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Temporal and spatial distributions of dissolved organic carbon and nitrogen in two small lakes on the Southwestern China Plateau

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Abstract Temporal and spatial distributions of dissolved organic carbon (DOC), dissolved organic nitrogen (DON), chlorophyll-*a* and inorganic nitrogen were investigated in two small mountainous lakes (Lake Hongfeng and Baihua), on the Southwestern China Plateau, based on almost 2 years' field observation. DOC concentrations ranged from 163 μM to 248 μM in Lake Hongfeng and from 143 μM to 308 μM in Lake Baihua, respectively, during the study period. DON concentrations ranged from 7 μM to 26 μM in Lake Hongfeng and from 14 μM to 47 μM in Lake Baihua. DOC showed vertical heterogeneity with higher concentrations in the epilimnion than in the hypolimnion during the stratification period. The DON concentration profiles appeared to be more variable than the DOC profiles. Apparent DON maxima occurred in the upper layer of water. In Lake Hongfeng, DOC concentration in the surface water was highest at the end of spring and early summer. DON concentration was 2–5 μM higher in May 2003 and in June 2004 than in adjacent months. DOC and chlorophyll-*a* concentrations were significantly correlated ($r = 0.79$, $P < 0.05$). The period of highest concentrations of DOC in Lake Hongfeng was also the season of concentrated rainfall. Algae activity and allochthonous input might result in an increase of DOC and DON concentrations together. In Lake Baihua, the maximum concentrations of DOC and DON in the surface water occurred simultaneously in May 2003 and February 2004. DOC concentrations were significantly correlated with DON ($r = 0.90$, $P < 0.01$), indicating the common sources. Allochthonous input, biological processes, stratification and mixing were the most important factors controlling the distributions and cycling of dissolved organic matter (DOM) and inorganic nitrogen in these two lakes. Inference from the corresponding vertical distributions of DOM and inorganic nitrogen indicated that DOM played potential roles in the internal loading of nitrogen and metabolism in the water body in these small lakes. The carbon/nitrogen (C/N) ratio showed a potential significance for tracing the source and biogeochemical processes of DOM in the lakes. These results are of significance in the further understanding of biogeochemical cycling and environmental effects of DOM and nitrogen in lake ecosystems.

Keywords Dissolved organic nitrogen · Dissolved organic matter · Lake · Southwestern China Plateau · Nitrogen

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Introduction

Naturally occurring dissolved organic matter (DOM) is one of the important constituents in natural water ecosystems. Previous studies have demonstrated that DOM shows strong reaction activity and significant eco-environmental effects in natural environments (e.g., Tanoue and Midorikawa 1995; Barber et al. 2001; Wu and Tanoue 2001; Doig and Liber 2006). DOM may control the transport, toxicity and fate of trace metals and organic contaminants in aquatic environments (e.g., Tanoue and Midorikawa

1995; Ohlenbusch et al. 2000; Wu and Tanoue 2001; Doig and Liber 2006) and influence acid–alkali balance, dissolved oxygen and biogeochemical cycling of nutrients (e.g., Ritchie and Perdue 2003; Kim et al. 2006). DOM can also serve as an energy source for aquatic food webs and as an influence on the biological activity of phytoplankton and bacteria (Thomas 1997). In addition, DOM is the precursor of trihalomethanes and other disinfection by-products that occur during the chlorination of drinking water (Palmstrom et al. 1988).

To better understand the role and biogeochemical cycling of DOM in aquatic systems, it is necessary to study the temporal and spatial distributions of DOM and corresponding influences. DOM in natural waters is a heterogeneous mixture of organic compounds. Therefore, dissolved organic carbon (DOC), dissolved organic nitrogen (DON), and carbon/nitrogen (C/N) ratios of DOM were often investigated when the cycling and dynamics of DOM were being studied (e.g., Hopkinson and Cifuentes 1993; Ogawa et al. 1999; Lobbes et al. 2000; Mash et al. 2004; Kim et al. 2006). DOM can be estimated from the measurement of carbon, because carbon is the major element in DOM. DOC concentrations have been widely determined in many lakes in the world and are obviously affected by lake and watershed characteristics (e.g., Hama and Handa 1983; Kim et al. 2000; Sugiyama et al. 2004). DOM in lake water has two sources: allochthonous and autochthonous. Allochthonous DOM mainly originates from the decomposition of plant debris and the humification of soil organic matter in natural conditions, but the influence of human activity needs to be considered if the lakes are close to cities. Several studies have been carried out to investigate the distribution and biogeochemical cycling of DOM, focusing on DOC (Hama and Handa 1983; Fukushima et al. 1996; Parks and Barker 1997; Kim et al. 2000; Sugiyama et al. 2004; Brooks et al. 2005), but relatively few studies have been reported on DON in lakes (Mash et al. 2004; Kim et al. 2006). At the same time, the importance of the C/N ratio in DOM has been emphasized in oceans (Hopkinson and Cifuentes 1993; Ogawa et al. 1999), but there have rarely been few detailed profiles of the C/N ratio in DOM in lakes.

