake were obviously more negative than those in the adjacent rivers, showing no water exchange occurred between the

rivers and the lake during this time (river–lake separation). In the flowing period (May), $\delta^{18}\text{O}$ and δD of the lake were close to those of the adjacent rivers, and the difference of $\delta^{15}\text{NH}_4^+$ or $\delta^{15}\text{NO}_3^-$ between the rivers and the lake was less, indicating river water inputs into Bang Lake (river–lake connection). During the flooding period (Jun–Jul), when the water level was at its highest, $\delta^{15}\text{NH}_4^+$ or $\delta^{15}\text{NO}_3^-$ in the rivers and the lake were distinctly different, showing incomplete water exchange between the rivers and lake during this stage, even though they seemed to be combined as a single waterbody. During the lowering period (Sep–Oct), $\delta^{15}\text{NH}_4^+$ or $\delta^{15}\text{NO}_3^-$ between the rivers and the lake was more variable, suggesting less water exchange between the rivers and the lake after river–lake separation.

Sources and transformations of DIN at different water levels in Bang Lake

The N isotope ratio ($^{15}\text{N}/^{14}\text{N}$) is widely used as an indicator of different N sources in aquatic environments (Kendall 1998, Chang et al. 2009, Gu 2012, Yoshikawa et al. 2016) because different forms of N from various sources have diverse N isotopic compositions. For example, $\delta^{15}\text{N}$ of synthetic fertilizers is $\sim 0\text{‰}$ (Marion et al. 2005), soil organic $\delta^{15}\text{N}$ ranges from 2‰ to 5‰ , $\delta^{15}\text{N}$ of animal waste ranges from 8‰ to 20‰ (Xie et al. 2007), $\delta^{15}\text{NH}_4^+$ in rainwater ranges from -30‰ to 0‰ , and $\delta^{15}\text{NO}_3^-$ in rainwater ranges from -10‰ to 2‰ (Zhao et al. 2011). Variability was apparent among $\delta^{15}\text{N}$ values from various N sources in the river catchments in this study (Table 2). $\delta^{15}\text{NH}_4^+$ values in urban sewage, fish pond or pig farm effluent, agricultural fertilizers, and rainwater were $25.4 \pm 3.3\text{‰}$, $>12\text{‰}$, $\sim 0\text{‰}$, and $-13.5 \pm 5.9\text{‰}$, respectively. $\delta^{15}\text{NO}_3^-$ from the pig farm was the heaviest ($7.0 \pm 2.3\text{‰}$) while that

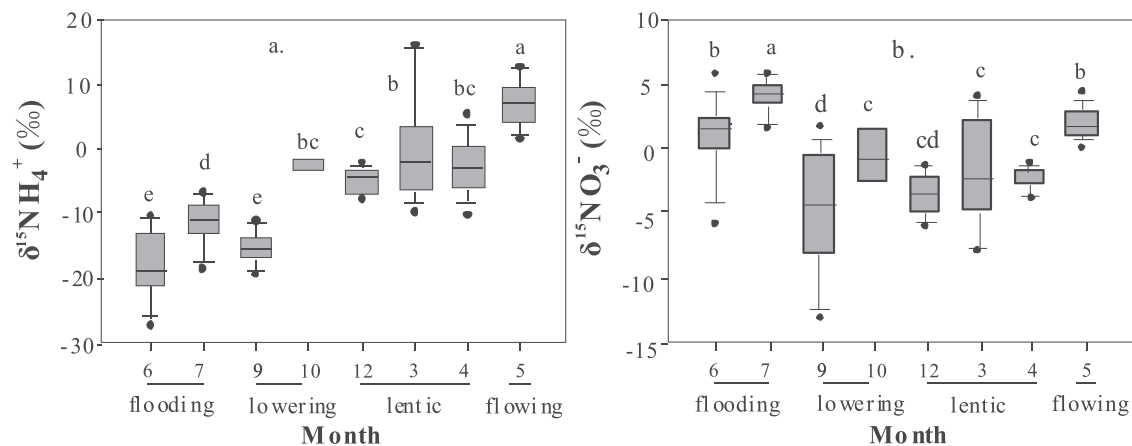


Figure 5. Monthly variation of (a) $\delta^{15}\text{NH}_4^+$ or (b) $\delta^{15}\text{NO}_3^-$ values with water levels in Bang Lake; the different letters of the boxplots indicate significant difference.

Table 2. Concentration and isotopic values of N origins along river catchments.

