Nutrient concentrations and fluxes in the upper catchment of the Miyun Reservoir, China, and potential nutrient reduction strategies

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Abstract The Miyun Reservoir is Beijing's main drinking water source. Increased nutrient levels in the reservoir have resulted in an increased risk of harmful algal blooms. One hundred ten water samples were collected at a range of spatial scales in the upper catchment of the Miyun Reservoir and were analyzed for total nitrogen (TN), nitrate (NO₃⁻-N), ammonium (NH₄⁺-N), total phosphorus (TP), and the potassium permanganate index (COD_{Mn}). Empirical equations were developed from relationships between nutrient concentrations and the main controls on nutrient, and were used to identify parts of the catchment that should be targeted with nutrient load reduction measures. Cropland was the main source of sediment for the streams, and much of the phosphorus was associated with sediment. The annual mean TP concentrations were closely correlated with both the annual mean suspended sediment concentrations and the ratio of the cropland area to the total basin area. There was a linear relationship between the annual mean TN concentration and the population density in the basins. Soil conservation may play an

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important role in reducing TP concentrations in the upper reaches of the Chao and Bai Rivers. It may be useful to (1) construct natural riparian buffers and vegetated buffers along croplands close to the watercourses, (2) implement management strategies to reduce nitrogen (N) fertilizer applications, and (3) construct additional wetlands to reduce nutrient loads in the study area.

Keywords Nutrient concentrations · Influencing factors · Empirical equations · Control measures · Miyun Reservoir

Introduction

Eutrophication refers to the nuisance enrichment of water bodies by excessive nutrients. Increased nutrient concentrations cause degradation of water bodies by, for example, increasing the growth of aquatic vegetation, which consumes dissolved oxygen when it dies and decays, and by promoting the development and persistence of many harmful algal blooms (Heisler et al. 2008; Lewitus et al. 2012) that can cause severe environmental problems. Eutrophication remains one of the most common water quality problems around the world and is an active area of scientific research (Smol 2008; Ferreira et al 2011; Ekholm and Lehtoranta 2012).

Since the 1980s, increasing population and social changes have resulted in steady increases in the demand for food and energy in China, which in turn have stimulated the development of agriculture and industry. Excessive amounts of anthropogenically produced nutrients have caused eutrophication in many lakes and reservoirs throughout China (Du et al 2011; Wu and Wang 2012; Zhao et al 2013), and nutrient enrichment of freshwaters continues to be one of the most serious problems for surface water management. Consequently, many studies have reported temporal and spatial variations in nutrient concentrations in large Chinese rivers and reservoirs (Domagalski et al 2007; Shen and Liu 2009; Yu et al 2010). Many researchers have simulated nutrient losses using models such as Soil and Water Assessment Tool (SWAT; Neitsch et al 2005) and Agriculture Non-Point Source Pollution model (AGNPS; Young et al 1987) to identify potential source areas of nutrient pollution and to provide suggestions for reducing non-point source (NPS) pollution. Wu and Chen (2013) used the SWAT model to calculate the spatial and seasonal distribution of nutrient loads in the East River Basin, while Shang et al. (2012) developed an improved SWAT-based computational framework for identifying critical source areas of agricultural pollution in the Lake Erhai Basin. Jiang et al. (2014) applied SWAT to estimate the impact of conservation tillage and fertilizer management on the reduction of total nitrogen (TN) and total phosphorus (TP) loads in the Liuxi River watershed. Yang et al. (2011) coupled the Xinanjiang and SWAT models to simulate nitrate $(NO_3^{-}N)$, ammonium $(NH_4^{+}N)$, and TP concentrations in runoff in the Songtao Watershed; results showed that, with a 40 % reduction in fertilization, TN and TP loads could be reduced by 7.51 and 7.76 %, respectively. Liu et al. (2008) calibrated and validated AGNPS with data collected from 10 storms in the Wuchuan Catchment.

Most of the physically based NPS models that are commonly used in China were developed in the USA. Although these models have been used in China, empirical research is limited and there is inadequate data to calibrate models or to derive export coefficients (Shen et al 2012). Agriculture in China is very different from that in the USA, so empirical data collected in the USA cannot be used in China; as such, many empirical studies of sediment and chemical runoff processes in Chinese agricultural systems are required at different scales (plot, field, catchment, river basin) (Ongley et al 2010) to support more accurate model calibration. In recent years, there has been a small, but growing, body of empirically based studies of nutrient loads relative to different land uses in China (Zhao et al 2011; Chen et al 2013; Lu et al. 2013b). However, because of the vast area and huge number of rivers and reservoirs involved, it will be very difficult to use the findings from empirical studies of NPS pollution in China to construct physically based NPS models for the foreseeable future. Given the urgent need to make rapid progress in the identification of nutrient source areas so that nutrient loads can be controlled, an alternative estimation and analysis method is required.

Beijing is one of the largest cities in the world and is currently facing a severe water shortage. The Miyun Reservoir is the main source of domestic water for Beijing. In recent years, nutrient levels in the water and sediment of the Miyun Reservoir have been documented and reported by researchers (Wang et al 2005, 2013a) and the factors that influence the seasonal, interannual, and spatial variations of nutrient concentrations in the reservoir have been discussed (Wang et al 2008). Moreover, the role that nutrients have played in stimulating the growth and proliferation of phytoplankton in the reservoir has been studied (Su et al 2013). Wang et al. (2013b) used a numerical model to simulate hydrodynamic and algal processes in the Miyun Reservoir. Levels of organic matter in the surface soils (Lu et al. 2013a) and NO₃⁻-N in groundwater (Wang et al 2011) in the upper river basin have also been studied. However, to date, with the exception of investigations of chemicals in river runoff at the reservoir inflows of the Chao and Bai Rivers (2012), most studies have been conducted in the Miyun Reservoir itself or at the field scale, and there have been relatively few reports of nutrients at the watershed or basin scale. Tang et al. (2011) used distributed hydrological simulations to examine the effect of land use change on TN and TP loads at the inflows of the Chao and Bai Rivers. Their results indicated that the major sources of uncertainty in model simulation may have been the result of the limited input data. Lack of information about nutrient concentrations and routing in the streams and rivers has made it difficult to develop methods for modeling nutrient dynamics in water bodies at different scales in this region.