The mineralization of organic matter in lakes has been thought to be an important source of internal loading of inorganic nutrients, such as nitrogen and phosphate. Traditionally, sedimentation, and the subsequent mineralization of particulate organic matter (POM), have been considered to account primarily for this process (Hutchinson 1938), but a recent study has disclosed that dissolved organic matter might play an important role in the hypolimnetic mineralization of carbon and nitrogen in a large lake (Kim et al. 2006). More studies about this topic are needed to verify this conclusion.

Reservoirs are now very common in many regions of the world, including China, and they represent an important and special type of lake nowadays. Many studies have been carried out in reservoirs (e.g., Parks and Barker 1997; Kim et al. 2000; Wu et al. 2001; Mash et al. 2004; Nakashima et al. 2007). In our study, almost 2 years' field observation was conducted in a two-reservoir system on the Southwestern China Plateau. The objectives of this study were to (a) investigate the temporal and spatial distributions of both DOC and DON, (b) study their influencing factors, and (c) discuss the relationship between DOM and inorganic nitrogen in these two lakes. This study could be helpful in the understanding of the biogeochemical cycling and environmental effects of DOM in lacustrine environments.

Sampling and analyses

Lake Hongfeng (HF) and Baihua (BH), located in the suburbs of Guiyang City, on the Southwestern China Plateau, are a two-reservoir system on the upper Maotiao River. The watershed is mainly underlain by carbonate rocks. Forest accounts for approximately 8.1% of the whole watershed area. Annual precipitation in the region of these two lakes is 1,200 mm year^{-1}. More than 55% of annual precipitation occurs from May to August (Zhang 1999). Lake Hongfeng has four main inflow rivers, which contribute the majority of the inflow water. The water discharged from the dam of Lake Hongfeng flows into Lake Baihua through a connected river and forms the major water source of Lake Baihua (Zhang 1999). The characterization of these two lakes and catchments has been reported in many previous studies (e.g., Zhang 1999; Wu et al. 2001; Xiao et al. 2002). These two reservoirs serve to control floods and provide water for electrical power generation, industrial and agricultural water supply and recreation. Lake Hongfeng is also the drinking water source for Guiyang City. The water level of these two lakes fluctuates in a significant range, influenced by water control of people and climate. The general hydrological characteristics of these two lakes are shown in Table 1.

Table 1 Hydrological characteristics of Lake Hongfeng and Lake Baihua

Hydrological characteristics	Hongfeng	Baihua
Watershed area (km^2)	1,596	1,895
Altitude (m)	1,227	1,188
Surface area at full capacity (km^2)	57.2	14.5
Length (km)	16	18
Average depth (m)	10.5	12.5
Hydraulic residence time (days)	119	37

Water samples were collected at 2 m or 3 m intervals with a Niskin water sampler from January 2003 to August 2004, every 2 months. We chose three sampling sites in this two-reservoir system: two sites, respectively, along the major axis of Lake Hongfeng and Lake Baihua (HF-S, BH-1) and one site on the connected river (Huaqiao) (Fig. 1). The temperature was instantly measured in the field. Water samples were immediately filtered through pre-combusted (450°C, 5 h) Whatman (GF/F) glass-fiber filters. The filtrate, used for DOC measurement, was collected directly into an acid-cleaned, pre-combusted (550°C, 5 h) brown glass bottle. These samples were acidified and kept at 4°C until analysis (principally within 100 h). For the measurement of other dissolved constituents [total dissolved nitrogen (TDN), nitrate (NO_3^-), ammonium ion (NH_4^+) and nitrite (NO_2^-)], the filtrate was contained in acid-washed polyethylene bottles and stored at 4°C. A chlorophyll-*a* sample was obtained by filtration of 1 l aliquots through Whatman (GF/F) glass-fiber filters. These samples were stored frozen until analysis.

