



Spatial and temporal distribution characteristics of different forms of inorganic nitrogen in three types of rivers around Lake Taihu, China

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Abstract

In order to control nitrogen (N) pollution of Lake Taihu, China, we studied the spatial and temporal distribution characteristics of inorganic N in inflowing rivers polluted by industry, agriculture, and domestic sewage during low, moderate, and high flow periods. The results showed that dissolved total nitrogen (DTN) was the main fraction of total nitrogen (TN) input from these rivers. Inflowing rivers had distinct impacts on TN, DTN, ammonium N (NH_4^+), and nitrate N (NO_3^-) concentrations of Lake Taihu during the low flow period. Particulate nitrogen (PN) had an impact on Lake Taihu during the three flow periods and all the three types of rivers would increase PN concentration in the lake. Rivers polluted by agriculture had the greatest impact on Lake Taihu's TN, DTN, NO_3^- , and dissolved inorganic N (DIN) concentrations, while rivers polluted by industry had the greatest impact on NH_4^+ concentration. Therefore, agriculture and industry should be key targets for nutrient reductions. The in-lake N concentrations were higher than those of inflowing rivers during moderate and high flow periods.

Keywords Inorganic nitrogen · Inflowing rivers · Industry · Agriculture · Domestic sewage · Eutrophication · Lake Taihu

Introduction

The Lake Taihu Basin is one of the most developed regions of China and reflects the rapid industrialization, urbanization, and agricultural intensification that play a vital role in China's social and economic development (Qin et al. 2007; Zhang et al. 2016). Lake Taihu is an important source of drinking water, and it serves flood control, tourism and recreation, shipping, and aquaculture activities (Qin et al. 2007). Since the 1980s, rapid economic development of the Lake Taihu Basin has

resulted in large amounts of industrial, agricultural, and urban pollutants discharged into the rivers discharging to the lake (Qin et al. 2007). With the deterioration of water quality, cyanobacterial harmful algal blooms (CHABs) have intensified, resulting in a significant decrease in ecological services provided by the lake (Zhang et al. 2016). In late May 2007, massive cyanobacterial blooms accumulated near the drinking water source of Wuxi City, China, led to the cutting off of drinking water supplies to approximately 2 million local residents for 1 week (Qin et al. 2010).

CHABs are the most conspicuous sign of eutrophication of freshwater ecosystems. Despite widespread efforts to reduce nutrient loading in many regions, eutrophication remains a growing problem (Schindler et al. 2008; Ibelings et al. 2016). Hydrologic and climatic factors play additional roles in the long-term changes of phytoplankton biomass and cyanobacterial bloom intensity, by modulating nutrient effects on phytoplankton growth and bloom potentials (Paerl et al. 2016a; Paerl 2017; Zhu et al. 2018a). Consequently, stricter nutrient reductions in the catchment, including nitrogen and phosphorus, are needed to ultimately control the cyanobacterial blooms in Lake Taihu (Paerl et al. 2011).

In many situations, reducing both nitrogen (N) and phosphorus (P) from external inputs provides the best opportunity to reduce phytoplankton biomass and bloom potentials (Paerl et al. 2016b). Increasing nutrient inputs worldwide has

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coincided with an unprecedented increase in CHABs, especially toxic, non-N₂-fixing cyanobacteria (Paerl et al. 2016b). Many lakes are increasingly impacted by N due in large part to the traditional focus on controlling only P inputs, while ignoring control of N inputs from watersheds, including N in urban runoff and increased use of fertilizers in agriculture (Paerl et al. 2016b). The average concentration of total nitrogen (TN) in Lake Taihu is approximately 2.62 mg/L, and the average concentration of total phosphorus (TP) is approximately 0.086 mg/L (Dai et al. 2016). Thus, TN is a highly significant source of nutrient pollution in Lake Taihu (Zhen and Zhu 2016).

Inflowing rivers are a dominant source of nutrient pollution in Lake Taihu. They provide the main water resources for domestic, industrial, and irrigation purposes; however, they are also easily polluted because of their critical roles in transporting municipal, industrial pollution, and runoff from agricultural lands (Xu et al. 2015; Zhou et al. 2016). Nitrogen from inflowing rivers in the northern region of Lake Taihu account for over 70% of the total N load in rivers around the lake (Li et al. 2016). Because of their impacts on the ecology, human health, and economic welfare, it is essential to prevent and control further water quality deterioration of the rivers.

To control N pollution of Lake Taihu, it is important to know the spatial and temporal distribution characteristics of N species in its inflowing rivers. In this study, inorganic N species in river water polluted by industrial, agricultural, and domestic sewage were measured during low, moderate, and high flow periods. Our findings provide background information relevant for implementing N input reductions needed to improve the Lake's water quality.

Material and methods

Sampling times and sites

The rainy season in the Lake Taihu Basin begins in April, with water levels usually reaching their peak in late July, while the lowest water levels occur between February and March every year, according to long-term data from the Lake Taihu Basin Authority of Ministry of Water Resources. The monthly rainfall in Lake Taihu Basin was 145 mm in February 2009, which was more than twice as high as that during the same period of previous years, and close to the maximum rainfall during the same period since 1956. The monthly rainfall in Lake Taihu Basin was 53 mm in May 2009, which was 54.0% less than that during the same period of previous years. The monthly rainfall in Lake Taihu Basin was 200.8 mm, which was 46% more than that during the same period of previous years. The water level of Lake Taihu in 2009 was 3.08 m at the beginning of the year and 3.25 m at the end of the year. The highest water level was 4.23 m in August and the lowest water level was 2.87 m in February. The annual average water level was 3.31 m, which

was close to the water level in May. Therefore, we chose February, May, and August 2009 represent low, moderate, and high flow periods. The specific sampling times were from 21 to 25 February, from 24 to 26 May, and from 26 to 28 August of 2009. Three types of rivers discharging to Lake Taihu were selected as study objects. The 1st (Zhihu Port), 2nd (Hengtang River), and 3rd (Wangyu River) were polluted by industry and they are located on the north side of Lake Taihu. The 4th (Xujiang River), 5th (Zhi Port), and 6th (Cao Port) were polluted by domestic sewage and they are located on the east side of Lake Taihu. The 7th (Zhongtang River), 8th (Changxing Port), and 9th (Jiapu Port) were heavily influenced by agricultural pollution and they are located on the southwest side of Lake Taihu. Three sampling sites were set up along the upper, middle, and lower reaches of each river, amounting to a total of 27 sample sites examined in our study (Fig. 1). The downstream sampling sites were at the intersection of each river and Lake Taihu.