To address this shortage of data and to develop a simple and rapid method for identifying the key controls on nutrient levels and the potential source areas of nutrients in the upper catchment of the Miyun Reservoir, water samples were collected from river basins at different scales and were analyzed for TN, NO_3^{-} -N, NH_4^{+} -N, TP, and the potassium

permanganate index (COD_{Mn}). In addition, agricultural production data were collected. After examining spatial and seasonal variations in nutrient fluxes and analyzing the factors that may have influenced nutrient concentrations, empirical equations were established to describe the functional relationships between nutrient concentrations and influencing factors. These equations were then used to suggest suitable management options for reducing nutrient loads in different watersheds.

Data and methods

The study area and data collection

The Miyun Reservoir was built in 1960 and is located approximately 85 km north of Beijing. The total storage capacity of the reservoir is about 4.4 billion m³. The reservoir is located at the junction of the Chao and Bai Rivers (Fig. 1) and has a drainage area of 15,788 km². The drainage areas of the Chao and Bai Rivers are 8575 and 6098 km², respectively, and represent 92.9 % of the total reservoir basin area. The annual mean temperature and precipitation in the basin are 7.6 °C and 531.0 mm, respectively. Approximately 70–80 % of the annual precipitation occurs between June and September.

Hydrological data, remote sensing images, and agricultural production data were collected for this study. The hydrological data included daily runoff records from 9 flow-gauging stations and daily precipitation records from 38 precipitation stations for 2008 and 2009 (Fig. 1). The agricultural production data included information on population, cropland area, fertilizer applications, and livestock/poultry numbers of 60 townships in the upper catchment of the Miyun Reservoir.

The administrative boundaries of the townships were based on watershed divides, and so they are closely related to the watershed boundaries; the population, cropland area, fertilizer application, and livestock/ poultry number data obtained from the townships were closely related to individual sub-catchments. Remote sensing images of the upper catchment of the Miyun Reservoir for 2007 with a resolution of 30×30 m were acquired. Land use information was assembled from Landsat ETM/TM images.

Sample collection and analysis

Eleven river sampling points were selected in the upper catchment of the Miyun Reservoir (Table 1). Five sampling points were chosen close to the reservoir inflows of the Bai, Chao, Qingshui, Baimaguan, and Mangniu Rivers to obtain more information about nutrient fluxes in the main rivers entering the Miyun Reservoir. Six sampling points were also located in the upper or middle reaches of the main rivers and their tributaries. The drainage areas of the sampling points ranged from 130 to 8056 km², and so a range of spatial scales was represented. Table 1 shows the watershed characteristics of the sampling points, including the drainage area (*A*), annual precipitation (*P*), population density in the basins (*D*), and the ratio of cropland to the total basin area (R_c).

Water samples were collected over a period of 1 year from July 2008 to June 2009. Samples were collected two times each month during the wet season (from June to September), and every 2 months during the dry season (from October to next May). The water was less than 2.5 m deep at most of the sampling sites, and the water samples were collected from a point 0.5 m below the water surface. Samples were collected from bridges to ensure that they were taken from a representative point in the main flow. Two samples were collected in clean 1-L polyethylene bottles at each site. One of the samples was analyzed unfiltered for TN, TP, and COD_{Mn} . The other sample was filtered through a 0.45-µm millipore filter before analysis for NO3-N and NH_4^+ -N. The filter paper and the retained sediment were dried at 105 °C for at least 6 h and then cooled prior to weighing. The sediment weight was used to calculate the suspended sediment concentration (C_S) . All water samples were acidified to pH <2 with highpurity HCl and then were stored in the freezer in clean polyethylene bottles until analysis (National Environmental Protection Office of China 2002).

Nitrogen (N) and phosphorus (P) concentrations are expressed in mg L⁻¹ of N or P. TN concentrations (C_{TN}) were determined by alkaline potassium persulfate digestion coupled with ultra-violet spectroscopy. NO₃⁻-N concentrations (C_{NO3-N}) were determined by ultraviolet spectrophotometry. NH₄⁺-N concentrations (C_{NH4-N}) were quantified photometrically with Indigo Phenol-Blue. TP concentrations (C_{TP}) were determined by the ammonium molybdate spectrophotometric method. COD_{Mn} is a water quality parameter; its

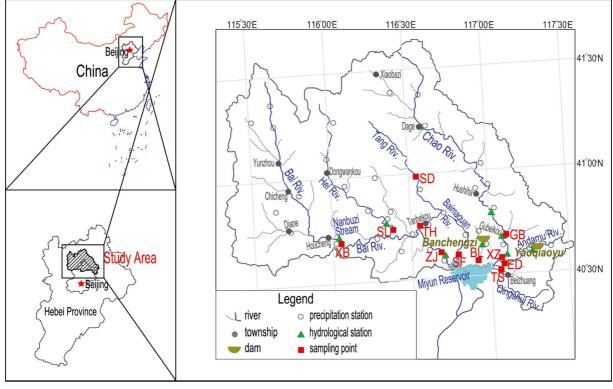


Fig. 1 The location of precipitation and flow measuring stations and river sampling points in the upper catchment of the Miyun Reservoir

concentrations reflect the concentration of oxygendemanding organic pollutants and reducing substances in a sample. Its concentrations (C_{COD}) were determined by the potassium permanganate titration method. $NO_3^{-}-N$, $NH_4^{+}-N$, and nitrite-nitrogen ($NO_2^{-}-N$) are forms of dissolved inorganic nitrogen commonly found in water bodies (Meybeck 1982; Torrecilla et al 2005). Earlier sampling in the Miyun Reservoir (Soil and Water

Table 1	River	sampling	points
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	Sampling Points				Watershed	characters		
No.	Name	Latitude	Longitude	River	A (km ²)	$P(\mathrm{mm})$	$D (\mathrm{km}^{-2})$	Rc (%)
1	Taishitun (TS)	40° 32′ 08.4″ N	117° 06′ 27.1″ E	Qingshui	536	739.0	65	6.11
2	Erdaohe (ED)	40° 34′ 05.7″ N	117° 08′ 47.6″ E	Andamu	368	506.8	63	5.36
3	Gubeikou (GB)	40° 41′ 24.1″ N	117° 09′ 18.5″ E	Chao	5123	494.5	65	8.29
4	Xinzhuang (XZ)	40° 35′ 06.4″ N	117° 07′ 55.6″ E	Chao	6098	493.9	69	8.10
5	Bulaotun (BL)	40° 33′ 53.3″ N	116° 59′ 15.7″ E	Mangniu	130	617.0	72	5.34
6	Shifo (SF)	40° 35′ 09.0″ N	116° 52′ 10.5″ E	Baimaguan	218	586.0	35	1.43
7	Zhangjiafen (ZJ)	40° 37′ 50.3″ N	116° 46′ 09.7″ E	Bai	8506	503.3	43	8.71
8	Tanghekou (TH)	40°43′ 47.4″ N	116° 37′ 59.4″ E	Tang	1257	428.5	32	2.94
9	Sidaoxue (SD)	40°57′ 56.3″ N	116° 37′ 29.1″ E	Tang	673	423.0	28	3.68
10	Shaliangzi (SL)	40°43′ 30.4″ N	116° 28′ 03.8″ E	Hei	1600	556.3	34	8.08
11	Xiabu (XB)	40°41′ 08.3″ N	116° 08′ 15.6″ E	Bai	4040	485.8	57	12.91

