



Source apportionment of nitrogen and phosphorus from non-point source pollution in Nansi Lake Basin, China

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Abstract

Nitrogen (N) and phosphorus (P) from non-point source (NPS) pollution in Nansi Lake Basin greatly influenced the water quality of Nansi Lake, which is the determinant factor for the success of East Route of South-North Water Transfer Project in China. This research improved Johnes export coefficient model (ECM) by developing a method to determine the export coefficients of different land use types based on the hydrological and water quality data. Taking NPS total nitrogen (TN) and total phosphorus (TP) as the study objects, this study estimated the contributions of different pollution sources and analyzed their spatial distributions based on the improved ECM. The results underlined that the method for obtaining output coefficients of land use types using hydrology and water quality data is feasible and accurate, and is suitable for the study of NPS pollution at large-scale basins. The average output structure of NPS TN from land use, rural breeding and rural life is 33.6, 25.9, and 40.5%, and the NPS TP is 31.6, 43.7, and 24.7%, respectively. Especially, dry land was the main land use source for both NPS TN and TP pollution, with the contributed proportions of 81.3 and 81.8% respectively. The counties of Zaozhuang, Tengzhou, Caoxian, Yuncheng, and Shanxian had higher contribution rates and the counties of Dingtao, Juancheng, and Caoxian had the higher load intensities for both NPS TN and TP pollution. The results of this study allowed for an improvement in the understanding of the pollution source contribution and enabled researchers and planners to focus on the most important sources and regions of NPS pollution.

Keywords Source apportionment · Non-point source pollution · Total nitrogen and phosphorus · Improved export coefficient model · Nansi Lake Basin

Introduction

Water crisis is one of the most serious problems that China is facing in the twenty-first century. This water crisis is reflected

not only by the lack of water quantity but also by the deterioration of water quality (Xia et al. 2012). In recent years, non-point source (NPS) pollution has become the major threat to quality of regional water resources as the point source (PS) pollution being controlled effectively (Chang et al. 2004; Min and Kong 2016). Nitrogen (N) and phosphorus (P) are main pollutants (Fu et al. 2015) and important indicators of water pollution monitoring in China. Large amounts of N and P in aquatic environments can cause a wide range of problems such as toxic algal blooms, oxygen depletion, and loss of biodiversity, which finally can degrade aquatic ecosystems and the quality of water used for drinking, industry, agriculture, recreation, and other purposes (Farenga and Daniel 2007; Ding et al. 2010; Ongley et al. 2010; Sun et al. 2012). Currently, more than 60% lakes in China are suffering different degrees of eutrophication, and over 50% of the problems are caused by NPS N and P pollution (Zhao et al. 2010; Xu et al. 2013; Ma et al. 2015). The recognition and control of NPS pollution, especially N and P loss, is the basic scientific problem and is of great scientific significance for improving the water environment of China.

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NPS pollution has the characteristics of randomness, universality, and uncertainty (Geng et al. 2012), and model simulations are common methods for its monitoring and assessment. Existing models can be classified into physical-based models and empirical models. Since 1970, various physical-based models have been developed to simulate nutrient transport based on hydrological processes. The represented models include ANSWERS (Beasley et al. 1980), SWRRB (Williams et al. 1985), SWAT (Arnold et al. 1993), HSPF (Bicknell et al. 2001), SPARROW (Schwarz et al. 2006), AGNPS (Yong et al. 1989), and BASINS (Whittemore 1998), etc. Although these models can provide acceptable simulation results compared to observed data (such as flow, N, and P loss), the values of many parameters cannot be obtained from field observation. Instead, these values must be determined through model calibration. Among empirical models, the export coefficient model (ECM) has widely been used in the study of NPS pollution at large-scale basins (Huang et al. 2012a; Zhang et al. 2014; Zhou et al. 2014). The initial ECM (Johnes, 1996) was built on the idea that the nutrient loads exported from a watershed is the sum of the losses from individual sources, such as land use, livestock, and rural living. In recent years, the ECM has been greatly improved in term of parameters and application ranges (Zhu et al. 2015). For example, transmission coefficient (Soranno et al. 1996; Xu et al. 2015), rainfall influence coefficient, watershed loss coefficient (Cai et al. 2004; Ren et al. 2015), and terrain factors (Noto et al. 2008; Zhu and Chang, 2008) were introduced to consider the influence of nutrient “source-sink” distance: the hydrological impacts and the pollutant loss in transport process and the influence of terrain. These studies not only promoted the application of ECM in NPS pollution load estimation but also laid the foundation for the estimation of NPS pollution output for the regions where lacking monitoring data. Export coefficients in the studies mentioned above are usually derived from literature and field experiments to determine the rate at which nutrients are lost from each identifiable source to the surface drainage network. Generally, the export coefficients of rural living and livestock can be determined from literature reviewing, because these coefficients do not vary greatly among areas (Yuan et al. 2017). However, export coefficients for different land use types cannot be taken from literatures because the reported values have generally been obtained in specific conditions. These values can only reflect features specific to those conditions and cannot provide sufficient accuracy when introduced to other conditions. Thus, field monitoring at the catchment outflow must be conducted to establish the nutrient loads annually transported into the target water body.