Sample collection and laboratory analysis

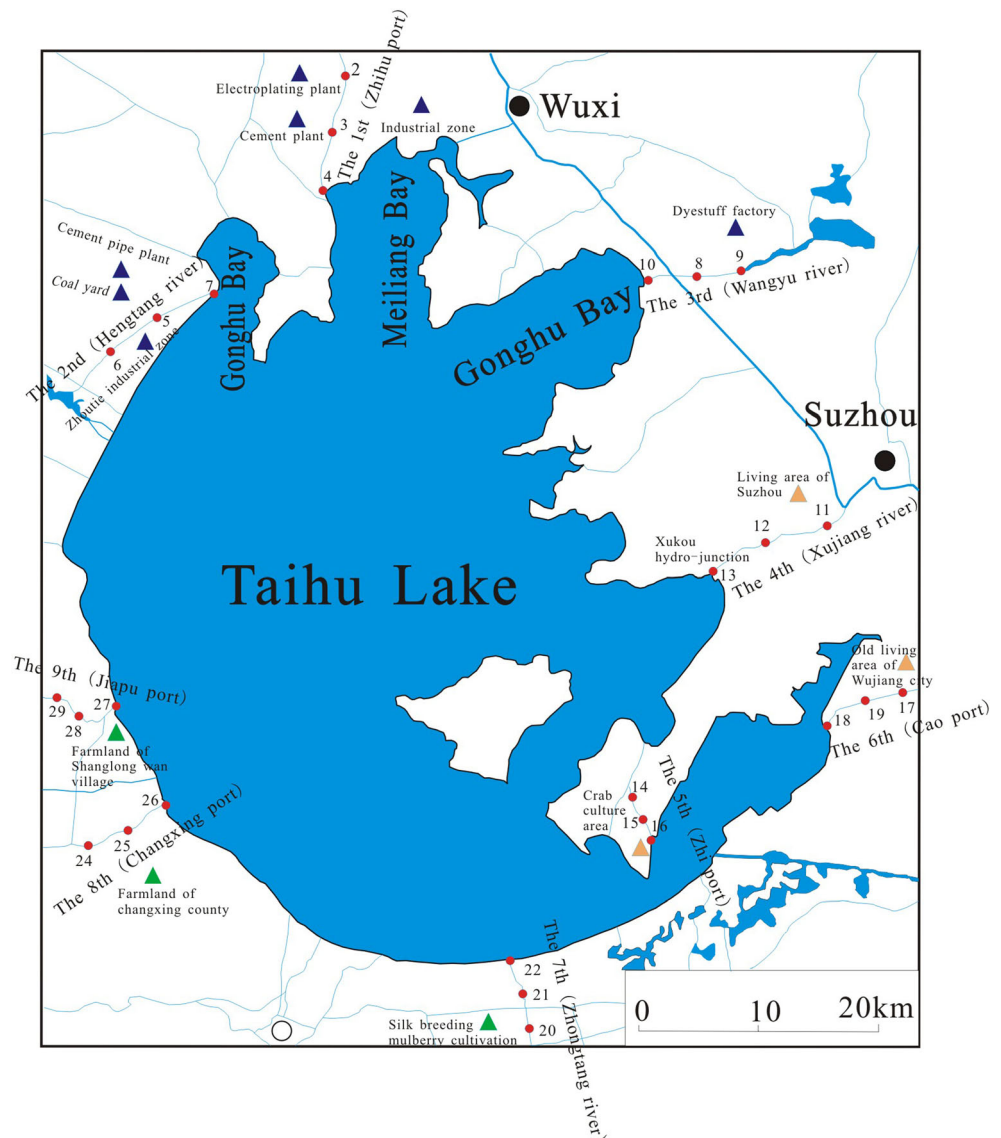
Water samples were collected at 0.5 m below the surface. Water temperature, pH, dissolved oxygen (DO), turbidity, and chlorophyll-*a* (Chl-*a*) were measured in situ using a Yellow Springs Instruments (YSI) 6600 V2 multi-sensor sonde. In the laboratory, a 25-mL raw water sample was digested by potassium persulfate, followed by TN measurements determined by spectrophotometry at 210 nm (Jin and Tu 1990). TP determinations were according to Jin and Tu (1990). For dissolved total nitrogen (DTN), a raw water sample was filtered through a Whatman GF/F glass fiber membrane (pre-combusted and cleaned with deionized water), followed by TN measurement. Dissolved total phosphorus (DTP) was determined by the TP procedure, but the digested solution was taken from the DTN measurement. A flow-injection system (Skalar Co., <http://www.skalar.com>) was used to determine ammonium N (NH₄⁺), nitrate N (NO₃⁻), and nitrite N (NO₂⁻) in Whatman GF/F membrane-filtered water samples. Particulate N (PN) was obtained by subtracting DTN from TN.

Results and discussion

Temporal and spatial variation in TN and DTN concentrations during three river flow conditions

TN and DTN concentrations of the 27 water samples declined gradually from the low flow period to the high flow period, and they showed very similar trends (Fig. 2). The average proportion of DTN in TN (DTN/TN) was 85.7% during low flow period, 56.3% during moderate flow period, and 68.4% during high flow period. While the average proportion of DTP/TP was 45.2% during low flow period, 38.6% during moderate flow period, and 25.7% during high flow period (Gao et al. 2016).

Fig. 1 Location of nine sampled rivers around Lake Taihu and 27 sampling sites of the rivers. Note: The 1st, 2nd, and 3rd were polluted by industry. The 4th, 5th, and 6th were polluted by domestic sewage. The 7th, 8th, and 9th were polluted by agriculture. Three sampling sites were set up along the upper, middle, and lower reaches of each river and the downstream sampling sites were at the intersection of each river and Lake Taihu. Blue triangle Represents the position of industrial pollution sources. Light orange triangle represents the position of domestic sewage pollution sources. Green triangle represents the position of agricultural pollution sources



Particulate phosphorus was the major component of TP, but DTN was the main component of TN in rivers around Lake Taihu. This indicates that P is more readily removed than N from river water by gravity settling within a river limited before it flows into Lake Taihu (Zhu et al. 2018b), especially during high flow. Therefore, more emphasis should be put on reducing discharge of N into rivers in order to reduce allochthonous N of Lake Taihu from inflowing rivers.

TN and DTN concentrations of the 4th, 5th, and 6th rivers decreased from upstream to downstream (Fig. 3). TN and DTN concentrations were 11.54 and 9.72 mg/L in the upstream, while 5.41 and 4.79 mg/L in the downstream segments of the Xujiang River (the 4th); 15.98 and 15.57 mg/L in the upstream, while 6.20 and 4.49 mg/L in the downstream segments of Zhi Port (the 5th); 7.70 and 7.12 mg/L in the upstream, while 3.14 and 2.00 mg/L in the downstream segments of Cao Port (the 6th). The data indicated that concentrations of TN and DTN naturally decreased by more than half from upstream to

downstream in these three rivers. This was most likely due to the Xujiang River, Zhi Port, and Cao Port being polluted by domestic sewage, and this type of river water has better self-purification ability than water polluted by agriculture and industry (Bayram et al. 2013; Brion et al. 2015). The main factors affecting the self-purification ability of river water include the following: (1) the types and properties of pollutants, the nature of water, aquatic organisms, dissolved oxygen in water, and other environmental factors; (2) self-purification of rivers mainly includes dilution, sedimentation, microbial decay and reaeration. Among these, sedimentation transports suspended matter from wastewater to the bottom of the river where the flow is gentle, and there is normally more suspended matter in the domestic sewage than in the industrial wastewater. Sedimentation plays an important role in the self-purification process of rivers polluted by domestic sewage. Microbial decay refers to the ability of heterotrophic bacteria, including pathogenic bacteria (e.g., coliforms) in feces to reproduce and