Protection Station of Beijing 2004; Xue et al 2012) indicated that NO_2^- -N concentrations were very low and did not contribute significantly to TN loads, so samples were not analyzed for NO_2^- -N concentrations.

Predicting nutrient concentrations from the influencing factors

The monthly flux F_{ij} (kg month⁻¹) of nutrients from the *i*th sampling point in the *j*th month was calculated by

$$F_{ij} = C_{ij} \cdot Q_{ij} \cdot 10^{-3} \tag{1}$$

where C_{ij} (mg L⁻¹) was the measured nutrient concentration for river runoff and Q_{ij} (m³) was the monthly total river runoff from the *i*th sampling point. The nutrient concentrations for the months when no samples were collected were derived by linear interpolation of the nutrient concentrations from adjacent months. The total annual flux (F_i) from the *i*th sampling point was then estimated using Eq. 2:

$$F_{i} = \sum_{j=1}^{12} F_{ij}$$
(2)

The annual mean nutrient concentrations for each sampling point were calculated by averaging the concentrations of all the samples that were collected during the year. The factors that may have influenced the spatial and seasonal variations in nutrient concentrations were divided into two categories, physical factors and human factors. Physical factors included precipitation, runoff, and drainage area characteristics. Human factors included ed population density and land use. Correlation analysis was carried out to determine the relationships between the factors and the nutrient concentrations, and correlations that were significant at the 95 % significance level were identified.

The information about the factors that might influence nutrient fluxes in the rivers was used to establish empirical equations to describe functional relationships between nutrient concentrations and the key controls. Management options to reduce nutrient loads in this region were also discussed.

Results

Spatial and seasonal variations in nutrient concentrations

Spatial variations in nutrient concentrations

Over the 1-year monitoring period, the average precipitation over the upper catchment of the Miyun Reservoir was 519.5 mm, which is very similar to the mean annual value (531.0 mm). Overall, 110 samples were collected from the sampling points during the year. Because of dams in the Banchengzi and Yaoqiaoyu Reservoirs in the upper reaches of the Mangniu and Andamu Rivers, respectively, the Mangniu River was dry at Bulaotun, and only two samples were collected from the Andamu River at Erdaohe. River water samples were collected throughout the year from all the other sampling locations. As a result, we have nutrient concentration data from nine sampling points.

The annual mean nutrient concentrations for the individual sites are shown in Table 2. The annual mean TP and NH_4^+ -N concentrations in the different rivers were similar. However, the average annual mean TN and NO_3^- -N concentrations (5.43 and 3.29 mg L⁻¹) at the two sampling points in the Chao River were higher than those in the other rivers. Further, the annual mean COD_{Mn} concentration (2.52 mg L⁻¹) of the Qingshui and Baimaguan Rivers that drain the area around the reservoir was higher than that of the main rivers.

Table 3 lists the relative contribution of each river to the total nutrient input entering the Miyun Reservoir. The Bai River accounted for 73.4 % of the total runoff, and for $61.3 \sim 73.7$ % (average 70.2 %) of the individual nutrient input to the Miyun Reservoir. The Chao River accounted for 22.7 and 25.0 % of the total runoff and total nutrient input entering the Miyun Reservoir, respectively. The rivers draining the area around the reservoir accounted for only 3.9 % of the runoff and 4.8 % of the total nutrient input that entered the reservoir. The Hei and Tang Rivers, the two major tributaries of the Bai River, accounted for 14.8 and 18.0 % of the total nutrient input, respectively.

Seasonal variations in nutrient concentrations

The mean nutrient concentrations for individual nutrients at the different sampling points during the four different seasons (spring, summer, autumn, winter) are

presented in Table 4. TN concentrations were highest in the autumn and were higher in the autumn and winter than in the other seasons; NH_4^+ -N concentrations were higher in the summer than in the other seasons. Seasonal variation in the NO_3^- -N concentrations was more difficult to identify.

TP and COD_{Mn} concentrations were highest in summer, when flows and suspended sediment concentrations were highest, and gradually decreased in the autumn and winter. Suspended sediment dynamics and the influence of residence time on organic matter concentrations may be important in explaining these trends and are considered further in the "Empirical equations associated with physical and human factors that may influence the variations in nutrient concentrations" section.

Empirical equations associated with physical and human factors that may influence the variations in nutrient concentrations

Influences on spatial variations in nutrient concentrations

Table 5 shows correlations between the annual mean nutrient concentrations and the influencing factors. The annual precipitation varied significantly between the sampled watersheds and ranged from 423 to 739 mm. However, the relationships between the annual mean nutrient concentrations and annual precipitation or annual runoff were not statistically significant. Differences in catchment characteristics and the magnitude of the suspended sediment concentrations in the rivers were more important. Table 5 indicates that catchment area (*A*) was the only controlling factor that was significantly correlated with the annual mean COD_{Mn} concentration ($C_{COD(a)}$). This was an inverse relationship (Fig. 2a), as follows:

$$C_{COD(a)} = 4.677 A^{-0.098}, r^2$$

= 0.656 (significant at the 0.01 level, n = 9).
(3)

This inverse relationship most likely reflects the fact that the main sources of COD_{Mn} were located in smaller catchments, and the oxygen demand progressively declined with residence time because of oxidation or deposition of particulate organic matter as the pollutants travelled downstream. The r^2 for this relationship was higher than those for the other quadratic function curves