NO_3^- concentration was measured by a chromatographic method according to Butt et al. (2001), with some modification, and the detection limit was 0.7 μM. NH_4^+ was determined by the indophenol blue method (Koroleff 1983), with the detection limit of 0.5 μM, and NO_2^-N concentrations were determined by spectrophotometric analysis [The State Environmental Protection Agency (SEPA) of China 2002], with the detection limit of 0.2 μM. Potassium persulfate oxidation ultraviolet (UV) spectrophotometry was employed to determine TDN concentrations, with the detection limit of 3.5 μM, and a 143 μM ammonium chloride (NH_4Cl) solution was used to monitor the recovery in the analytical process (SEPA 2002). DON concentration was calculated as differences between TDN and total inorganic nitrogen (NO_3^-, NO_2^- and NH_4^+). DOC was measured by high-temperature catalytic oxidation, with a High TOC II analyzer (Elementar, Germany). Potassium hydrogen phthalate was used as standard. The relative standard deviation (RSD) of the replicate measurements ($n = 5$) of DOC was less than 2%, and the detection limit was 16 μM. Chlorophyll-*a* concentration was measured by acetone-extraction spectrophotometry (Jin and Tu 1990). Dissolved oxygen (DO) concentration was determined by the classical iodometric method.

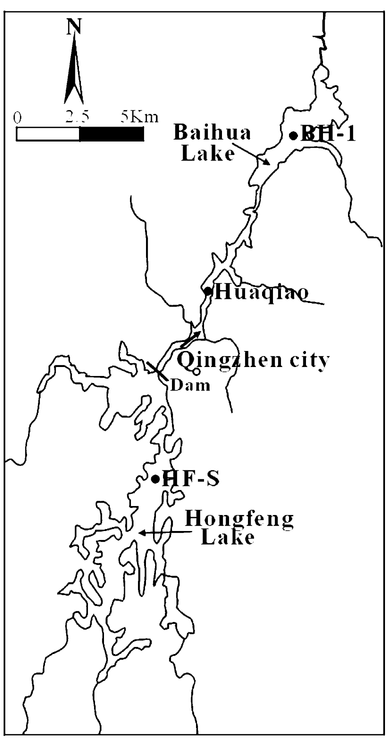


Fig. 1 Map of sampling sites (*solid circles*) in Lake Hongfeng and Baihua, and their connected river. The *arrow* in the connected river shows the direction of water flow

Results and discussion

Limnological characteristics of Lakes Hongfeng and Baihua

The pH value of surface water raged from 7.9 to 8.6 and 7.8 to 8.4 in Lake Hongfeng and Lake Baihua, respectively, in 2003. These two lakes exhibited stratification during the summer season. The spatio-temporal distribution patterns of DO and temperature in the water column were almost similar in these two lakes (Fig. 2). Distinct stratification was evident from May, and the hypolimnion was anoxic. The stratification phenomenon did not completely disappear until November. The thermocline occurred at the depth of 4 m in July at Lake Baihua. It appeared that both small lakes showed strong seasonal anoxia in the hypolimnion during the summer season.

Figure 3 shows the vertical and temporal distributions of NO_3^-, NO_2^- and NH_4^+ in Lake Hongfeng and Lake Baihua. NO_2^- and NH_4^+ were minor forms of dissolved inorganic nitrogen, while NO_3^- was the predominant form, accounting for approximately 52–98% (84% on average) of dissolved inorganic nitrogen in the surface water of these two lakes. The maxima of NO_2^- and NH_4^+ occurred in the bottom water during the stratification period, suggesting the regeneration of inorganic nitrogen. Maximum NO_3^- occurred at 8–16-m depth between March and July in Lake Hongfeng and at 0–12-m depth between May and September in Lake Baihua.

Vertical and temporal distribution of DOC and DON

During the study period, DOC concentrations ranged from 163 μM to 248 μM in Lake Hongfeng and from 143 μM to