Nansi Lake is the largest freshwater lake of North China, and is also the pivot for the East Route of South-North Water

Transfer Project (ERSNWTP). It is important to note that Nansi Lake is the largest drink water source of Shandong province, and pumping stations in the Nansi Lake will be used to control water storage and water transfer of ERSNWTP. Therefore, the water quality of the Nansi Lake Basin is important for the health of people in many cities along the ERSNWTP, and determines the success of the project. According to the Chinese national plan for pollution treatment during the water transfer process of ERSNWTP, the water quality of the Nansi Lake needs to reach class III. However, water quality of Nansi Lake was inferior to class V overall, and total nitrogen (TN) and total phosphorus (TP) in the local lake exceed four times and two times of the demand standard of class III when the ERSNWTP was first launched in 2002. The water quality control of the Nansi Lake Basin was later remarked by experts as the Toughest Pollution Management Issue in the world (Wang et al. 2015; Liu et al. 2016). The main water pollution in Nansi Lake Basin is NPS pollution, and the contribution rate of NPS TP and TN pollution research more than 60% (Shen et al. 2008). Therefore, many scholars have been involved in the concern of NPS pollution control in Nansi Lake Basin (Huang et al. 2012b; Liu et al. 2012; Lin and Jing 2014). Therefore, it is of great significance to investigate and analyze the NPS pollution sources and their contribution ratios. The purposes of this paper are (1) to improve the ECM by developing a method to determine the export coefficients of NPS pollution from different land use types based on the hydrological and water quality data, (2) to simulate the contribution ratios of NPS TN and TP pollution from different sources in district level based on the improved ECM, and (3) to recognize the spatial characteristics of NPS TN and TP pollution. This study can not only provide the scientific data for the pollution control of Nansi Lake Basin but also present methods and results for similar simulation on NPS pollutant load of other ungauged watersheds of China.

Source apportionment based on the improved ECM

Export coefficient model (ECM)

The ECM, which is based on the idea that the nutrient load exported from a watershed equals the sum of the losses from individual sources, has been used in many studies. The individual sources consist of land use, livestock, rural living, and atmospheric deposition. The model allows accurate estimation and analysis of nutrient pollutants (Johnes, 1996). The ECM is outlined as follows:

$$L = \sum_{i=1}^n (E_i A_i (I_i)) + P \quad (1)$$

where L is loss of nutrient (kg), E_i is export coefficient for nutrient source i (kg/per head/year or kg/km²/year), A_i is area of the catchment occupied by land use type i (km²), or number of livestock type i , or of people, I_i is the input of nutrients to source i (kg), and P is the input of nutrients from precipitation (kg), which can be calculated as follows:

$$P = k \times V \times \alpha \tag{2}$$

where, k is the concentration of nutrient, V is the amount of rainfall (m³), α is runoff coefficient.

Improved export coefficient model (IECM)

Usually, the pollutants produced by various pollution sources in a certain basin will be collected by the surface runoff, and then entered the river or lake receiving the surface runoff. ECM model refined the relationship between NPS pollution sources and their output quantity, but did not consider the influence of rainfall runoff on pollution outputs from various types of land use and nutrient sources. Therefore, the calculating process of ECM model did not include the interception of soil and vegetation, groundwater infiltration, biochemical reaction, adsorption, degradation, etc. Moreover, the actual formation process of agricultural NPS pollution is complex, due to the randomness and intensity heterogeneity of rainfall, as well as the uncertainty of pollutant migration and transformation pathways (Ouyang et al. 2010; Zhang et al. 2010). In this study, we used the pollutant producing coefficient (λ) of sub-basins to represent the comprehensive influences of output intensity in rainfall runoff process and pollutant migration losses. The improved ECM (IECM) is expressed as follows:

$$L = \sum_{m=1}^n L_m \lambda_m \tag{3}$$

where L is the total loss of a certain basin(kg), L_m is loss of nutrient in sub-basin m (kg), λ_m is the pollutant producing coefficient of nutrient in sub-basin m , and n is the number of sub-basin.