Table 2 Annual mean nutrient concentrations at the different sampling points

Sampling points Site	Site	XB	SL	HT	SD	ZJ	SF	GB	XZ	TS	Average
	River	Bai	Hei	Tang	Tang	Bai	Baimaguan	Chao	Chao	Qingshui	Í
	Drainage area (km ²⁾	4040	1680	1257	673	8506	228	5123	8609	536	15788
Annual mean	TN	4.37 ± 0.94	3.62 ± 1.23	2.81 ± 0.67	2.28 ± 1.49	2.85 ± 1.08	3.95 ± 1.98	5.47±1.88	5.43 ± 2.32	3.32 ± 1.51	3.79 ± 1.49
concentration	NO ^{3-N}	2.97±0.77	2.70 ± 0.91	$2.08 {\pm} 0.48$	1.31 ± 0.45	$2.06 {\pm} 0.72$	2.85 ± 1.38	$3.79 {\pm} 1.10$	$3.29 {\pm} 0.87$	2.37±1.29	$2.60 {\pm} 0.89$
(mg L ⁻¹)	NH_4^+-N	$0.169{\pm}0.082$	0.169 ± 0.082 0.138 ± 0.069	$0.147 {\pm} 0.059$	0.121 ± 0.070 0.111 ± 0.042		0.119 ± 0.042	0.119 ± 0.042 0.130 ± 0.048	0.115 ± 0.047	0.119 ± 0.048	$0.130 {\pm} 0.056$
	TP	0.043 ± 0.026	0.043 ± 0.026 0.030 ± 0.022		0.020 ± 0.014 0.024 ± 0.018	0.025 ± 0.017	0.022 ± 0.018	0.034 ± 0.026	0.026 ± 0.020	0.032 ± 0.026	0.028 ± 0.021
	COD_{Mn}	2.30 ± 1.04 2.50 ± 1.21	2.50 ± 1.21	2.10 ± 1.20	$2.10\pm1.20 \qquad 2.76\pm1.95 \qquad 1.93\pm0.79$	1.93 ± 0.79	2.74 ± 1.40	2.00 ± 0.93 1.81 ± 0.93		$2.29 {\pm} 0.85$	2.27 ± 1.13

Table 3 The contribution of the individual rivers to the nutrient input to the Miyun Reservoir

River ^a	Nutrient	load (t a^{-1})				Relative contribution of each river to the total (%)				
	TN	NO ₃ ⁻ -N	NH4 ⁺ -N	ТР	COD _{Mn}	TN	NO ₃ ⁻ -N	NH4 ⁺ -N	ТР	COD _{Mn}
Hei ^b	226.0	192.2	8.5	1.97	170.3	13.8	15.9	12.0	15.1	17.2
Tang ^b	240.3	192.1	12.3	2.29	224.6	13.8	23.0	12.8	17.6	22.7
Bai	1150.4	922.6	38.7	9.43	729.0	66.0	72.6	61.3	72.4	73.7
Baimaguan	52.6	44.5	1.5	0.30	38.1	3.2	2.7	2.8	2.3	3.9
Chao	646.9	407.2	12.2	3.04	203.5	29.1	22.9	34.4	23.3	20.6
Qingshui	28.4	24.1	0.9	0.25	18.9	1.7	1.8	1.5	1.9	1.9
Nutrients entered Miyun reservoir	1878.2	1398.3	53.3	13.02	989.6	100.0	100.0	100.0	100.0	100.0

^a The total nutrient fluxes entering the Miyun Reservior represent the sum of the inputs from the Bai, Chao, Baimaguan, and Qingshui Rivers. The Mangniu River was dry at Bulaotun because of the dams located in the upper reaches

^b The Hei and Tang Rivers are tributaries of the Bai River and their contribution is included in the total for the Bai River

or linear equations, so a power function can be used to describe the relationship between $C_{COD(a)}$ and A.

Considerable amounts of P are transported in association with suspended sediment (Ellison and Brett 2006; Berretta and Sansalone 2011; Rodríguez-Blanco et al 2013). Relative to other land use types, cropland is the main source of sediments and nutrients in the study area (Li et al 2013) because of frequent disturbances of the land surface and the relatively low vegetation cover (Miao et al 2012). Therefore, TP concentrations were higher in catchments that had a higher proportion of cultivated land than in those with less cultivated land. Equation 4 demonstrates that the annual mean TP concentrations ($C_{TP(a)}$) increased as both the annual mean suspended sediment concentrations ($C_{S(a)}$, g L⁻¹) and the ratio of the cropland area to the total basin area (R_C , %) increased:

$$C_{TP(a)} = 0.010C_{S(a)} + 0.0012R_C + 0.019, r^2$$

= 0.748 (significant at the 0.01 level, n = 9). (4)

Figure 2b shows that the annual mean TN concentration ($C_{TN(a)}$) increased as the population in the watershed (D) increased.

$$C_{TN(a)} = 0.051D + 1.339, r^2$$

= 0.558 (significant at the 0.05 level, $n = 9$). (5)

The variation in the annual mean NO₃⁻N concentrations was also closely related to *D*. Concentrations of Nrelated pollutants are generally elevated in areas of high population density. Large towns with higher population densities are mainly located in the lower reaches of the rivers; these places have greater amounts of garbage, domestic sewage, and excreta from livestock and poultry.

Factors influencing seasonal variations in nutrient concentrations

Daily mean river flow data (Q) was available for five sampling points (Table 6). C_{COD} was significantly correlated with Q at the 99 % level at three of these sampling points and at the 95 % level at the other two points. At the three sampling points where C_S concentrations were appreciable and where there was substantial variation between the samples, C_{COD} was only significantly correlated with C_S at the 95 % level at sites GB and SL. The average correlation coefficient between C_{COD} and Q was greater than that between C_{COD} and C_{S} . The increases of C_{COD} relative to Q differed between the sampling points (Fig. 3). C_{COD} increased at a slower rate at ZJ and XZ than at XB, GB, and SL. ZJ and XZ were at the outlets of the Bai and Chao Rivers, respectively, and the annual mean COD_{Mn} concentrations at these sites were lower than at the sampling points in the upper branches or tributaries.