Sample types	NH ₄ -N (mg/L)	NO ₃ -N (mg/L)	δ ¹⁵ NH ₄ ⁺ (‰)	δ ¹⁵ NO ₃ ⁻ (‰)
Rainwater (Apr–Jun)	3.01 (0.98)	0.80 (0.53)	-13.47 (5.87)	-3.2 (1.23)
Fish-pond	1.29 (0.54)	0.46 (0.15)	13.47 (1.01)	-7.20 (2.22)
Urbanized-sewage outfalls	7.12 (1.83)	0.47 (0.17)	25.41 (3.34)	-2.91 (1.53)
Vegetable-field runoff	2.34 (0.48)	3.60 (0.62)	-2.02 (1.53)	2.45 (1.07)
Rice-field runoff	1.81 (0.31)	4.01 (0.82)	-3.01 (1.23)	2.09 (1.02)
Pig-farm outfalls	4.76 (2.56)	1.92 (1.04)	19.25 (3.22)	7.00 (2.30)

from the fish pond was the most depleted ($-7.2 \pm 2.2\text{‰}$); δ¹⁵NO₃⁻ of rainwater, urban sewage, and agricultural fertilizers ranged from -4‰ to 4‰ .

The physical and chemical parameters of waterbodies are important controls on N cycling in aquatic environments. For example, nitrification needs abundant DO, >0.5 mg/L, and a preferred temperature of 20–30 °C, whereas its reaction declines at temperatures <15 °C and stops below 5 °C. Denitrification most likely operates under alternating anaerobic and aerobiotic conditions (DO <0.5 mg/L) and high NO₃⁻ concentrations (Ogilvie et al. 1997, Xiao and Liu 2010). The water temperature ranged from 20 to 30 °C (except in winter when it was 12.0 ± 0.5 °C), DO was abundant (8–10 mg/L), pH changed little, and dissolved N concentrations were high at different water levels in Bang Lake (Table 3). By combining these parameters with δ¹⁵N values of pollution origins and adjacent rivers, we were able to identify the main sources of DIN and the dominant N transformation processes at different water levels in Bang Lake.

During the flooding period at the highest water level, comparison of the δ¹⁵NH₄⁺ or δ¹⁵NO₃⁻ of rainwater ($-13.5 \pm 5.9\text{‰}$ or $-3.2 \pm 1.2\text{‰}$, respectively) with δ¹⁵NH₄⁺ (-27.1‰ to about -7.0‰) of Bang Lake suggested that NH₄⁺ was mainly from rainwater. δ¹⁵NO₃⁻ values of the lake (mostly between 0.1‰ and 5.9‰) suggested that NO₃⁻ was mainly from agriculture and/or livestock effluent (Table 2) in retained river water. The high temperature, abundant DO, and wet conditions in summer meant that mineralization and nitrification occurred readily in the internal lake (Knoepp and Swank 2002, James 2010), and that the DIN concentrations (Table 3) and isotopic compositions (Fig. 5a–b)

were higher in July than in June. During this period, retained river waters mixed with rainwater in Bang Lake.

During the lowering period, the retained lake water flows rapidly into Poyang Lake, and therefore the δ¹⁵N values rapidly decreased in September and were much lower than those in June and July (Fig. 5a–b). The DIN isotopic compositions differed significantly between the lowering period and the flood period because the lake water was at its highest water level for 3 months, resulting in accelerated ammonification and nitrification under conditions of abundant DO and preferential temperature. The water level continued to decline to a depth of ~1 m, and the flow velocity also decreased. In these conditions, internal biogeochemical activities in the lake were still strong in autumn, and heavy isotopes were also gradually transformed (Kendall 1998), generally resulting in increased N isotopes in October.

During the lentic period, most of the δ¹⁵N values were negative and did not differ significantly from those during the lowering period (Oct), indicating that the lake's internal biogeochemical reactions were slow under winter and early spring low temperatures. The δ¹⁵NH₄⁺ values of some water samples were notably high (e.g., 15.6‰, 14.8‰, 4.4‰), possibly reflecting direct sources of anthropogenic pollution during early spring (Mar). The increasing N concentrations in the lake were likely the result of evaporation and the decreased water area in the shallow wetland (<0.8 m).

During the flowing period in early summer, rainfall is abundant and river water gradually flows over the natural dam into the lake. The higher δ¹⁵NH₄⁺ ($6.9 \pm 3.4\text{‰}$) and δ¹⁵NO₃⁻ ($8.0 \pm 2.6\text{‰}$) values indicated that agricultural pollution and human and/or livestock wastewater in runoff from land were the main sources of N during this period.

The influence of hydrological processes on N pollutant biogeochemistry in Bang Lake

Bang Lake, as a floodplain lake with restricted river–lake connections, experiences large fluctuations in water level. Hydrology and biogeochemistry of the lake differ from those of closed lakes where variations in precipitation cause only slight changes in water levels. For example,

Table 3. Monthly physical and chemical parameters in Bang Lake (average [SD]).