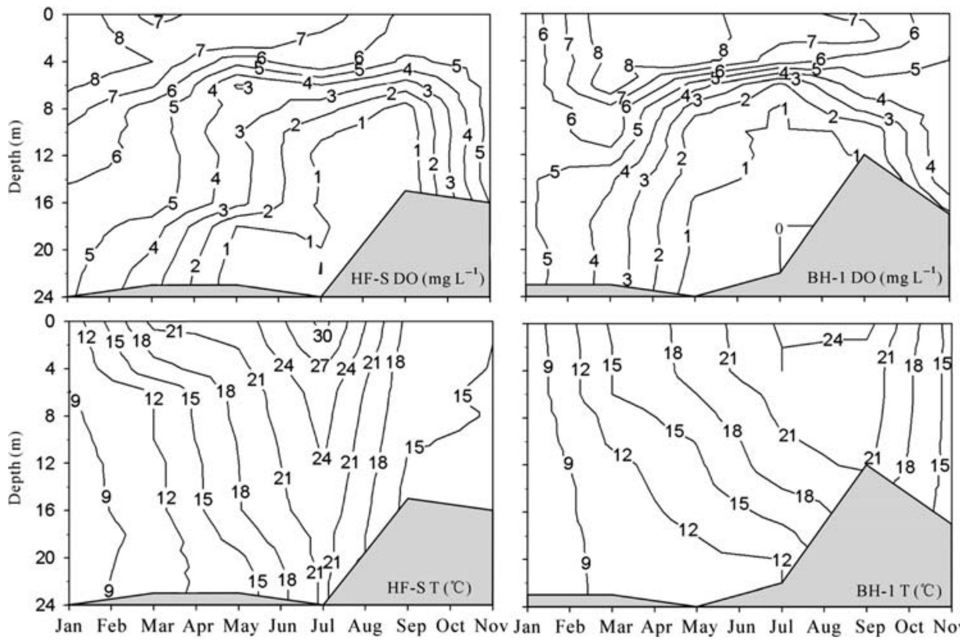


Fig. 2 Seasonal changes of vertical distributions of DO and temperature in Lakes Hongfeng and Baihua in 2003. In the text, the surface is a water depth of 0 m. The scales for the *X* and *Y* axes are the same for all contours. The *values* of these contours indicate the corresponding values of the appointed parameters. Note that the water depth during every month was not same

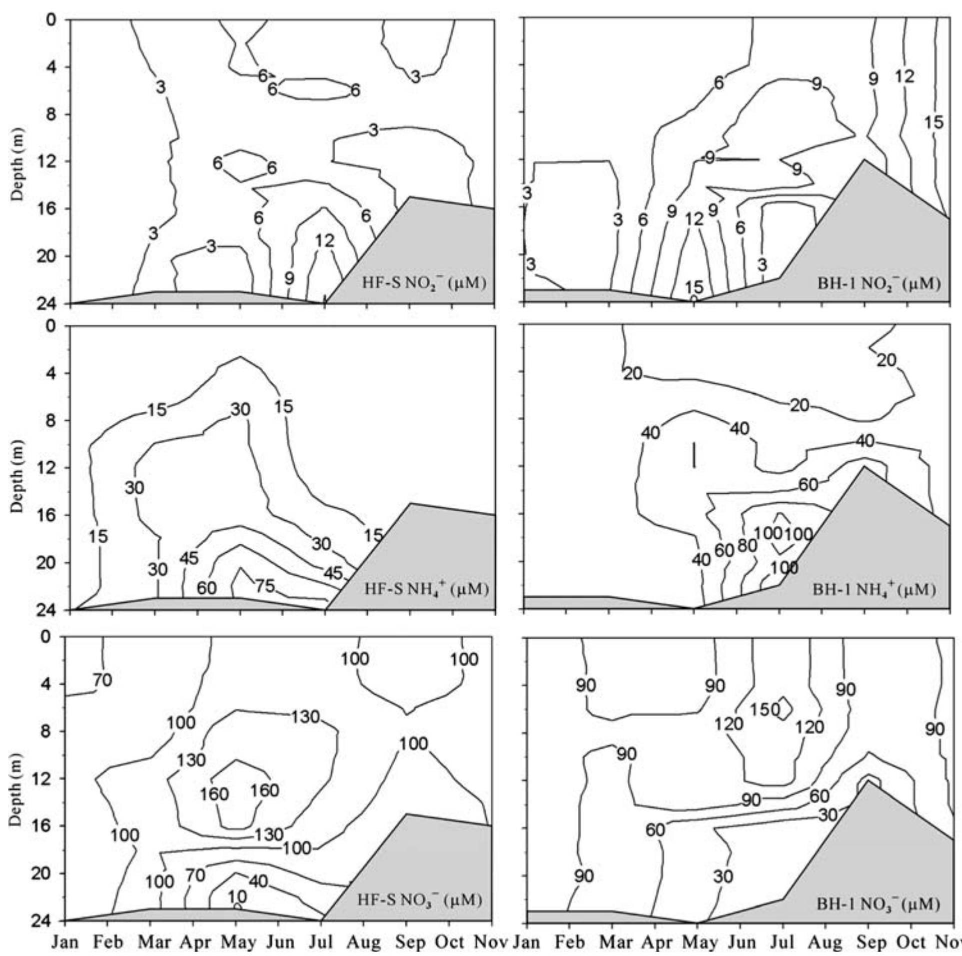


Fig. 3 Seasonal changes of vertical distributions of NO_2^-, NH_4^+ and NO_3^- in Lakes Hongfeng and Baihua in 2003. In the text, the surface is a water depth of 0 m. The scales of the *X* and *Y* axes are the same for all contours. The *values* of these contours indicate the corresponding values of the appointed parameters. Note that the water depth during every month was not same