Suspended sediment concentrations in the water samples (C_S) had an influence on seasonal variations in the TP concentrations. TP concentrations in the water samples (C_{TP}) were significantly correlated with C_S at XB, GB, and SL, where C_S varied widely

Table 4	easonal van	ation in nutrient	Table 4 Seasonal variation in nutrient concentrations (mg L^{-1}) at the different sampling points	mg L^{-1}) at the di-	fferent sampling	points					
Season	Nutrients	Sampling points	its								Average
		XB	SL	TH	SD	ZJ	SF	GB	ZX	TS	
Spring	NT	4.25 ±0.73	3.05 ± 0.38	5.15±0.18	4.77±0.48	1.78 ± 0.64	2.87±0.45	2.91 ± 0.23	3.17 ± 0.17	2.98±0.42	3.44±1.12
)	$NO_{3}^{-}N$	$3.35 {\pm} 0.87$	2.28 ± 1.49	4.45 ± 0.10	3.76 ± 0.25	$1.51 {\pm} 0.66$	2.68 ± 1.04	2.44 ± 1.29	2.60 ± 1.20	1.34 ± 0.19	2.71 ± 1.44
	NH4 ⁺ -N	0.115 ± 0.016	$0.089 {\pm} 0.075$	0.101 ± 0.007	0.101 ± 0.057	$0.105 {\pm} 0.089$	$0.096 {\pm} 0.018$	0.098 ± 0.079	0.058 ± 0.056	0.045 ± 0.006	0.090 ± 0.050
	TP	$0.029 {\pm} 0.001$	$0.017 {\pm} 0.007$	$0.020 {\pm} 0.007$	0.017 ± 0.004	$0.015 {\pm} 0.007$	0.017 ± 0.001	0.017 ± 0.011	0.006 ± 0.006	$0.018{\pm}0.003$	$0.017 {\pm} 0.007$
	$\mathrm{COD}_{\mathrm{Mn}}$	$2.15 {\pm} 0.18$	1.75 ± 0.34	2.07 ± 0.23	$1.56 {\pm} 0.27$	$1.36 {\pm} 0.59$	$1.25 {\pm} 0.88$	$1.48 {\pm} 0.70$	1.02 ± 0.32	1.49 ± 0.21	$1.57 {\pm} 0.46$
Summer	NT	$4.46 {\pm} 0.69$	$2.69 {\pm} 0.51$	$4.70 {\pm} 0.58$	4.48 ± 0.42	$1.71 {\pm} 0.65$	$2.60{\pm}2.84$	3.26 ± 1.38	3.31 ± 1.82	2.64 ± 2.37	3.32 ± 1.65
	$NO_{3}^{-}NO_{3}$	$3.27 {\pm} 0.70$	$1.86 {\pm} 0.90$	3.75 ± 0.45	3.15 ± 0.52	1.35 ± 0.73	$1.90 {\pm} 1.87$	$2.76 {\pm} 0.65$	2.51 ± 0.98	2.33 ± 2.27	2.54 ± 1.12
	NH4 ⁺ -N	0.193 ± 0.113	0.125 ± 0.090	0.155 ± 0.073	0.128 ± 0.045	0.119 ± 0.044	0.142 ± 0.046	$0.154 {\pm} 0.042$	0.129 ± 0.037	0.136 ± 0.024	0.142 ± 0.066
	TP	0.049 ± 0.036	0.029 ± 0.031	0.037 ± 0.022	0.029 ± 0.026	0.027 ± 0.023	0.023 ± 0.025	0.036 ± 0.029	0.029 ± 0.019	0.050 ± 0.032	$0.034 {\pm} 0.028$
	$\mathrm{COD}_{\mathrm{Mn}}$	$2.60 {\pm} 0.99$	2.23 ± 1.70	2.31 ± 1.41	2.02 ± 1.74	$3.30 {\pm} 0.92$	2.44 ± 0.89	2.69 ± 1.27	$3.57 {\pm} 0.53$	2.87 ± 0.88	2.67 ± 1.22
Autumn	IN	5.02 ± 0.95	3.99 ± 0.44	7.63 ± 0.44	$8.66 {\pm} 3.19$	$3.34{\pm}1.34$	$3.68 {\pm} 1.07$	5.71 ± 2.65	5.65±2.06	$4.66 {\pm} 0.41$	5.37±1.78
	$NO_{3}^{-}NO_{3}$	$2.18 {\pm} 0.56$	$2.60 {\pm} 0.30$	$3.82 {\pm} 0.19$	$3.48 {\pm} 0.09$	1.03 ± 0.28	2.42 ± 0.38	$3.19 {\pm} 0.02$	$3.94 {\pm} 0.20$	2.33 ± 0.32	2.78 ± 0.93
	NH4 ⁺ -N	$0.166 {\pm} 0.031$	0.115 ± 0.057	0.125 ± 0.075	0.122 ± 0.127	0.125 ± 0.009	$0.159 {\pm} 0.015$	0.143 ± 0.047	$0.146 {\pm} 0.062$	0.127 ± 0.046	0.136 ± 0.055
	TP	0.032 ± 0.009	0.018 ± 0.003	$0.039 {\pm} 0.001$	0.028 ± 0.003	$0.020 {\pm} 0.003$	$0.019 {\pm} 0.001$	0.020 ± 0.005	0.014 ± 0.013	0.013 ± 0.007	0.023 ± 0.011
	$\mathrm{COD}_{\mathrm{Mn}}$	$1.98{\pm}1.18$	$1.55 {\pm} 0.29$	1.75 ± 1.29	1.82 ± 2.90	$2.76 {\pm} 0.48$	$1.89 {\pm} 1.70$	$2.26 {\pm} 0.78$	2.51 ± 1.46	1.79 ± 0.44	2.04 ± 1.34
Winter	IN	4.91 ± 2.07	3.22 ± 3.48	6.27 ± 0.96	7.43 ± 0.95	2.52 ± 1.06	$3.60{\pm}2.71$	4.80 ± 3.94	4.84 ± 4.31	4.55 ± 1.72	4.68 ± 2.54
	$NO_{3}^{-}NO_{3}$	1.83 ± 0.76	$1.80 {\pm} 0.60$	$2.89 {\pm} 0.73$	$3.10 {\pm} 0.38$	0.72 ± 0.81	2.25 ± 1.45	2.27±0.44	2.91 ± 0.24	2.44 ± 1.19	2.25 ± 0.93
	NH4 ⁺ -N	$0.108{\pm}0.057$	0.082 ± 0.008	0.082 ± 0.003	0.091 ± 0.007	0.065 ± 0.032	$0.142 {\pm} 0.069$	0.108 ± 0.020	$0.100 {\pm} 0.020$	0.078 ± 0.014	0.095 ± 0.033
	TP	0.025 ± 0.004	0.008 ± 0.011	$0.016 {\pm} 0.004$	0.005 ± 0.004	$0.008 {\pm} 0.003$	$0.008 {\pm} 0.007$	0.013 ± 0.016	0.005 ± 0.001	0.008 ± 0.010	0.011 ± 0.009
	$\mathrm{COD}_{\mathrm{Min}}$	0.91 ± 0.17	$0.90 {\pm} 0.27$	$1.08 {\pm} 0.15$	$1.00 {\pm} 0.26$	$1.24 {\pm} 0.18$	1.01 ± 0.12	$1.86 {\pm} 0.04$	$1.26 {\pm} 0.27$	$1.26 {\pm} 0.68$	1.17 ± 0.36
Data are sl August 1,	nown in "ave and August	rage±standard d 16, 2008; June 1	Data are shown in "average±standard deviation." Sampling dates are in the following different seasons: two times in spring, April 1 and May 31, 2009; five times in summer, July 1, July 16, August 1, and August 16, 2008; June 16, 2009; three times in autumn, September 16, and October 1, 2008; two times in winter, December 1, 2008 and February 1, 2009	ing dates are in the nes in autumn, S	le following diffe eptember 1, Sep	erent seasons: tw tember 16, and 0	o times in spring, October 1, 2008;	April 1 and May two times in wir	31, 2009; five tin nter, December 1	mes in summer, J , 2008 and Febr	uly 1, July 16, uary 1, 2009