Determination of the export coefficients for different land use types

The water quality data of monitoring points in the outlets included PS and NPS pollutants. The NPS pollutant discharge mainly occurred in the flood season, while the PS pollutant emission was relatively stable during the year because PS pollutants were controlled by local governments to meet local pollutant standard.

Therefore, the monitoring data of water quality in dry season can be treated as the amount of PS pollutant discharge after the degradation of river.

Therefore, the export coefficients of different land use types were determined based on the mass balance of the pollutants using hydrological and water quality data. The determination process was shown as follows:

First, the export coefficient of a specific land use type was defined as E_i , ($i = 1, 2, 3, \dots, n$), with n types of land use. Next, m small sub-basins ($m > n$) are chosen from the entire research area and the NPS pollution yield equation for each sub-basin is established as follows:

$$L = PS + \lambda L_0 + \lambda \sum_{i=1}^n [E_i \cdot A_i(I_i)] + \lambda P \tag{4}$$

Where L is the loss of nutrients (kg), PS is the point source pollution load (kg), L_0 is the annual NPS pollution loads from rural domestic waste and livestock (kg), E_i is export coefficient of land use type i in the watershed (kg/km²/year), A_i is area of land use type i (km²), and I_i is annual input load of the NPS pollutants from land use type i (kg).

The left side of Eq. (4) can also be expressed as follows:

$$L = C^* Q \tag{5}$$

where C is the annual mean observed concentration of the nutrients in the outlet of the sub-basin (g/L), and Q is the annual total stream flow measured in the outlet (m³).

The PS pollution load and the annual NPS pollution loads from rural domestic waste and livestock on the right side of Eq. (4) can be calculated as follows (Wu et al. 2015):

$$PS = \frac{C_d * Q_d}{D_d} \times 365.25 \tag{6}$$

where C_d is mean observed concentration of the NPS pollutants during the dry season in the outlet of the sub-basin (g/L), Q_d is total flow volume during the dry season at the outlet (m³), and D_d is the number of days in the dry season. So the annual NPS pollutants is the difference of the total pollutants and the PS pollution load (PS).

$$NPS = L - PS \tag{7}$$

$$L_0 = \sum_{j=1}^n [E_j \cdot A_j(I_j)] \tag{8}$$

where E_j is the export coefficient of humans or livestock, A_j is the population or number of livestock, and I_j is the input nutrient load from humans or livestock.

Then, Eq. (4) can be transformed to:

$$C^*Q = \frac{C_d^*Q_d}{D_d} \times 365.25 + \lambda \sum_{j=1}^n [E_j \cdot A_j(I_j)] + \lambda \sum_{i=1}^n [E_i \cdot A_i(I_i)] + \lambda P \quad (9)$$

With the analysis described above, we can obtain m equations for the m sub-basins, and the following equation group is established as follows:

$$\begin{cases} C_1^*Q_1 = \frac{C_{d1}^*Q_{d1}}{D_{d1}} \times 365.25 + \lambda_1 \sum_{j=1}^n [E_j \cdot A_{j1}(I_{j1})] + \lambda_1 \sum_{i=1}^n [E_i \cdot A_{i1}(I_{i1})] + \lambda_1 P_1 \\ C_2^*Q_2 = \frac{C_{d2}^*Q_{d2}}{D_{d2}} \times 365.25 + \lambda_2 \sum_{j=1}^n [E_j \cdot A_{j2}(I_{j2})] + \lambda_2 \sum_{i=1}^n [E_i \cdot A_{i2}(I_{i2})] + \lambda_2 P_2 \\ \dots\dots\dots \\ C_m^*Q_m = \frac{C_{dm}^*Q_{dm}}{D_{dm}} \times 365.25 + \lambda_m \sum_{j=1}^n [E_j \cdot A_{jm}(I_{jm})] + \lambda_m \sum_{i=1}^n [E_i \cdot A_{im}(I_{im})] + \lambda_m P_m \end{cases} \quad (10)$$

The export coefficient, E_i , for the different land use types can be deduced through optimization using the genetic algorithm (GA). In order to be comparable, the results with previous studies, this paper does not consider the impact of precipitation.

Materials and processing

Study area

Nansi Lake (South Four Lakes), located in Shandong Province of China, is one of the top ten freshwater lakes of China and the largest shallow freshwater lake of north China (Fig. 1). Nansi Lake Basin covers part of four cities, including the cities of Jining, Heze, Zaozhuang, and Taian with an area about 29,000 km² (Fig. 1). The

west part is the alluvial flood plain of Yellow River with the low-lying east to the west, and the east part consists low mountain hills and piedmont alluvial plains with the low-lying north to the south. The region has monsoon climate of medium latitudes with the average temperature range of 13.3–15 °C, an average frost-free period of 199–210 days, and average annual rainfall range of 590–820 mm. The total amount of water is 55.9×10^8 m³ including surface water resource of 22.5×10^8 m³. There are 6 major soil types including brown soil, moisture soil, solonchak soil, sand soil, sand ginger black soil and paddy soil. The main land use types are cultivated land (dry land and paddy land) and construction land, accounting for 67.7 and 16.7% in 2010. There were 23 counties (including cities), and the total population was 22.7×10^6 with 72.6% agricultural population in 2012.