308 μM in Lake Baihua. DOC concentrations in these two lakes were lower than those of some boreal lakes (more than 300 μM; Schindler et al. 1997), but were higher than those of some large lakes, such as Lake Biwa (mean value approximately 110 μM in the epilimnion; Kim et al. 2006) and Lake Baikal (90–110 μM; Yoshioka et al. 2002). DOC concentrations in the Japanese eutrophic lakes Kasumigaura and Suwa were 100–300 μM (Hama and Handa 1983; Fukushima et al. 1996), which were comparable to those of Lake Hongfeng and Lake Baihua. Vertical and temporal distributions of DOC and DON are apparently observed in Lake Hongfeng and Lake Baihua (Fig. 4). In both lakes the concentrations of DOC showed vertical heterogeneity, with higher concentrations in the epilimnion than in the hypolimnion during the summer stratification period. Variations in concentrations of DOC over depth were less pronounced during the overturn period (November and January) than during the stratification period. These results are in agreement with those of some previous reports concerning DOC distribution in lakes (Wetzel 1972; Sugiyama et al. 2004; Mostofa et al. 2005; Kim et al. 2006), although the vertical heterogeneity of DOC concentrations was not significant in shallow Lake Suwa, Japan (Hama and Handa 1983). An increase in DOC concentrations was sometimes found near the lake bottom during the summer season (2004 in Lake Hongfeng and 2003 in Lake Baihua), and this might be attributed to the degradation of sinking organic particles, as noted in Lake Soyang, Korea, by Kim et al. (2000).

DON concentrations ranged from 7 μM to 26 μM in Lake Hongfeng and from 14 μM to 47 μM in Lake Baihua. These concentrations were higher than those of coastal seas (mean value approximately 10 μM; Bronk 2002) and Lake Biwa (mean value approximately 8.35 μM in the epilimnion; Kim et al. 2006). DON accounted for 5–22% of TDN in Lake Hongfeng and 7–33% in Lake Baihua. Although DON frequently forms the largest part of TDN in many natural waters (average about 60–69%; Bronk 2002), it sometimes accounts for a small part of the TDN, such as 12% in the Southern Ocean (Ogawa et al. 1999), which is similar to our results. In both lakes the vertical gradient pattern of DON concentrations from surface to deep water appeared to be more variable than the DOC profiles. However, apparent DON maxima occurred in the upper 4–6-m layer of Lake Hongfeng and in the surface water of Lake Baihua. DON concentrations in the hypolimnion were generally lower, especially during the stratification period. Similar vertical distributions of DON were reported in ocean water columns, where the highest DON concentration was observed in the surface water (Charles et al. 1997; Ogawa et al. 1999). In Lake Biwa, the maxima of DON occurred in the upper 15–20-m layer between June and October (Kim et al. 2006), which was similar to our results. As for DOC, increased DON concentrations were found near the lake bottom during the summer season in Lake Hongfeng.

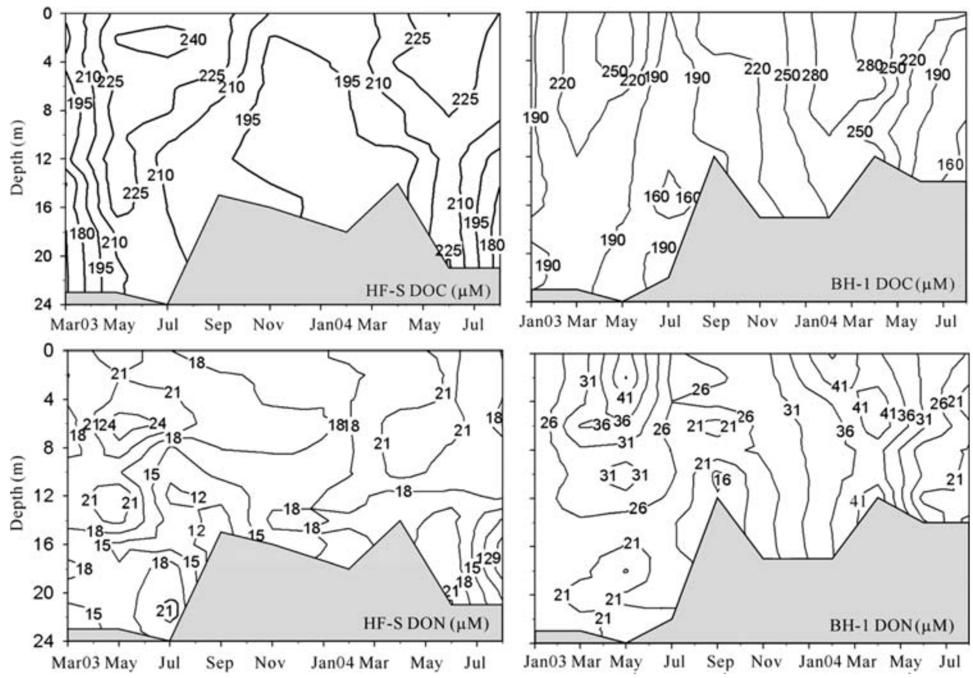
Although the vertical distributions of DOC and DON were similar between Lake Hongfeng and Lake Baihua, the temporal distributions of DOC and DON in surface water were different.