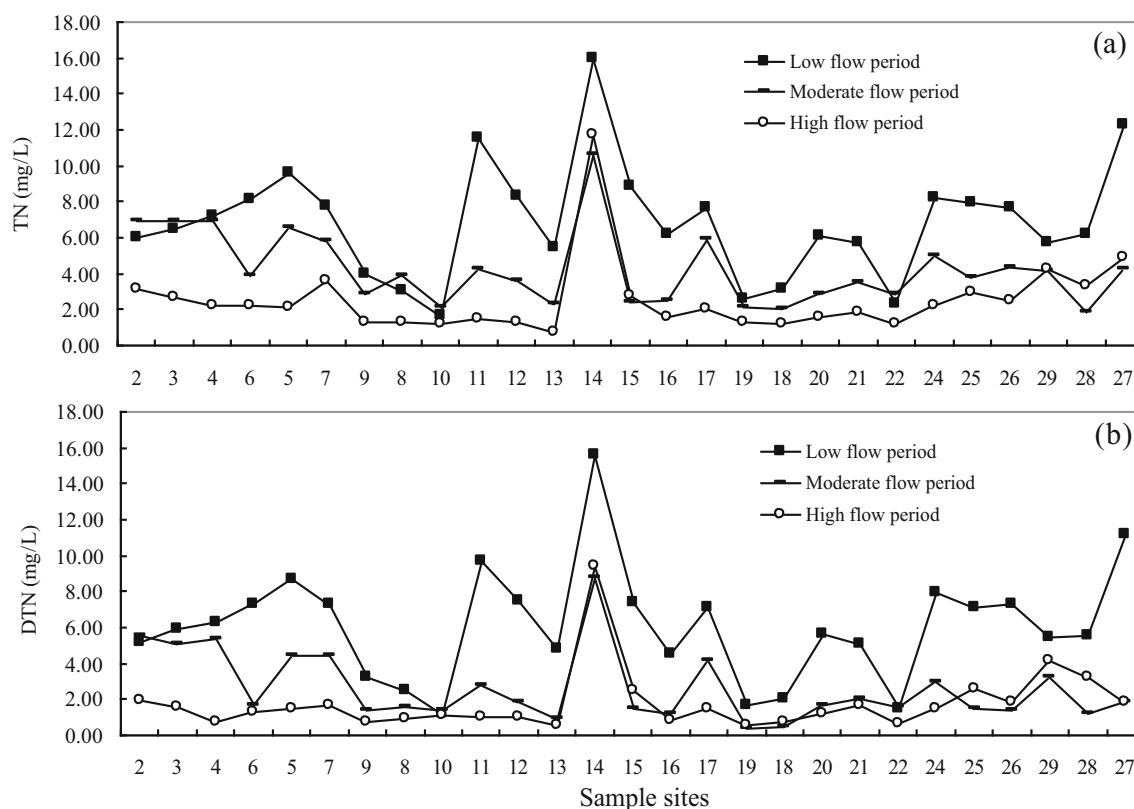


Fig. 2 TN and DTN concentrations of sampling sites in rivers around Lake Taihu during low, moderate, and high flow periods

increase with the increase of nutrients (food) in rivers. The number of heterotrophic bacteria is gradually reduced to the natural level with the gradual decrease of nutrients as well as the reproduction and ingestion of protozoa. The organic matter delivered by wastewater and their degradation product are good nutrient for saprophytic microorganisms. After entering the river, organic matter can be completely degraded and transformed into inorganic matter following microbial breakdown. The self-purification of microorganism in river water is also closely related to domestic sewage inputs.

There was not regular increase or decrease change in TN and DTN concentrations from upstream to downstream in rivers polluted by industry (the 1st, 2nd, and 3rd), which differed from those in rivers polluted by domestic sewage. Many factories were moved to the upper and middle reaches of Zhihu Port after the merger of Yangshan and Luoshe districts. There were electroplating plant (upstream), industrial zone, and cement plant (middle stream) drainages near the sampling sites of Zhihu Port (the 1st). The Hengtang River (the 2nd) is located in the Zhoutie industrial zone of Yixing and the middle stream sampling site was near a cement pipe plant and a coal yard. The main industry near the Wangyu River (the 3rd) upstream sampling site was a dyestuff factory. The source of pollutants contained in industrial wastewater was very complex, and there was a great difference in pollutants transported after discharge (Sun et al. 2016; Ukah et al. 2018), so the unpredictable change from upstream to downstream in rivers polluted by industry

appeared to stem from the diversification of industry along the rivers. TN and DTN average concentrations in rivers polluted by several industries during low, moderate, and high flow periods are shown in Table 1. N concentrations will probably only decrease if industrial discharge standards are strictly enforced and significant polluting industries cease discharging.

TN and DTN concentrations in upstream and downstream of rivers polluted by agriculture were closely related to specific agricultural activities. Figure 3 shows that TN and DTN concentrations in the upper reaches of the 7th river were higher than those in the lower reaches during the low flow period. The upper reaches of Zhongtang River (the 7th) are in Dongshanglin Village of Huzhou City, where silkworm breeding and mulberry cultivation are prominent. Mulberry cultivation has high N requirements. Most of the N fertilizer for mulberry cultivation is applied in winter (late November) and spring (February). As a result, TN and DTN concentrations in the upper reaches of the 7th river were higher than those in the lower reaches during the low flow period. TN concentrations in the lower reaches of Jiapu Port (the 9th) were higher than those in its upper reaches during all flow periods, especially during the low flow period in February. The Jiapu Port passes through the farmland of Shanglongwan Village and flows into Lake Taihu. It appears that the elevated N concentrations were the result of spring plowing and fertilization. TN and DTN concentrations reached 12.24 and 11.13 mg/L, respectively, in the lower reaches of Jiapu Port during the low flow period. Improved fertilizer