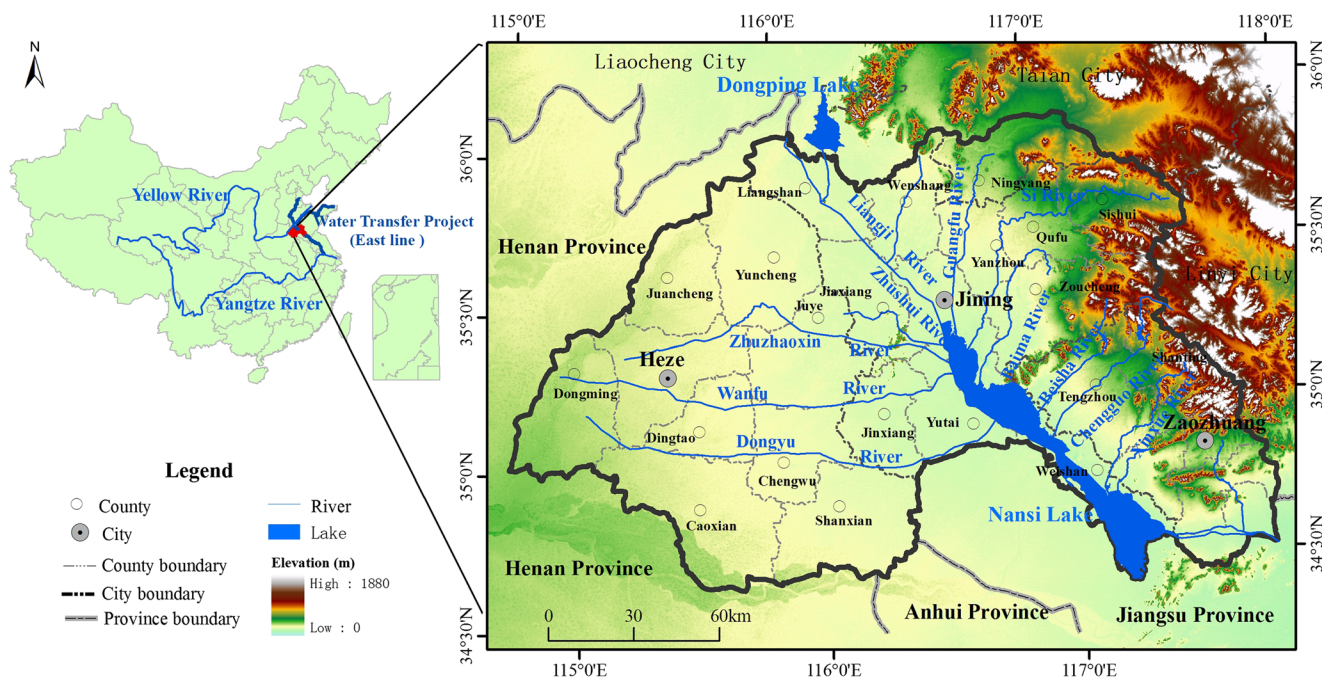


Fig. 1 Location of Nansi Lake Basin

Data sources and preprocessing

This research took NPS TN and TP pollutant as the study objects, and estimated the average source profiles in the period 2008–2012. The data used to estimate source profiles of NPS TN and TP pollution in Nansi Lake Basin included the social-economic data, land use data, digital elevation model (DEM), hydrological data, and water quality monitoring data.

Social-economic data included rural life data and agricultural breeding data of different counties, which were obtained from statistical yearbook of Shandong province from 2008 to 2012. Rural life data include the permanent rural population (RLP) and agricultural breeding data. Agricultural breeding data included four types (livestock breeding data (BLB), pig breeding data (PIB), breed data of sheep and goats (SGB), and poultry breeding data (PTB)). The annual average values from 2008 to 2012 for rural life data and agricultural breeding data were calculated and used to evaluate the amount of TN and TP pollution in Nansi Lake Basin, respectively.