 Table 5
 Correlations between annual mean nutrient concentrations and key influencing factors

Influencing factors	Annual mea	Annual mean nutrient concentrations (mg L^{-1})							
	TN	NO ₃ ⁻ -N	NH4 ⁺ -N	TP	CODMn				
Annual precipitation (P)	0.053	0.159	-0.281	0.210	0.136				
Annual river runoff (Ry)	-0.082	-0.080	-0.174	-0.057	-0.590				
Drainage area (A)	0.385	0.329	-0.158	0.178	-0.792**				
Annual mean suspended sediment concentrations $(Cs(a))$	0.531	0.618	0.522	0.779*	-0.190				
Density of people in the basins (D)	0.747*	0.686*	-0.041	0.580	-0.624				
The ratio of cropland area to total basin Area (R_C)	0.440	0.425	0.426	0.826**	-0.473				

*Correlation is significant at the 0.05 level (two-tailed); **correlation is significant at the 0.01 level (two-tailed)

(Table 6); C_{TP} also increased as Q increased at the three sampling points:

$$C_{TP} = 0.005C_S + 0.004Q + 0.015, r^2$$

= 0.629 (significant at the 0.01 level, n = 36). (6)

When a river's flow rate is high, the river has a greater ability to transport sediments. Increased concentrations of suspended particles provide additional potential for P delivery. Dam and reservoir construction on streams and rivers may result in decreased suspended sediment loads in a water body (Miao et al 2011). However, sediment stored in the bottom of a reservoir can be re-suspended when the flow rate is high and endogenous P may be released into the water (Chung et al 2009).

N is highly soluble and exists in different forms in water bodies, including NO_3^- -N and NH_4^+ -N. Thus, N is likely to be delivered to the watershed outlets in its dissolved form, without suspended sediments or organic particles. TN, NO_3^- -N, and NH_4^+ -N concentrations were not statistically correlated with Q or C_S (Table 6).

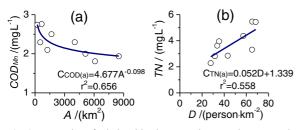


Fig. 2 Examples of relationships between the annual mean nutrient concentrations and key influencing factors

Discussion

Potential nutrient sources

Potential terrestrial sources of nutrients in the upper catchment of the Miyun Reservoir include industrial wastewater, domestic sewage, livestock/poultry breeding, and fertilizer applications (Table 7). The amount of nutrients that originated from domestic sewage, livestock/poultry breeding, and fertilizer applications was calculated from the information about population, fertilizer applications, and livestock/ poultry numbers for each township in the study area. Quantities of industrial wastewater discharged annually were reported by the Soil and Water Protection Station of Beijing (2004).

Since the 1980s, industrial development has been restricted in the upper catchment of the Miyun Reservoir to protect water quality. Approximately 4362 t of TN and 344 t of TP were derived from industrial wastewater each year, which represented only 9.1 and 4.5 % of the total TN and TP loads from the upper catchment, respectively. Further, only 4.7 % of the total TN loads and 5.8 % of the total TP loads were from domestic sewage (Table 7). As sewage treatment facilities are constructed in the main townships in the lower reaches of the Bai and Chao Rivers, discharges of domestic sewage are expected to reduce in the future; domestic and industrial point sources will eventually be a minor proportion of the total nutrient load in the study area.

Profits from crop cultivation, fruit farming, and livestock and poultry breeding are the main source of income for the local inhabitants. Since the late 1980s, as the population has increased, higher crop yields have

Sampling	g points	Correlatio	n with Q				Correlatio	n with C_S			
Name	River	NO ₃ ⁻ -N	NH4 ⁺ -N	TN	ТР	COD _{Mn}	NO ₃ ⁻ -N	NH4 ⁺ -N	TN	ТР	COD _{Mn}
XB	Bai	-0.006	-0.252	-0.512	0.704**	0.769**	-0.094	-0.238	-0.219	0.698**	0.379
ZJ	Bai	0.417	-0.049	0.178	0.630*	0.890**	а	a	a	а	а
GB	Chao	-0.015	0.445	0.086	0.739**	0.678*	-0.165	0.417	-0.565	0.833**	0.684*
XZ	Chao	-0.002	0.163	-0.167	0.720**	0.576*	а	а	а	а	а
SL	Hei	0.320	-0.032	-0.055	0.515	0.72**	0.095	-0.066	-0.097	0.793**	0.608*

Table 6 Correlations between nutrient concentrations and (a) the daily mean river flow (Q) and (b) suspended sediment concentrations (C_S) for the five sampling stations with flow records

*Correlation is significant at the 0.05 level (two-tailed)

** correlation is significant at the 0.01 level (two-tailed)

^a Cs was very low. As an independent variable, the range of variation was limited, and a correlation analysis was not conducted

been achieved by increased fertilizer applications. Fertilizer applications represent 28.6 and 10.0 % of the total TN and TP loads in the study area, respectively. Since the early 1990s, the local government has encouraged development of animal husbandry, meaning that livestock and poultry numbers have increased rapidly in the study area. Using information on livestock numbers and the typical TN and TP loads in excrement from an individual unit of livestock or poultry (Supplementary Table S1), annual TN and TP loads associated with excrement have been estimated at 57.6 and 79.7 % of the total TN and TP loads, respectively.