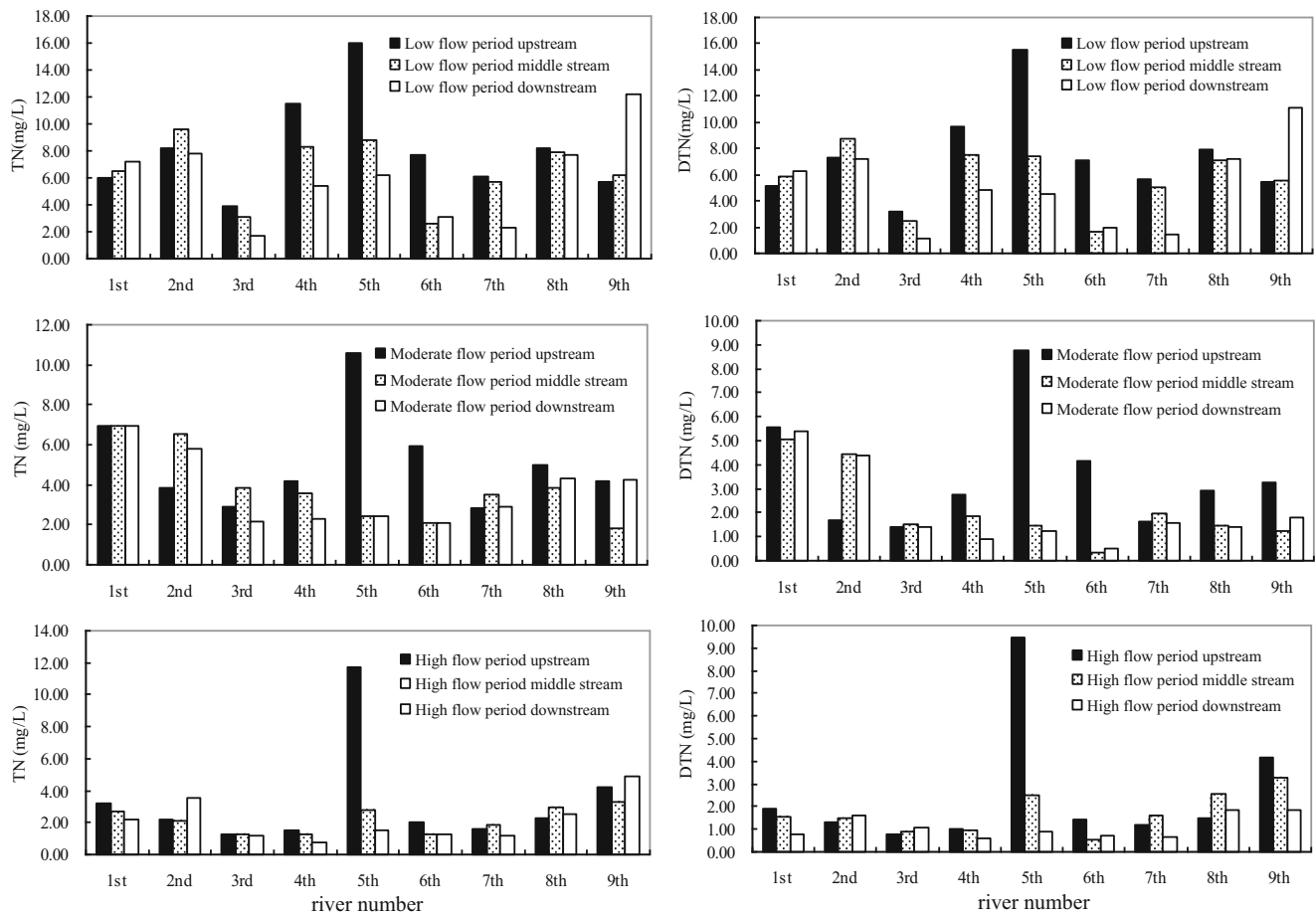


Fig. 3 TN and DTN concentrations changes from upstream to downstream in rivers around Lake Taihu during low, moderate, and high flow periods

management (increasing the utilization rate of N fertilizer and application of organic fertilizer) may be the key to reducing N concentrations of rivers polluted by agriculture.

With regard to impacts on Lake Taihu, N concentrations in the downstream segments of the rivers are most important. Table 2 lists TN and DTN concentrations in the downstream

of the three types of rivers. This indicates that TN and DTN concentrations in the lower reaches of rivers polluted by domestic sewage were the lowest in comparison with those of rivers polluted by industry and agriculture, although TN and DTN concentrations in typical domestic sewage were very high. In spring tillage season (low flow period), TN and DTN

Table 1 TN and DTN average concentrations in three types of rivers

Sampling rivers		Low flow period		Moderate flow period		High flow period	
		TN (mg/L)	DTN (mg/L)	TN (mg/L)	DTN (mg/L)	TN (mg/L)	DTN (mg/L)
Polluted by industry	The 1st	6.56 ± 0.64	5.78 ± 0.56	6.94 ± 0.03	5.34 ± 0.27	2.68 ± 0.48	1.41 ± 0.59
	The 2nd	8.51 ± 0.94	7.76 ± 0.83	5.40 ± 1.39	3.52 ± 1.57	2.63 ± 0.82	1.47 ± 0.17
	The 3rd	2.90 ± 1.14	2.30 ± 1.03	2.96 ± 0.84	1.46 ± 0.07	1.26 ± 0.06	0.92 ± 0.17
Polluted by sewage	The 4th	8.41 ± 3.07	7.34 ± 2.47	3.35 ± 0.99	1.84 ± 0.94	1.18 ± 0.40	0.86 ± 0.23
	The 5th	10.34 ± 5.06	9.15 ± 5.75	5.16 ± 4.69	3.81 ± 4.31	5.35 ± 5.58	4.26 ± 4.56
	The 6th	4.46 ± 2.82	3.60 ± 3.05	3.37 ± 2.23	1.67 ± 2.17	1.53 ± 0.47	0.90 ± 0.48
Polluted by agriculture	The 7th	4.70 ± 2.11	4.06 ± 2.26	3.08 ± 0.35	1.74 ± 0.22	1.55 ± 0.32	1.16 ± 0.51
	The 8th	7.91 ± 0.26	7.44 ± 0.44	4.38 ± 0.60	1.94 ± 0.86	2.55 ± 0.34	1.97 ± 0.56
	The 9th	8.04 ± 3.64	7.37 ± 3.36	3.40 ± 1.37	2.10 ± 1.04	4.15 ± 0.79	3.09 ± 1.18

Numbers in italics indicate distinct difference between the upper and lower reaches of the rivers

Table 2 TN and DTN concentrations in the downstream sampling sites of rivers and corresponding sampling sites of Lake Taihu

Downstream site (lake site)		Low flow period		Moderate flow period		High flow period	
		TN (mg/L)	DTN (mg/L)	TN (mg/L)	DTN (mg/L)	TN (mg/L)	DTN (mg/L)
Polluted by industry	1st (6#)	7.24 (6.37)	6.29 (5.91)	6.91 (3.61)	5.39 (3.24)	2.21 (2.65)	0.77 (1.34)
	2nd (17#)	7.79 (4.31)	7.25 (3.94)	5.80 (5.84)	4.41 (4.98)	3.58 (2.98)	1.63 (2.35)
	3rd (14#)	1.69 (1.11)	1.18 (0.75)	2.16 (2.80)	1.42 (2.41)	1.20 (1.58)	1.09 (1.16)
Polluted by sewage	4th (28#)	5.41 (0.98)	4.79 (0.73)	2.27 (1.92)	0.89 (1.75)	0.74 (0.96)	0.59 (0.64)
	5th (26#)	6.20 (1.34)	4.49 (0.77)	2.46 (1.76)	1.21 (1.50)	1.54 (1.13)	0.87 (0.97)
	6th (25#)	3.14 (1.71)	2.00 (0.91)	2.07 (1.12)	0.51 (0.75)	1.23 (1.61)	0.71 (1.19)
Polluted by agriculture	7th (24#)	2.28 (1.73)	1.46 (0.92)	2.90 (2.04)	1.58 (1.73)	1.20 (0.82)	0.63 (0.75)
	8th (20#)	7.67 (2.00)	7.25 (1.21)	4.30 (4.07)	1.42 (3.10)	2.51 (3.17)	1.83 (2.95)
	9th (20#)	12.24 (2.00)	11.13 (1.21)	4.23 (4.07)	1.80 (3.10)	4.90 (3.17)	1.85 (2.95)

Numbers in italics indicate TN or DTN concentration in Lake Site was higher than that in downstream site of river

concentrations in the downstream of rivers polluted by agriculture were highest, but those in the downstream of rivers polluted by industry during the moderate flow period had the greatest impact on Lake Taihu. The impact of rivers polluted by industry and agriculture on Lake Taihu was similar during the high flow period because of the large water volume.