Land use data in China are generally updated every 5 years. This study selected the land use data of 2010 to represent the average land use situation from 2008 to 2012. The land use data of 2010 were obtained from the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (<http://www.resdc.cn/b-f/b-f.asp>). This land use map was based on the 1:100000 land use/cover data of the Nansi Lake Basin and surrounding areas. Then, it was revised according to field survey. In accordance with the IECM and existing literature, the land uses of the Nansi Lake Basin were divided into six types: paddy land (PL), dry land (DL), woodland (WL), grassland (GL), water body and wetland (WB), and construction land (CL).

DEM data with a 30-m spatial resolution were downloaded from the National Aeronautics and Space Administration website (<https://reverb.echo.nasa.gov>). The map was revised by removing the background values. Then the slope data were calculated using surface analysis of ArcGIS Spatial Analyst.

Hydrological and water quality monitoring data from 2008 to 2012 were obtained from the weekly report on automatic water quality monitoring at key cross-sections of major rivers released by Shandong Environmental Monitoring Center. The hydrological and water quality variables included TN, TP, and stream flow and water flow rate.

In addition, the boundaries of Nansi Lake Basin were generated based on the administrative boundaries of counties and districts of the Shandong Municipality. The raster data were all resampled to 30 m. All the maps were projected into equal-area maps through Albers projection for the subsequent spatial analysis.

Table 1 Basic conditions of the 11 sub-basins in Nansi Lake Basin

Position	Code	Name	Area/km ²	Annual rainfall/mm	Monitor point	Runoff volume/10 ⁸ m ³	Average slope/°	Land use composition
Western part of Nansi Lake Basin	I	Dongyu River	4328.2	647.9	Xiyao	0.3	0.5	CL 80%, BL 17%
	II	Wanfu River	1592.1	660.4	Gaohe bridge	0.1	0.6	CL 81%, BL 15%
	III	Zhuzhaoxin River	3885.4	645.8	Yulou	0.6	0.5	CL 78%, BL 17%
	IV	Zhushui River	464.5	682.5	105 road bridge	0.1	1.0	CL 78%, BL 15%, GL 4%
	V	Liangji River	1955.7	682.5	Liji	0.7	0.6	CL 80%, BL 15%
Eastern part of Nansi Lake Basin	VI	Guangfu River	1373.1	682.5	Xishifo	0.7	0.8	CL 80%, BL 17%
	VII	Si River	2518.4	773.8	Yingou	0.6	2.5	CL 64%, GL 14%, BL 12%, WL 7%
	VIII	Baima River	759.9	682.5	Malou	0.4	0.7	CL 70%, BL 15%
	IX	Beisha River	1096.2	775.2	Wangchao bridge	0.2	3.0	CL 53%, GL 27%, BL 8%, WL 5%
	X	Chengguo River	808.3	817.9	Qunle bridge	1.1	2.8	CL 56%, GL 20%, BL 14%, WL 5%
	XI	Xinxue River	806.3	820.8	Ruhukou	2.4	4.9	CL 41%, GL 31%, WL 15%, BL 9%