In Lake Hongfeng, DOC concentrations in the surface water were highest at the end of spring and early summer, with maxima occurring in May 2003 (maxima lasted until September) and in June 2004 (Fig. 5a). This trend is similar to that observed in Lake Biwa and Lake Kasumigaura, in Japan, with DOC concentrations in the epilimnion increasing from spring to fall (Imai et al. 2001; Kim et al. 2006). The seasonal variation of DON in the surface water of Lake Hongfeng was not so obvious (Fig. 5a). DON concentrations were 2–5 μM higher in May 2003 and in June 2004 than in adjacent months.

In Lake Baihua, the maximum concentrations of DOC and DON in the surface water occurred simultaneously in May 2003 and February 2004 (Fig. 5b). DON concentrations were 10–17 μM higher than in adjacent months. DOC concentrations were significantly correlated with DON ($r = 0.90$, $P < 0.01$), indicating the common sources. Further discussion will be presented later.

It has been reported that DOC can be released during phytosynthesis by living algae and during decomposition of dead algae (Mague et al. 1980; Norrman et al. 1995). Chlorophyll-*a* can be used to represent algae biomass. Because the Secchi disk depths (used for the in situ measurement of clarity of surface waters) were always less than 4 m (Zhang 1999) and the maximum concentration of chlorophyll-*a* usually occurred at 2 m or 3 m depth, the average chlorophyll-*a* concentration in the 0–3 m water layer was used to compare DOC and DON. In Lake Hongfeng, the maximum concentrations of chlorophyll-*a* in the surface layer occurred from May to July 2003 and August 2004 (Fig. 5a), indicating an increase in algal biomass. DOC, DON and chlorophyll-*a* concentrations in the surface water varied almost simultaneously (Fig. 5a). DOC and chlorophyll-*a* concentrations in the surface water of Lake Hongfeng were significantly correlated ($r = 0.79$, $P < 0.05$). This suggested that algal biological activities might contribute to DOC and DON in the surface water. On the other hand, the contribution of the allochthonous input to DOC and DON cannot be ignored in lacustrine environments. Large increases of DOC have been detected in rivers and streams with flow in the monsoon season (Hinton et al. 1997; Kim et al. 2000). In Lake Hongfeng's watershed, the rainfall was concentrated from April to July 2003 (Fig. 6), and the highest concentrations of DOC appeared in Lake Hongfeng during this period. Therefore, the terrestrial DOM brought through rivers into lakes

Fig. 4 Seasonal changes of vertical distributions of DOC and DON concentrations in Lakes Hongfeng and Baihua from January 2003 to August 2004. In the text, the surface is a water depth of 0 m. The scales for the *Y*-axes are the same for all contours. The *values* of these contours indicate the corresponding values of the appointed parameters. Note that the water depth during every month was not same