Monthly	June	July	September	October	December	March	April	May
Water level	flooding		lowering		lentic			flowing
Depth (m)	3.5 (0.7)	5.0 (1.2)	1.9 (0.4)	0.8 (0.2)	0.5	0.5	0.5	3.0 (0.8)
T (°C)	27.5 (0.8)	29.8 (0.5)	27.2 (0.6)	20.7 (2.3)	12.0 (0.5)	18.5 (0.5)	23.5 (0.5)	25.5 (0.7)
pH	8.5 (0.3)	7.8 (0.5)	7.8 (0.6)	7.8 (0.4)	7.1 (0.6)	7.5 (0.1)	7.5 (0.2)	7.6 (0.5)
DO (mg/L)	8.5 (1.7)	8.2 (0.5)	8.3 (0.5)	10.0 (1.05)	9.0 (0.9)	9.6 (0.7)	10.1 (0.4)	8.4 (0.4)
NH ₄ -N (mg/L)	0.24 (0.04)	0.30 (0.05)	0.29 (0.06)	0.32 (0.04)	0.69 (0.37)	0.77 (0.41)	0.52 (0.14)	0.48 (0.19)
NO ₃ -N (mg/L)	0.15 (0.04)	0.48 (0.23)	0.09 (0.02)	0.11 (0.11)	0.20 (0.10)	0.28 (0.14)	0.17 (0.09)	0.14 (0.05)

White et al. (2008), in their study of 16 natural lakes in the Laurentian Great Lakes Region where water level changed by <0.75 m, demonstrated that the biochemistry of the lakes was restricted by internal physical-chemical parameters, such as Ca^{2+} , conductivity, pH, and SO_4^{2-} .

Hydrological processes in floodplain systems can considerably influence nutrient dynamics (Baldwin and Mitchell 2000, James 2010, Welti et al. 2012). Bang Lake may be recognized as either an open, closed, or semi-closed system, depending on the water level. When closed, considerable differences occurred among the $\delta^{18}\text{O}$, δD , and $\delta^{15}\text{N}$ values in water in both the lake and the adjacent rivers, findings consistent with river-lake separation in the lentic period. The maximum nutrient concentrations and eutrophication were also observed in Bang Lake in this period due to the considerable amount of evaporation and/or the release from interstitial water resulting from wind wave actions in the shallow lake (Hu et al. 2005, Qin and Zhu 2006, Liang et al. 2015). By contrast, the $\delta^{18}\text{O}$, δD , and $\delta^{15}\text{N}$ values of the open lake water and adjacent rivers were similar, indicating discharge from the rivers into the lake (river-lake connection) during the flowing period; during this period, N pollutants mainly comprised agricultural, domestic, and/or livestock and poultry wastewater in runoff from the river catchments' surfaces. When semi-closed, water exchange with the lake water was incomplete, and the $\delta^{15}\text{N}$ values and solute N concentrations differed from those in adjacent rivers. The semi-closed state can be separated into 2 stages. During the first, the water level reaches a maximum during the 2–3-month flooding period, and the main sources of N are rainfall retained water from inflowing rivers and strong biogeochemical reactions in the internal lake (Rodgers et al. 2005). During the second stage, the water level declines and lake water flows out rapidly, causing gradual separation of the river from the lake; internal biogeochemical reactions (ammonification and nitrification) are crucial for N transformations in this stage.

Conclusions

The hydrological processes at the different water levels were described by the stable isotopes (δD , $\delta^{18}\text{O}$, $\delta^{15}\text{N}$), and the source and transformation processes of DIN were traced by $\delta^{15}\text{NH}_4^+$ or $\delta^{15}\text{NO}_3^-$ in Bang Lake. During the rainy season, the main sources of DIN are agriculture and/or domestic and livestock wastewater in surface runoff from the catchments' surfaces. During the flooding stage, river water and plentiful rainwater remain in the lake for 3 months when the flow is slow; the main

source of NH_4^+ is rainwater while NO_3^- is mainly from agriculture and/or livestock wastewater in retained river water; and mineralization and nitrification occur in the internal lake. When lower rainfall causes the water levels to fall, the river is separated from the lake, and mineralization and nitrification are the most important N transformations in the internal lake. As the water level continues to fall, Bang Lake becomes a closed wetland; during this stage, the N concentrations reach a maximum, but biogeochemical reactions are weak. This study will help in managing nutrient input and in controlling the pollution in the floodplain lake. Moreover, field studies on the transport and transformation of inorganic nitrogen and the effects of hydrological variations on the nitrogen cycling in floodplain lakes would be greatly useful for future study.

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