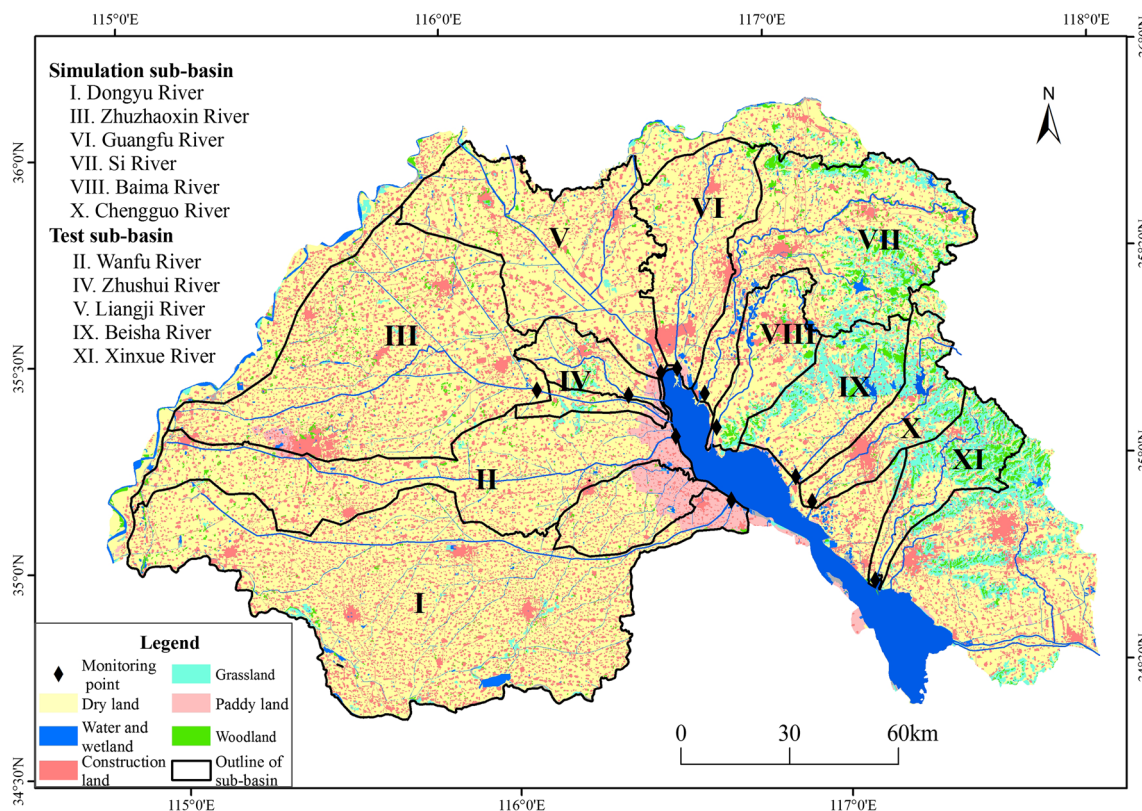


Fig. 2 Land use map and the 11 selected sub-basins

Calculation process of source apportionment based IECM

Watershed delineation

Eleven sites with monthly records of TN and TP pollution data were selected along the stream lines of 11 rivers. These rivers included Dongyu River, Wanfu River, Zhuzhaoxin River, Zhushui River, Liangji River, Guangfu River, Si River, Baima River, Beisha River, Chengguo River, and Xinxue River by Shandong Environmental Monitoring Center. Eleven sub-basins were delineated based on the digital elevation model (DEM, 30 m) data by taking water quality monitoring sections as outlets using ArcSWAT software (<http://swat.tamu.edu>). The basic conditions of the 11 sub-basins were shown in Table 1 and Fig. 2.

Determining coefficients of IECM

The pollutant producing coefficients (λ) were defined by the loss rates of NPS pollutant. Previous study had revealed N and P loss rates from different land use types under different precipitation conditions (Cheng et al. 2006). The nutrient loss rates under the precipitation range between 600 and 800 mm were selected as the pollutant producing coefficients for different land use

types in the Nansi Lake Basin. The export coefficients of rural life and rural breeding came from Pollutant Discharge Standard of Chinese Livestock and Poultry Breeding Industry (GB 18596-2001) (Table 2).

The export coefficients of different land use types were calculated through Eq. (10) and the calculation process were as follows: Firstly, the TN and TP pollution loads were calculated using the export coefficients of rural life and agricultural breeding and their quantity. Secondly, the TN and TP pollution loads were assigned to rural residential land as the pollution intensity. Thirdly, the TN and TP pollution loads were assigned to sub-basins according to the area of rural residential land. Finally, the export coefficients of different types of land use (Table 3) were calculated through Eq. (10) combined with the pollutant producing coefficients using the data of six sub-basin (Dongyu River, Zhuzhaoxin River, Guangfu River, Si River, Baima River, and Chengguo River). And then, the TN

Table 2 Export coefficients of rural living and rural breeding

Type	Agricultural breeding kg/year				Rural life kg/year
	BLB	PIB	SGB	PTB	RLP
TN	1.58	6.11	0.45	0.23	0.028
TP	0.16	1.01	0.17	0.045	0.012

Table 3 Comparison of output coefficients for various land use types with those of other regions

Region	References	Type	DL	PL	WL	GL	CL	WB
Hei River basin	Li and Zhuang, 2003	TN	29		2.38	10	–	–
		TP	0.9		0.15	–	–	–
Upper reaches of Yangtze River	Shen 2008	TN	29		2.38	–	11	10.6
		TP	0.9		0.15	0.2	0.24	4.3
Middle and lower reaches of Hanjiang River	Shi et al. 2002	TN	18.85	26.37	6.79	11.5	6.81	–
		TP	4.08	2	1.53	1.55	0.16	–
Baixiang River basin	Ren et al. 2015	TN	23.2		2.5	6	13	11
		TP	1.61		0.15	1.65	0.5	0.2
Beijing City	Liu et al.2011b	TN	20.2	–	4.38	6.26	–	–
		TP	2.06	–	0.35	0.68	–	–
Xili reservoir area	Li et al. 2016	TN	11.20	14.86	4.22	6.30	3.24	–
		TP	1.54	1.68	0.59	0.72	0.1	–
Henan city	Liu et al. 2016	TN	27.74		4.38	–	–	–
		TP	1.92		0.35	–	–	–
China	Li and Mao 2008	TN	12	–	1.9	3.2	–	–
		TP	1.1	–	0.05	0.18	–	–
Nansi Lake Basin		TN	9.23	11.63	4.25	6.04	2.89	1.93
		TP	1.52	1.64	0.44	0.89	0.27	0.12

and TP pollution loads from different sources and different counties were calculated and presented in Figs. 5, 6, and 7.