The annual TN and TP inputs entering the Miyun Reservoir from the rivers were 1878.2 and 13.02 t and only accounted for 3.91 and 0.17 % of the total TN and TP loads from the upper catchment of the Miyun Reservoir, respectively. The runoff depth in the upper catchment of the Miyun Reservoir was only 29.8 mm during the monitoring period,

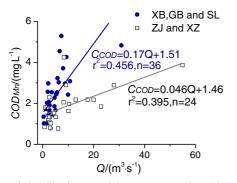


Fig. 3 Relationships between COD_{Mn} concentrations (C_{COD}) and discharge (Q)

meaning that the runoff coefficient was 0.057. Ma et al. (2010) estimated that the impact of human activities, including direct withdrawal of water, accounted for 23 % of the decrease in water flowing into the Miyun Reservoir, while the indirect impact of human activities through land use and vegetation change accounted for 18 % of the decrease. Other studies have reported increased in-stream TN retention with decreased stream water discharge (Lu et al. 2013a, b); therefore, the amount of TN and soluble nutrients that are transported downstream may decrease as the stream flow in the study area decreases. Implementation of soil-water conservation measures has effectively reduced the downstream transport of eroded soil particles. The suspended sediment concentrations in both the Chao and Bai Rivers have decreased dramatically since 1990 (Fig. 4a). The decrease in the amount of suspended sediment may result in significant reductions in downstream transport of sediment-associated P.

In this study, nutrient concentrations were not monitored for most of the storm runoff events. River flow and sediment transport are significant in storm events, so nutrient inputs from rivers to the Miyun Reservoir may be underestimated, especially the TP load. At the annual scale, most P is exported in a few runoff events that account for a small percentage of the water volume (Bowes et al 2009). For example, in a mixed land use catchment in Galicia (NW Spain), 35 % of particulate phosphorus load and 19 % of dissolved phosphorus load occurred in only 2 % of the time over a whole year (Rodríguez-Blanco et al 2013). **Table 7**The contribution of in-dividual sources to the terrestrialnutrient loads of the upper catch-ment of the Miyun Reservoir

Potential nutrient sources	Nutrient loa	ds (t a^{-1})	Relative cor source to the	ntribution of each e total (%)
	TN	ТР	TN	TP
Industrial wastewater	4362.0	344.0	9.1	4.5
Domestic sewage	2246.3	449.3	4.7	5.8
Livestock and poultry breeding	27,718.4	6139.8	57.6	79.7
Fertilizer application	13,757.8	767.1	28.6	10.0
Total	48,084.5	7700.2	100.0	100.0

Options for improving water quality

Based on the empirical equations derived in this study and the analysis of potential nutrient sources, suggestions have been made about how to reduce nutrient loads in the different watersheds. To estimate interannual dynamics in nutrient concentrations, historical nutrient data were collected for the outflows of the Chao and Bai Rivers (Xue et al 2012). The same calculation methods were used in this study and the earlier study, and annual nutrient concentrations are the average of the monthly data (Fig. 4).

Transport in association with sediment plays an important role in P delivery in the study area (Eqs. 4 and 6), and cropland is the main source of sediment and nutrients (Li et al 2013). Therefore, the P load could be reduced if soil loss from cropland was reduced. Since the 1980s, soil conservation practices, including returning farmland to forestland, constructing stone-faced terraces, and tree planting in fish-scale pits, have been progressively introduced in the upper catchment of the Miyun Reservoir. Consequently, sediment concentrations in the streams and rivers have gradually decreased. There is a weak linear decreasing trend in TP concentrations (Fig. 4b, significance level p > 0.1),

which demonstrates the effectiveness of soil conservation practices.

The ratio of cropland to the total area is still relatively high within the upper reaches of the Chao and Bai Rivers (Fig. 5a), where the high risk of soil erosion could result in increased suspended sediment concentrations in streams. Soil conservation practices will significantly reduce P concentrations in the streams, especially in the townships with relatively high R_C values.

Population increases may result in increased N loads and elevated TN concentrations in water bodies (Eq. 5). From 1990 to 2009, the population of the upper river basin of the Miyun Reservoir increased by 11.5 %, and TN concentrations also showed a significant increasing linear trend (significance level p=0.01) in both the Chao and Bai Rivers (Fig. 4c). Livestock/poultry husbandry and fertilizer applications were the main sources of N pollutants. Where population densities are high, such as in the Chao and Oingshui Basins (Fig. 5b), most livestock excrement is spread on fields as manure. Annual TN loads associated with excrement (E_{TN}) generated by each township have been estimated from livestock numbers and the typical TN loads of excrement from an individual unit of livestock or poultry (Supplementary Table S1). Calculated TN loads are positively related

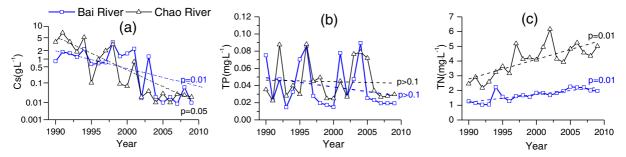


Fig. 4 Linear trends in the annual **a** suspended sediment, **b** TP, and **c** TN concentrations at the outflows of the Chao and Bai Rivers (data were provided by Xue et al. (2012))

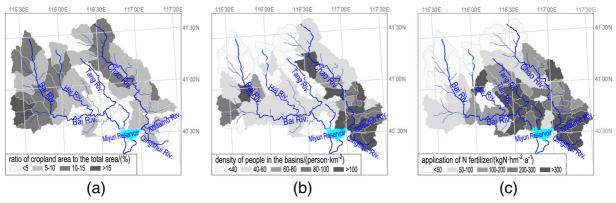


Fig. 5 Spatial distribution of the a ratio of cropland area to the total area, b population density, and c N fertilizer applications

with population (P_O) (Fig. 6). Polluted water from livestock or poultry husbandry can be treated by sewage treatment equipment, which would improve the water supply and drainage systems. Practical technologies, such as biogas fermentation and the organic fertilizer products, can also be used to treat excrement.

N fertilizer applications to cropland are relatively high in the townships on the north shore of the Miyun Reservoir and in the Qingshui, Andamu, Tang, and Tian River basins, which are in the lower reaches of the Chao and Bai River basins (Fig. 5c). Excessive N fertilizer applications may result in higher groundwater NO₃⁻-N concentrations in this region (Wang et al 2011) that could directly threaten the water quality of the reservoirs, especially in the areas that border the Miyun Reservoir. N losses in these watersheds could be significantly reduced by using fertilizer management techniques, such as split applications (Richardson et al 2008).