In Table 2, we compared TN and DTN concentrations in downstream sampling sites of these rivers with those (routine monitoring data of Taihu Laboratory for Lake Ecosystem Research) in the corresponding sampling sites of Lake Taihu. All the TN and DTN concentrations in downstream of these rivers were higher than those in the corresponding lake area during the low flow period. Therefore, these rivers had a distinct impact on TN and DTN concentrations in Lake Taihu during the low flow period. Rivers polluted by agriculture had the greatest impact on Lake Taihu's TN and DTN concentrations, followed by rivers polluted by industry, while rivers polluted by domestic sewage had the least impact. The internal N concentration in Lake Taihu was highlighted during moderate and high flow periods: TN concentrations of half the rivers and DTN concentrations of almost all the rivers at their intersection with Lake Taihu were lower than those in the corresponding lake area. It showed that TN and DTN concentrations of Lake Taihu had already been very high and that efforts must be taken to reduce internal N while controlling allochthonous N, including dredging and removing N by collecting aquatic vegetation and biota. Only by combining reductions in external N inputs and reducing internal N sources will eutrophication of Lake Taihu be mitigated.

Temporal and spatial variation characteristics of NH_4^+ , NO_3^- , and NO_2^- concentrations in three river types

Dissolved inorganic N in water mainly exists as NH_4^+ , NO_3^- , NO_2^- , and N_2 . Aquatic organisms can use the first three forms

of N, except only N_2 -fixing cyanobacteria can utilize N_2 (Dodds et al. 2002; Smith 2003). NH_4^+ and NO_3^- are the main inorganic N sources to support algal growth. Cyanobacteria and phytoplankton typically favor NH_4^+ uptake over NO_3^- because NO_3^- enters the cyanobacteria cell via active transport and it must be reduced to NO_2^- and then to NH_4^+ by nitrate-reductase and nitrite-reductase, respectively, before it can be assimilated (Syrett and Morris 1963; Raven et al. 1992; Addy et al. 2017).

NH_4^+ is produced by microbial decomposition of N-containing organic matter. Therefore, elevated NH_4^+ concentrations are indicative of water enriched in organic matter. Allochthonous N entering the system is often dominated by NO_3^- (Chaffin and Bridgeman 2014), although organic N can be a significant fraction, especially in anthropogenically disturbed watersheds, where animal and other waste products are discharged to streams and rivers (Osburn et al. 2016). N-limited *Microcystis* blooms in the Klamath River, California, USA showed the same growth yield as additions of NH_4^+ , NO_3^- (Moisander et al. 2009). In Eastern US lakes, enrichment of NO_3^- stimulated *Microcystis* growth more frequently than did NH_4^+ (Davis et al. 2010). An experiment on N utilization by N-limited cyanobacteria occurring during blooms (Lake Erie's Maumee and Sandusky Bays and Grand Lake St. Marys in Ohio, USA) demonstrated that additional NO_3^- and NH_4^+ exacerbated *Microcystis* blooms (Chaffin and Bridgeman 2014). Thus, both NO_3^- and NH_4^+ need to be targeted (reduced) to alleviate cyanobacteria blooms. Osburn et al. (2016) reported that organic N, especially dissolved organic N, should also be targeted for reduction.

Substandard discharge of industrial wastewater, a rapid increase in population and relatively weak treatment of municipal domestic sewage, and excessive applications of chemical fertilizer will lead to the increase of NH_4^+ and NO_3^- in rivers (Van Drecht et al. 2009; Billen et al. 2013; Zhang et al. 2015). Figure 4 shows the variation of NH_4^+ and NO_3^- concentrations from upstream to

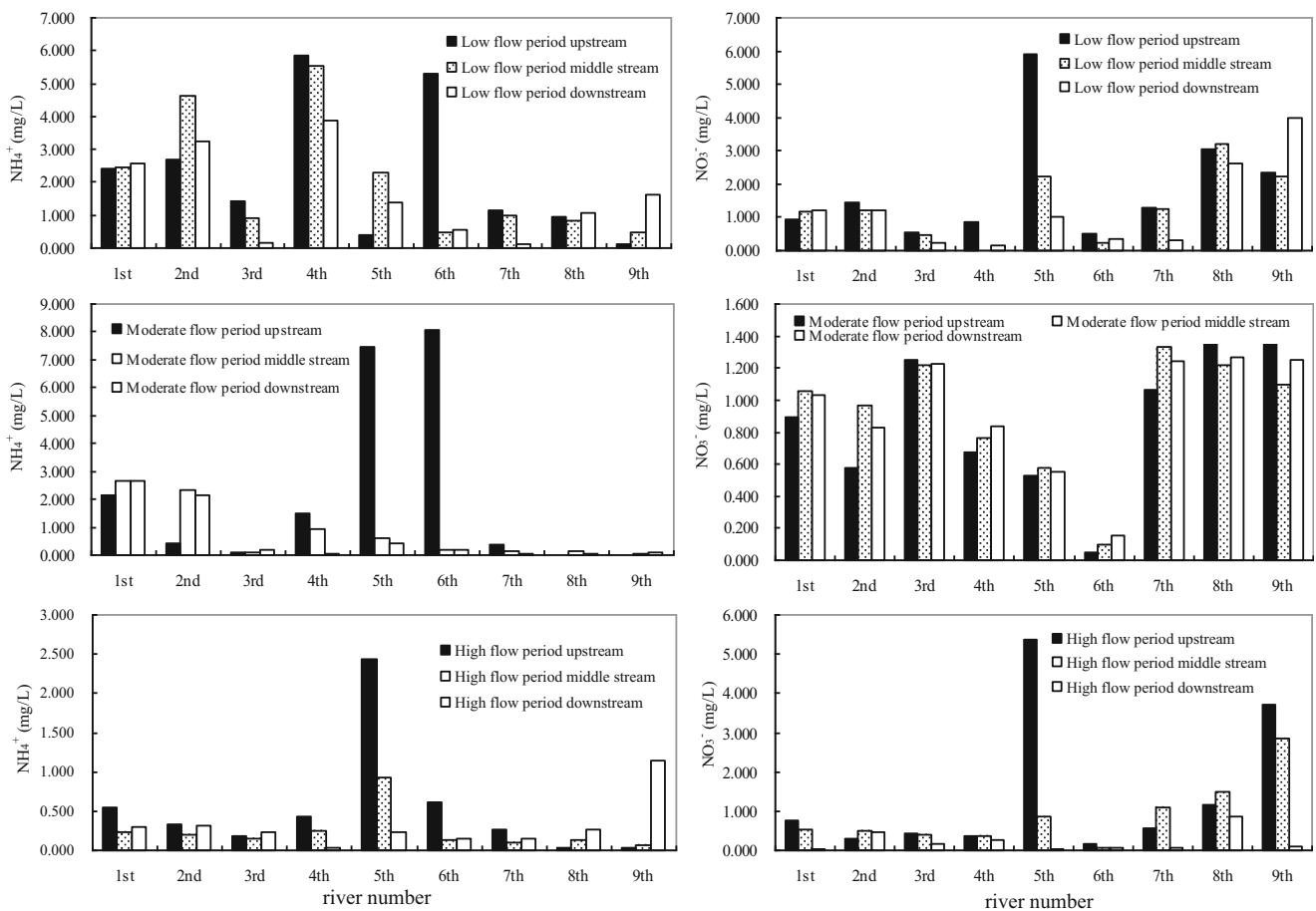


Fig. 4 NH_4^+ and NO_3^- concentrations changes from upstream to downstream in rivers around Lake Taihu during low, moderate, and high flow periods

downstream in the rivers during low, moderate, and high flow periods. NH_4^+ concentration showed an obvious declining trend from upstream to downstream in rivers polluted by domestic sewage except for the 5th river during the low flow period. The rivers polluted by industry and agriculture showed no regularity from upstream to downstream. NH_4^+ concentration in the upper reaches of Zhongtang River (the 7th) was higher than that in the lower reaches during low flow period. NH_4^+ concentration in the lower reaches of Jiapu Port (the 9th) was higher than that in the upper reaches during all flow periods, especially in February. The variations of NH_4^+ concentrations in the 7th and 9th rivers were very similar to TN and DTN (Fig. 3), maybe it was also caused by mulberry growing in Dongshanglin Village and the farmland of Shanglongwan Village.