Results and discussion

Composition of non-point pollution

The total pollution amount and point source pollution loads of the 11 sub-basins were calculated by Eqs. (5) and (6), respectively. The amount of NPS pollution was obtained by the difference between the total pollution and the point source pollution. The proportions of NPS TN and TP pollution for the 11 sub-basins were calculated and shown in Fig. 3. The proportion range of NPS TN pollution was 45.3~86.7%, and the total proportion was 65.9% in Nansi Lake Basin. The proportion range of NPS TP pollution was 48.3~86.0%, and the total proportion was 66.8%. These indicated that the main pollution sources for TN and TP in Nansi Lake Basin was from NPS pollution, which are in accordance with results reported by Zhang et al. (2016), Shen et al. (2008) and Wei et al. (2013).

Model verification and uncertainty analysis

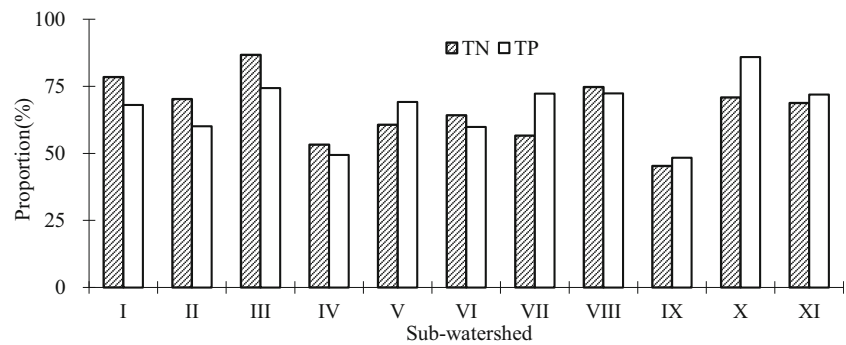
The total amount of NPS TN pollutant entering into the rivers (considering river entry coefficients of 0.3 for land use and 0.6 for rural living and breeding) in Nansi Lake Basin was 50,662 t/a, which had relative errors of -3.12 and -18.3% compared with the research results of Lin and Jing (2014) and Huang

et al. (2012b), respectively. The total amount of NPS TP pollutant entering into the rivers in Nansi Lake Basin was 8421 t/a, which had relative errors of 1.1 and 13.5% compared with the research results of Lin and Jing (2014) and Huang et al. (2012b), respectively. Therefore, estimated NPS TN and TP results by IECM were reliable and met the analytical requirements of the study.

The export coefficients of different land use types are important parameters for constructing ECM. In order to test the export coefficients of different land use types, the amount of NPS TN and TP pollutants of the five test sub-basins (Wanfu River, Zhushui River, Liangji River, Beisha River, and Xinxue River) were estimated using the calculated export coefficients of different land use types (Table 3) and export coefficients of rural living and rural breeding (Table 2). The calculated NPS TP and TN pollution loads were compared with those values obtained from Eqs. (5), (6), and (7), and the results were shown in Fig. 4. The relative error value ranges for NPS TN and TP pollution were -20.7~18.0% and -12.7~14.3% respectively. It is proved that the output coefficients of different land use types calculated is reasonable and reliable, which is exactly consistent with the output requirement of the model, and it can be further applied and popularized in the research area.

Comparing to export coefficients for various land use types of other regions reported in literatures (Table 3), export coefficients for different land use types in Nansi Lake Basin were relatively smaller, and the export coefficient values of dry land and paddy land were half times smaller than those of literatures. Therefore, the estimated results were bound to be wrong if the export coefficients for different land use types were taken from literatures.

Fig. 3 Proportions of NPS TN and TP pollution of the 11 sub-basins



However, the trend that export coefficients of cultivated land were larger than those of woodland and grassland.

It is known that livestock production in China has been industrializing and the treatment of rural domestic sewage has been paid more and more attention by local governments. Thus, manure discharges and rural domestic discharges are important point sources of nutrients in water systems. However, the rate of rural domestic sewage treatment is only 11% in China (Liu et al. 2013) and the manure sewage treatment rate was 46% in Shandong province (Shandong Provincial Government 2016). In this research, the pollutant loss rates were used to represent the average proportion of livestock manure and domestic sewage into the river in a certain sub-basin, which can estimate the amount of pollutants entering the rivers. It should be noted that the pollutant loss rate into the river (λ) used in Eq. (4) did not fully consider the environmental heterogeneity among catchments and ignored the atmospheric deposition. Thus, it is not surprising that the relative error was maintained at a relatively high level or even increased in some cases. It is expected that the results would be better if the pollutant loss rate (λ) values could be modified with the catchments variations.