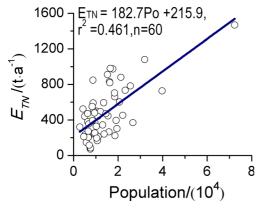


Fig. 6 The relationship between annual TN loads from livestock and poultry excrement (E_{TN}) and the population (P_O) of each township

The population densities and N fertilizer applications are higher in the Qingshui River Basin and the lower reaches of the Chao River Basin, so pollutant inputs to the rivers should be higher than in other basins. Natural riparian buffers and constructed vegetated buffers could effectively reduce downstream nutrient delivery from cropland near the rivers (Supplementary Fig. S1) (Richardson et al, 2008). Vegetation has relatively high dissolved N requirements, but lower dissolved P requirements (Mengel and Kirkby 1982; Dosskey et al 2010). N uptake rates by forest and herbaceous vegetation in N-enriched riparian zones of up to 170 kg N ha⁻¹ year⁻¹ have been reported and accounted for the major proportion of the total nutrient load input to these riparian zones (Peterjohn and Correll 1984; Hefting et al. 2005). Restoration of natural wetlands and construction of further wetlands at the points where the Chao and Qingshui Rivers enter the Miyun Reservoir could reduce nutrient transport into the reservoir.

Applicability and limitations of the empirical equations

Equations 3-6 offer a simple, robust, and rapid method for identifying the key controls on nutrient levels and should help to identify both the nutrient sources and the areas of a catchment that should be targeted by mitigation measures. This method does not require complex software with user interfaces, or detailed information about the study area.

Eutrophication is a nationwide problem that has threatened public health in China for some time, so nutrient load reduction strategies should be enforced as soon as possible. Empirical models may be used to calculate nutrient loads or concentrations in watersheds and basins where little data is available. The export coefficient model (ECM) was recently used to calculate watershed (Lu et al. 2013b) or basin-scale (Ma et al 2011) nutrient loads in China. While the methodology is easy to use, the export coefficients are inherently highly variable. Moreover, the choice of an export coefficient from the literature is inevitably subject to considerable uncertainty (Wu et al 2012). Chen et al. (2013) used a modified load apportionment model to identify point and diffuse source nutrient inputs to the Changle River, while their study did not successfully identify the factors that might have influenced nutrient variations. Specific knowledge on pollutants and their controls should be used to guide the choice of nutrient load reduction strategies in the early stages of the eutrophication process. Statistical relationships should be established between nutrient concentrations and loads, and natural and human-related factors.

The equations developed in our study were based on a limited set of environmental and physical factors, which simplified the approach for grouping physical processes and provided generalized response estimates. These simplified techniques, because of their inherent generalizations, have limited descriptions of the processes that are associated with pollutant movement, transport, transformation, and delivery that may influence seasonal variations in nutrient concentrations. TN concentrations were highest in the autumn (Table 4), which begins at the end of the wet season. This suggests that the water supplied to the rivers in the autumn is mainly derived from shallow groundwater. This shallow groundwater passes slowly through the soil and subsurface zone, and its TN concentration increases because of mobilization of synthetic N fertilizer and decomposition of soil organic matter. Furthermore, river flows are relatively stable in the autumn, which means that it is convenient for livestock and poultry watering. The combination of the relatively low flow rates, the limited potential for dilution, and the excrement inputs from livestock most likely resulted in increased TN concentrations. In addition, elevated TN concentrations in the autumn and winter may reflect the lower temperatures, which minimize biological uptake (Panno et al 2008). The reasons for the seasonal variation in the concentrations of the different forms of N varied between the determinants considered (Neitsch et al 2005). Variations in N forms during the summer wet season reflected inputs of eroded soil particles to the rivers and nitrification in the groundwater that supplied the rivers. After the summer, NH_4^+ -N concentrations gradually decreased (Table 4). However, seasonal variation in NO_3^- -N concentrations was more complex.

Equation 6 reflects the relationship between TP concentrations and river flow. The sampling interval must be sufficiently short so that a representative range of storm events is captured at the study sites. The twicemonthly monitoring during the wet season may not have been sufficient to adequately characterize the high-range TP concentration-flow relationships. Inclusion of sampling data for high-flow events at monitoring sites would increase the robustness of the empirical equations.

Conclusions

In the upper catchment of the Miyun Reservoir, sedimentassociated P exerts an important influence on seasonal and spatial variations in TP concentrations. Annual mean TP concentrations ($C_{TP(a)}$) were well correlated with annual mean suspended sediment concentrations ($C_{S(a)}$) and the ratio of the cropland area to the total basin area (Rc) as follows: $C_{TP(a)}=0.010C_{S(a)}+0.0012R_{C}+0.019$ ($r^{2}=0.748$, significance level p=0.01). C_{TP} increased as C_{S} and river flow (Q) increased: $C_{TP}=0.005C_{S}+$ 0.004Q+0.015 ($r^{2}=0.629$, significance level p=0.01).

There was a linear relationship between the annual mean TN concentrations $(C_{TN(a)})$ and the population density (*D*) of the basins as follows: $C_{TN(a)}=0.051D+1.339$ ($r^2=0.558$, significance level p=0.05). Organic compounds and reducing substances in streams and rivers are likely to be deposited or oxidized when transported over longer distances. As such, the annual mean COD_{Mn} concentration ($C_{COD(a)}$) was well correlated with the drainage area (*A*) as follows: $C_{COD(a)}=4.677A^{-0.098}$ ($r^2=0.656$, significance level p=0.01).

Soil conservation will most likely play an important role in reducing TP concentrations in the upper reaches of the Chao and Bai Rivers. To reduce N in the study area, (1) natural riparian buffers and vegetated buffers could be constructed on cropland close to the channels in the Chao and Qingshui River Basins, (2) fertilizer management plans could be used to support reduction of N fertilizer applications in the areas surrounding the Miyun Reservoir, and (3) natural wetlands could be restored and further wetlands could be constructed at the inlets of the Chao and Qingshui Rivers to the Miyun Reservoir. Acknowledgments Funding for this research was provided by "the National Natural Science Foundation of China (No. 41401560) and National Basic Research Program of China (2007CB407203)." We are very grateful to Des Walling for insightful comments and help in modifying the English language of the manuscript.

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