The highest NH_4^+ average concentrations (2.856 mg/L during low flow period, 2.146 mg/L during moderate flow period, 0.581 mg/L during high flow period) were in the rivers polluted by domestic sewage, followed by those (2.271 mg/L during low flow period, 1.415 mg/L during the moderate flow period, 0.277 mg/L during high flow period) in the rivers polluted by industry. The lowest NH_4^+ average concentrations (0.813 mg/L during low flow period, 0.099 mg/L during moderate flow period, 0.242 mg/L during high flow period) were in the rivers

polluted by agriculture. High NH_4^+ concentrations are characteristic of human and animal excreta and domestic sewage inputs.

The NH_4^+ average concentration in rivers polluted by industry (the 1st, 2nd, 3rd) and domestic sewage (the 4th, 5th, 6th) decreased with low, moderate, and high flow periods. However, they were higher during the low and high flow periods in rivers polluted by agriculture, because February (low flow period) and August (high flow period) were fertilization periods for wheat and rice in Lake Taihu Basin. Other studies also showed that the highest agriculture nitrogen losses occurred during spring and autumn (Bonaiti and Borin 2010; Pfannerstill et al. 2016).

Table 3 lists NH_4^+ concentrations in the downstream sampling sites of these rivers. According to Environmental Quality Standards for surface water of PRC (GB3838-2002), NH_4^+ concentrations in the Zhihu Port (the 1st) and the Hengtang River (the 2nd) were up to grade V (grade V criterion is NH_4^+ concentration > 2.0 mg/L) during low and moderate flow periods. They were between grade I and grade II ($0.15 \text{ mg/L} < \text{NH}_4^+$ concentration $\leq 0.5 \text{ mg/L}$) only during the high flow period. By comparison, NH_4^+ concentrations in rivers polluted by industry had greater impact on Lake Taihu. However, NH_4^+ concentrations in the lower reaches of the Wangyu River (the 3th) had been improved to between grade I and grade II from

Table 3 NH_4^+ and NO_3^- concentrations in the downstream sampling sites of rivers and corresponding sampling sites of Lake Taihu

Downstream site (lake site)		Low flow period		Moderate flow period		High flow period	
		NH_4^+ (mg/L)	NO_3^- (mg/L)	NH_4^+ (mg/L)	NO_3^- (mg/L)	NH_4^+ (mg/L)	NO_3^- (mg/L)
Polluted by industry	1st (6#)	2.580 (3.14)	1.225 (1.46)	2.660 (0.70)	1.035 (1.76)	0.296 (0.46)	0.041 (0.41)
	2nd (17#)	3.235 (1.80)	1.215 (1.68)	2.155 (2.39)	0.831 (1.76)	0.316 (0.84)	0.457 (0.49)
	3rd (14#)	0.148 (0.45)	0.249 (0.11)	0.205 (0.42)	1.223 (0.99)	0.240 (0.37)	0.159 (0.51)
Polluted by sewage	4th (28#)	3.880 (0.38)	0.139 (0.16)	0.031 (0.29)	0.840 (0.71)	0.040 (0.31)	0.277 (0.11)
	5th (26#)	1.392 (0.33)	1.034 (0.17)	0.398 (0.29)	0.553 (0.54)	0.238 (0.32)	0.021 (0.44)
	6th (25#)	0.554 (0.56)	0.361 (0.24)	0.164 (0.48)	0.158 (0.09)	0.144 (0.43)	0.056 (0.23)
Polluted by agriculture	7th (24#)	0.126 (0.61)	0.323 (0.28)	0.043 (0.36)	1.224 (0.78)	0.153 (0.40)	0.054 (0.33)
	8th (20#)	1.076 (0.23)	2.614 (0.41)	0.041 (0.24)	1.269 (1.94)	0.273 (0.43)	0.869 (1.16)
	9th (20#)	1.613 (0.23)	3.971 (0.41)	0.079 (0.24)	1.254 (1.94)	1.139 (0.43)	0.090 (1.16)

Numbers in italics indicate NH_4^+ or NO_3^- concentration in Lake Site was higher than that in downstream site of river

grade V (Gao et al. 2011) during all flow periods after the comprehensive water environment improvement of Wangyu River was implemented between 2008 and 2009. NH_4^+ concentration in the downstream region of the Xujiang River (the 4th) was very high, leading to grade V conditions during low flow period at the Xukou sampling site (hydro-junction). NH_4^+ concentration in the downstream of Zhi Port (5th) almost reached grade IV (NH_4^+ concentration ≤ 1.5 mg/L) during low flow period, because the sampling site was a crab culture area. As for most rivers (not affected by shipping, aquaculture, and other factors) polluted by domestic sewage, NH_4^+ concentrations at the intersections of river and Lake Taihu would gradually be reduced to between grade I and grade II due to river self-purification ability. NH_4^+ concentrations in rivers polluted by agriculture were also between grade I and grade II at their entrance to Lake Taihu, except for the downstream sampling site of the 9th river that just passed through the farmland of Shanglong Village. Based on Table 3, NH_4^+ concentrations in the corresponding lake area showed no obvious differences among low, moderate, and high flow periods, and they were generally below 0.5 mg/L (grade II). Several high NH_4^+ concentrations appeared in north Zhushan Bay (17#) and Meiliang Bay (6#) of Lake Taihu, but the Chl-*a* concentrations were only 11.90 and 4.24 $\mu\text{g/L}$ during the moderate flow period, 17.19 and 7.81 $\mu\text{g/L}$ during the low flow period at 17# and 6# sampling sites according to routine monitoring data of Taihu Laboratory for Lake Ecosystem Research.

When comparing NH_4^+ concentrations in the downstream sampling sites of these rivers with those in the corresponding sampling sites of Lake Taihu (Table 3), most downstream riverine NH_4^+ concentrations were obviously higher than those in the corresponding lake area only during the low flow period. By comparison, the rivers polluted by industry had a greater impact on Lake Taihu's NH_4^+ concentration than other rivers.

The change in NO_3^- concentration from upstream to downstream in these rivers differed from TN, DTN, and NH_4^+ , and

there was no regularity observed, even in rivers polluted by domestic sewage (see Fig. 4). This phenomenon was probably caused by the transformations among various dissolved inorganic nitrogen (i.e., NH_4^+ , NO_2^- , and NO_3^-). Waters polluted by fecal organic nitrogen or protein catabolism metabolites would undergo decomposition to NH_4^+ (Hotta et al. 2007; Ebeling et al. 2006), and then NO_2^- by nitrifying bacteria (*Nitrosomonas*), finally NO_3^- was produced by *Nitrobacter* (Mook et al. 2012). Whether there were new organic pollutants entering the water and the degree of river self-purification all affect NO_3^- concentration. The most significant feature was that the NO_3^- average concentrations in rivers polluted by agriculture were the highest (2.253 mg/L during low flow period, 1.277 mg/L during moderate flow period, 1.319 mg/L during high flow period). The NO_3^- average concentrations in rivers polluted by domestic sewage were 1.252 mg/L during low flow period, 0.471 mg/L during moderate flow period, and 0.834 mg/L during high flow period. In rivers polluted by industry, values were 0.938 mg/L during the low flow period, 1.007 mg/L during moderate flow period, and 0.401 mg/L during the high flow period, respectively. Agricultural N fertilizer is generally applied as $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and organic N. In whatever form it is applied, soil microorganisms ultimately generate NH_4^+ and NO_3^- , which are biologically available, including the target crop (Mayzelle 2013). Nitrate ions and soil both have negative charges, so nitrate ions are not adsorbed to soil particles and are easily leached into rivers from agriculture soil. As a result, the NO_3^- average concentrations in rivers polluted by agriculture around Lake Taihu were the highest.