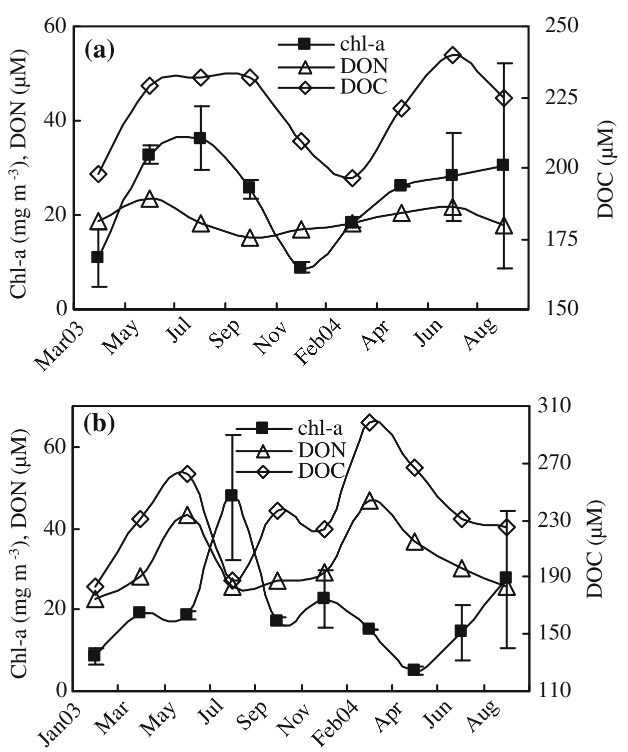


Fig. 5 Temporal variations of surface DOC, DON and average chlorophyll-*a* (*Chl-a*) concentrations in the surface layer (0–3 m) water in **a** Lake Hongfeng and **b** Lake Baihua from January 2003 to August 2004. See the text for the reason why we chose 0–3 m average value to represent the surface chlorophyll-*a* concentrations. Errors are standard deviations for individual depths

during heavy rainfall might also result in an increase in DOC and DON concentrations in the surface water. It has been reported that the C/N ratio of organic matter can be used as a tracer of its source. Terrestrial organic matter usually has a higher C/N ratio than does autochthonous organic matter (Ertel et al. 1986; Cowie and Hedges 1994). Mash et al. (2004) found that autochthonous DOM had a lower C/N ratio than allochthonous DOM in Arizona reservoirs, in the United States of America (USA. The low C/N ratio of DOM in May 2003 in Lake Hongfeng might indicate a relatively large contribution of autochthonous sources to DOM, while a relatively higher C/N ratio in June 2004 (Fig. 7) might imply comparatively large contribution of allochthonous sources, when the maximum DOC concentrations are observed. Therefore, in Lake Hongfeng, algal activity and allochthonous input might result in an increase of DOC and DON concentrations together, but further investigations are needed to determine the relative contributions of each factor in these lakes.

In Lake Baihua, the seasonal variation of chlorophyll-*a* in surface water was quite different from that of DOC and DON. Chlorophyll-*a* concentrations were considerably lower when DOC and DON maxima appeared (Fig. 5b). Except in summer, the DOC concentrations in the surface water of Lake Baihua (from 183 μM to 299 μM) were higher than those in Lake Hongfeng (from 181 μM to 240 μM), while DOC concentrations in the surface water at the Huaqiao site (from 209 μM to 568 μM), which was located on the river connecting these two lakes, were much higher than those of these two lakes. The domestic sewage of Qingzhen City, which was characterized by high DOC and DON concentrations and low C/N ratio (Li et al., unpublished data), flowed into the river connecting these two lakes, before the Huaqiao site, through a river. It might

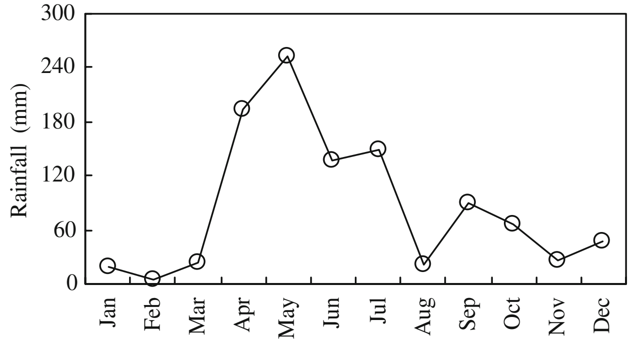


Fig. 6 Rainfall for each month in the lake region during 2003 (data are obtained from the Qingzhen city hydrometric station)

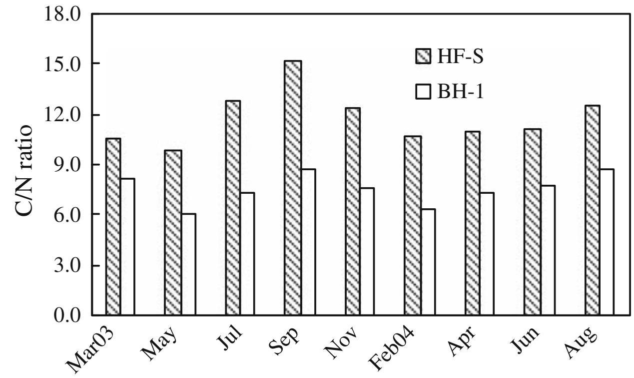


Fig. 7 C/N ratios of DOM in the surface water of Lakes Hongfeng and Baihua from March 2003 to August 2004