Table 3 lists NO_3^- concentrations in downstream sampling sites of these rivers and those in the corresponding lake area. It can be seen that NO_3^- concentrations in rivers polluted by agriculture were the highest, followed by those in the rivers polluted by industry. NO_3^- concentrations in rivers polluted by domestic sewage had no significant difference with those in the corresponding lake area during the low and moderate

flow periods. The greatest impact on Lake Taihu was from rivers polluted by agriculture during low flow period.

The continuous ingestion of food containing NO_3^- and NO_2^- additives also brings potential hazards for human health. Nitrite (NO_2^-) is the main toxic agent as it can react with secondary or tertiary amines to produce nitrosamines which are potential mutagenic, carcinogenic, and teratogenic (Wang et al. 2006; Almeida et al. 2007). Nitrite can also combine with hemoglobin to form methemoglobin and result in anoxia (Camargo and Alonso 2006; Mook et al. 2012). It is unstable and converted easily into NH_4^+ or NO_3^- by microorganism or oxidant, and NO_2^- is also used as N source by algae. NO_2^- concentrations had no regularity from upstream to downstream of the rivers because of its instability. They were lower than NH_4^+ and NO_3^- concentrations in all the sampling sites in any flow period. The highest NO_2^- average concentrations (0.105 mg/L during low flow period, 0.250 mg/L during moderate flow period, and 0.105 mg/L during high flow period) were in the rivers polluted by industry. NO_2^- average concentrations in rivers polluted by domestic sewage were 0.087, 0.092, and 0.064 mg/L, respectively, during low, moderate, and high flow periods. They were 0.079, 0.022, and 0.121 mg/L, respectively, during low, moderate, and high flow periods in rivers polluted by agriculture. NO_2^- concentration is not limited in the Environmental Quality Standards for surface water of PRC (GB3838-2002), although it is toxic to human health; maybe it is because NO_2^- is unstable and easily converted (Ellis et al. 1998; Moorcroft et al. 2001; Almeida et al. 2007). The European Community has established the maximum allowable levels of nitrites in drinking water at 0.1 ppm (Almeida et al. 2007). Some studies showed that NO_2^- can be harmful to fish when it exceeded 0.06 mg/L (Eddy and William 1987). When we compared NO_2^- concentrations in the downstream segments of these rivers, we found higher values (0.436, 0.271 mg/L) appeared in the downstream sampling sites of rivers (the 1st and 2nd) polluted by industry and the other high values (0.245, 0.235 mg/L) appeared in rivers polluted by agriculture. As a result, NO_2^- concentrations in rivers polluted by domestic sewage had less impact on Lake Taihu than those in rivers polluted by industry and agriculture.

Temporal and spatial variation characteristics of dissolved inorganic nitrogen concentrations in three river types

Dissolved inorganic nitrogen (DIN) includes NH_4^+ , NO_3^- , and NO_2^- . The average proportion of DIN in TN (DIN/TN) of all these rivers was 49.1% during low flow period, 48.7% during moderate flow period, and 45.7% during high flow period. The highest DIN average concentrations were in the rivers polluted by domestic sewage; they were 4.196, 2.710, and 1.479 mg/L, respectively, during low, moderate, and high flow periods. DIN average concentrations in rivers polluted by industry were 3.315, 2.672, and 0.783 mg/L, respectively, during low, moderate, and

high flow periods. The relatively lowest DIN average concentrations were 3.144, 1.398, and 1.682 mg/L, respectively, during low, moderate, and high flow periods in rivers polluted by agriculture. DIN average concentrations in three types of rivers decreased with low, moderate, and high flow periods in almost all cases.

We compared DIN concentrations in downstream segments of these rivers with those in the receiving lake water (Table 4). Most of the downstream DIN concentrations were higher than those in the lake during the low flow period while the opposite was true during moderate and high flow periods. When comparing DIN concentrations in the downstream of these rivers during low flow period, rivers polluted by agriculture had the largest impact on Lake Taihu. Table 3 and Table 4 show that NH_4^+ was the chief component of DIN in the downstream of rivers polluted by domestic sewage during low flow period but NO_3^- was the bulk of DIN in the downstream of rivers polluted by agriculture during low and moderate flow periods.

Targeted approaches should be chosen to cut down allochthonous DIN from inflowing rivers according to the main N species of three types of rivers during low flow period. Biological nitrogen removal (BNR) processes will be needed to remove inorganic nitrogen of domestic sewage and constructing wetlands or drainage ditches serve as treatment facilities for agricultural wastes (Bosch et al. 2013; King et al. 2016; Paerl et al. 2018). Drainage ditches are often at the interface between terrestrial and larger water bodies such as rivers and lakes (Kumwimba et al. 2016). Vegetated drainage ditches have received attention in recent years for their successful retention and removal of agricultural contaminants (Moore et al. 2010; Elsaesser et al. 2013; Iseyemi et al. 2016; Kumwimba et al. 2017). Some studies showed that drainage ditches were a good way to remove NH_4^+ from untreated domestic sewage if they contained *Cladophora* sp. as a resident species (Zhang et al. 2009; Liu and Vyverman 2015; Kumwimba and Zhu 2017).

Temporal and spatial variation characteristics of PN concentrations in three river types

PN is essential factors in nutrient loading and act as potential sources of dissolved nutrients (Zuo et al. 2016; Duan et al. 2016). The average proportion of PN in TN (PN/TN) was 14.3% during the low flow period, 43.7% during the moderate flow period, and 31.6% during the high flow period. PN/TN in the rivers during the moderate and high flow periods was higher than that during the low flow period. This may be due to the soil erosion caused by more water volume during moderate and high flow periods. Figure 5 shows the variation of PN/TN from upstream to downstream in the rivers during low, moderate, and high flow periods. In most cases PN/TN in the downstream were higher than that in the upstream (Fig. 5). Along the flow direction, DIN decreased with the growth of primary and secondary producers (Burford et al. 2008), PN/TN increased with the decrease

Table 4 DIN concentrations in the downstream sampling sites of rivers and corresponding sampling sites of Lake Taihu

Downstream site (lake site)		Low flow period DIN (mg/L)	Moderate flow period DIN (mg/L)	High flow period DIN (mg/L)
Polluted by industry	1st (6#)	3.968 (4.747)	4.131 (2.492)	0.356 (0.963)
	2nd (17#)	4.563 (3.513)	3.257 (4.340)	0.852 (1.405)
	3rd (14#)	0.401 (0.564)	1.460 (1.468)	0.413 (0.893)
Polluted by sewage	4th (28#)	4.101 (0.546)	0.871 (1.009)	0.324 (0.438)
	5th (26#)	2.530 (0.506)	1.020 (0.839)	0.264 (0.766)
	6th (25#)	0.934 (0.808)	0.332 (0.577)	0.210 (0.699)
Polluted by agriculture	7th (24#)	0.457 (0.900)	1.294 (1.154)	0.217 (0.733)
	8th (20#)	3.758 (0.649)	1.310 (2.190)	1.387 (1.783)
	9th (20#)	5.819 (0.649)	1.356 (2.190)	1.325 (1.783)

Numbers in italics indicate DIN concentration in Lake Site was higher than that in downstream site of river

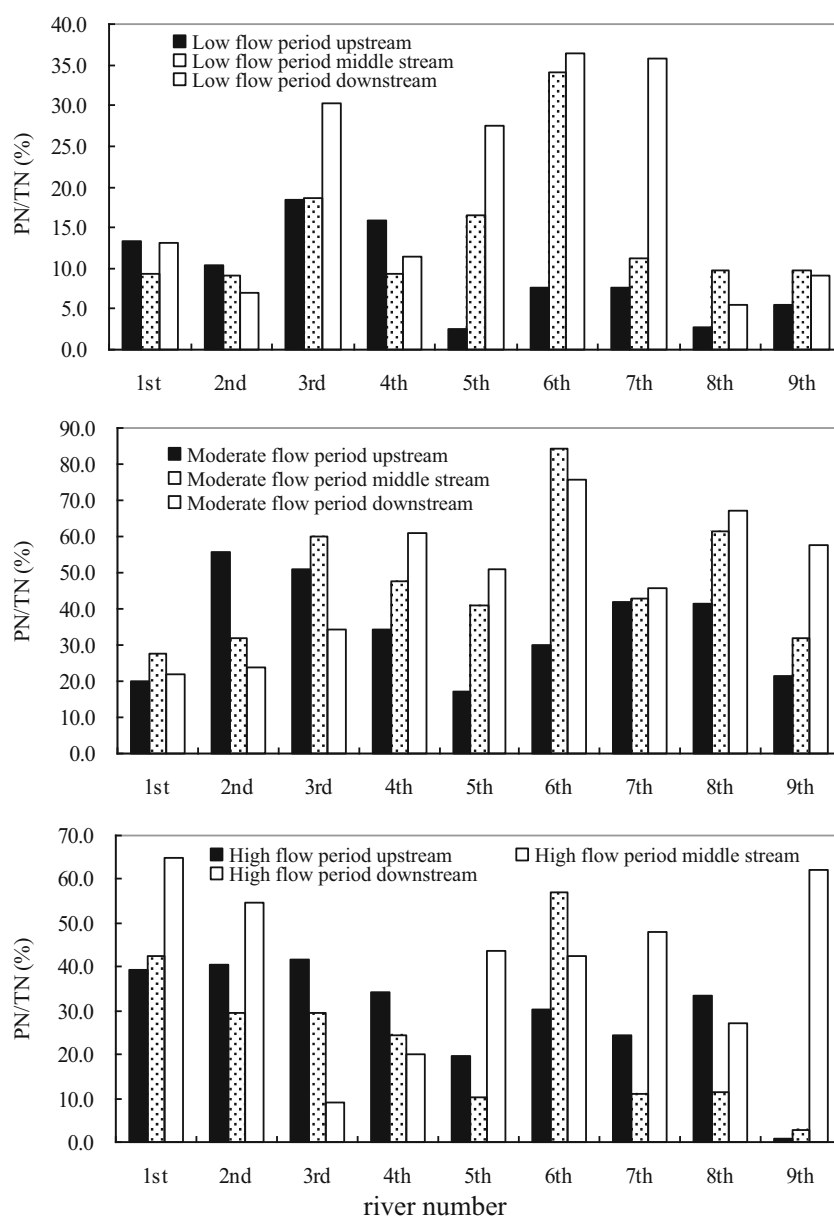
Fig. 5 PN/TN changes from upstream to downstream in rivers around Lake Taihu during low, moderate, and high flow periods

Table 5 PN concentrations in the downstream sampling sites of rivers and corresponding sampling sites of Lake Taihu

Downstream site (lake site)		Low flow period PN (mg/L)	Moderate flow period PN (mg/L)	High flow period PN (mg/L)
Polluted by industry	1st (6#)	0.94 (0.46)	1.52 (0.37)	1.44 (1.31)
	2nd (17#)	0.54 (0.37)	1.39 (0.86)	1.95 (0.63)
	3rd (14#)	0.51 (0.36)	0.74 (0.39)	0.11 (0.42)
Polluted by sewage	4th (28#)	0.62 (0.25)	1.38 (0.17)	0.15 (0.32)
	5th (26#)	1.71 (0.57)	1.25 (0.26)	0.67 (0.16)
	6th (25#)	1.14 (0.80)	1.56 (0.37)	0.52 (0.42)
Polluted by agriculture	7th (24#)	0.81 (0.81)	1.32 (0.31)	0.57 (0.07)
	8th (20#)	0.42 (0.79)	2.88 (0.97)	0.68 (0.22)
	9th (20#)	1.11 (0.79)	2.43 (0.97)	3.05 (0.22)

of DIN. Another reason was that PN itself increased from up-stream to downstream with the soil erosion around the rivers.

We compared PN concentrations in downstream segments of these rivers with those in receiving lake water (Table 5). It can be seen that almost all the downstream PN concentrations were higher than those in the lake during low, moderate, and high flow periods. PN had an impact on Lake Taihu and all the three types of rivers would increase PN concentration in the lake. It is therefore necessary to reduce PN concentration before the rivers flow into Lake Taihu. Setting up vegetated drainage ditches or wetland (Coveney et al. 2002) at the interface between the rivers and the lake may be also a good option for reducing PN concentration.

Conclusions

- (1) DTN was the main component of TN in rivers around Lake Taihu and it is difficult to remove, so more emphasis should be put on reducing discharge of N into rivers.
- (2) Inflowing rivers had distinct impacts on TN, DTN, NH_4^+ , and NO_3^- concentrations of Lake Taihu during low flow period. Rivers polluted by agriculture had the greatest impact on the Lake's TN, DTN, NO_3^- , and DIN concentrations while rivers polluted by industry had the greatest impact on NH_4^+ concentration. Therefore, agriculture and industry should be key targets for N removal. Improved fertilizer management and industrial discharge standards should be strictly enforced. Rivers polluted by domestic sewage had a lower impact on N concentration of Lake Taihu because of riverine self-purification ability from upstream to downstream when compared with rivers polluted by agriculture and industry.
- (3) NO_3^- was the chief component of DIN in the downstream of rivers polluted by agriculture and NH_4^+ formed the bulk of DIN in the downstream of rivers polluted by domestic sewage during low flow period. PN had an impact on Lake Taihu during the three flow periods and all the three types of rivers would increase PN concentration in the lake.

Therefore, targeted approaches should be chosen to effectively remove allochthonous N of Lake Taihu from inflowing rivers. Drainage ditches with effective nutrient-scavenging plants in them could be a solution.

- (4) The internal N concentrations of Lake Taihu were higher than those of inflowing rivers during moderate and high flow periods. Efforts should focus on reducing internal N while controlling allochthonous N, including dredging and removing N by collecting aquatic vegetation and biota. Only by combining reductions in external N inputs and reducing internal N sources will eutrophication of Lake Taihu be mitigated.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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