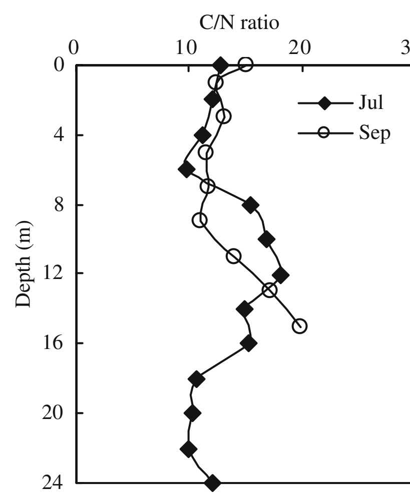


Fig. 8 Vertical profiles of C/N ratios of DOM in July and September 2003 at the HF-S site. In the text, the surface is a water depth of 0 m

be assumed that the domestic sewage was the major allochthonous source of DOM to Lake Baihua and resulted in increased DOC and DON concentrations. The C/N ratio of DOM in the surface water of Lake Baihua was much lower than that of Lake Hongfeng, and C/N ratios were much lower than in other months when the maximum DOC and DON concentrations were observed (May 2003 and February 2004), reflecting the influence by allochthonous input of DOM, viz. domestic sewage (Fig. 7). In summer, the water from Lake Hongfeng was discharged from the dam more frequently, so that the allochthonous DOM input to Lake Baihua was diluted.

The relationship between DOM and inorganic nitrogen

Some previous studies have demonstrated that DOM may play important roles in hypolimnetic metabolism and internal loading of nitrogen and carbon in stratified water bodies (Houser et al. 2003; Kim et al. 2006). Although we could not make rigorous calculations about the contribution of DOM to the internal loading of nitrogen in Lakes Hongfeng and Baihua, our study still showed some relationships between DOM and inorganic nitrogen from their spatio-temporal distributions in Lake Hongfeng.

In response to both biological activity and hydrographical properties, DOC, DON and inorganic nitrogen gradients developed in the water column during the stratification period (e.g., July and September 2003). NO_3^- concentration increased from the surface to 12 m depth, while DOC and DON concentrations generally decreased in the same water column in July (Figs. 3, 4). The relatively low NO_3^- concentration in surface water might have resulted from the use of NO_3^- by algae in the euphotic zone, while the aerobic mineralization of DOM (together with POM) might have contributed to an increase of NO_3^- in the 4–12-m layer, the same mechanism as brought forward by Lehmann et al. (2004) in Lake Lugano. Meanwhile, from 6 m to 12 m the C/N ratio of DOM increased with depth (Fig. 8), which suggested the mineralization of DOM, since DOM degradation caused an increase in the C/N ratio (Herczeg 1988; Charles et al. 1997). NO_3^- concentration gradually decreased over depths below 12 m, while NH_4^+ accumulated in the same water column in July (Fig. 3). The decrease in NO_3^- may have been caused by the intense microbial mineralization of organic matter (POM and DOM) in this anoxic layer (below 12 m) using NO_3^- as electron acceptors, and the anoxic mineralization of organic matter led to the accumulation of ammonium ions. It seemed that 12 m was the redoxcline in the July water column. In September, organic matter mineralization still existed and led to maximum NO_3^- concentration in the intermediate waters (approximately 7 m depth; Fig. 3). Below 9 m the C/N ratio of DOM increased with depth (Fig. 8), which might have been caused by the mineralization of DOM or the use of DON by heterotrophic microbes. During the overturn period of the water body in November, all the gradients of

DOM and inorganic nitrogen collapsed (Figs. 3, 4). This distribution pattern of DOM and inorganic nitrogen revealed that the stratification of lake waters greatly influences the transport and transformation of DOM and inorganic nitrogen in lakes, and DOM plays potential roles in the internal loading of nitrogen and metabolism of the water body in small lakes.

These results exhibited some relationships among spatio-temporal distributions of DOC, DON, chlorophyll-*a* and inorganic nitrogen, as well as significant influences by lake hydrochemistry. Allochthonous input, biological processes, stratification and vertical mixing were the most important factors controlling the distributions and cycling of DOM and inorganic nitrogen in these two small lakes. Terrestrial input and biological processes probably significantly affected the temporal distribution of DOC and DON, while biological processes of DOM and stratification mainly influenced the spatial distribution of DOC, DON and inorganic nitrogen. C/N ratio showed a potential significance for tracing the source and biogeochemical processes of DOM in lakes.

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