The improved ECM is used in this research is a statistical model to estimate pollutant output of sub-basins rapidly by ignoring the mechanism process of NPS formation and transmission. However, NPS pollution has the characteristics of spatial diversity and complex process mechanism, and is affected by atmosphere, precipitation, topography, and other factors. The model structure of ECM determines that the estimated results were the average output level of pollution

sources with no spatial difference. Therefore, the estimated results had certain uncertainty, and the ECM can only be used as a statistical model to quickly estimate the pollutant outputs.

Source apportionment of NPS TN and TP pollution in Nansi Lake Basin

The amounts and contribution rates of NPS TN and TP pollution from different types of sources (including different types of land use, rural breeding, and rural life) were calculated based on ICEM. The total amounts of TN and TP pollution in Nansi Lake Basin were 50,662 and 8421 t, respectively. The output of NPS TN pollution was about six times of that of NPS TP pollution, which was close to reported NPS TN/TP ratios (ranging from 5 to 10) (Han et al. 2011). Comparing to the ratio of 8 from previous results for northern China, the NPS TP emissions were higher. The reason was that the amount of NPS TP pollution calculated in this study included those from rural breeding which has higher TP loss. If NPS TP pollution from rural breeding was ignored, the NPS TN/TP ratio was 7.8, which was closer with the reported ratio of northern China.

The amount of NPS TN and TP load produced by different types of pollution sources were calculated and shown in Fig. 5. The major source of NPS TN was rural living, which accounted for 40.5% of the total, followed by land use (33.6%), and rural breeding (25.9%). Among those seven land use types, dry land contributed the most (81.3%). Wheat and maize were the dominant crops in dry land. For the economic

Fig. 4 The observation and simulation of NPS TN and TP pollution in five test sub-watersheds

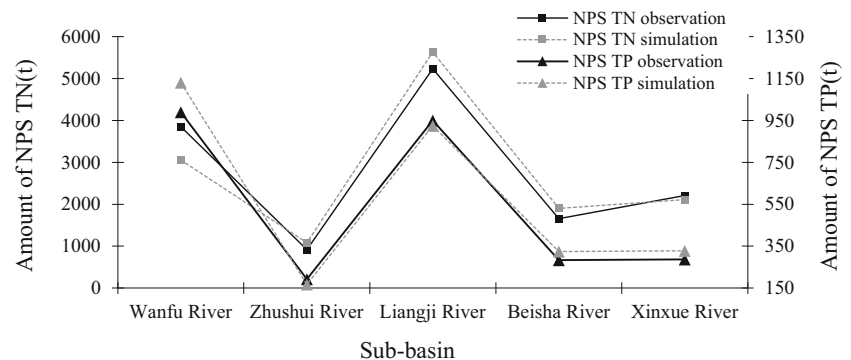
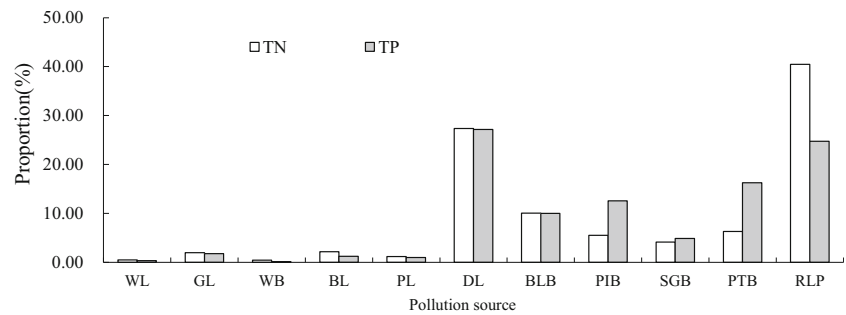


Fig. 5 NPS TN and TP load contribution rates from different kinds of pollution sources



yield from these crops, the use of fertilizers may have raised environmental contamination (Liu et al. 2011a). Furthermore, crop residue, which is not systematically managed, is a N-rich pollution source. It has been documented that regardless of the residual level, conventional tillage is less effective at reducing N in water and sediments than no-till systems (Mostaghimi et al. 1992; Liu et al. 2012). Conventional tillage is just the primary tillage pattern in Nansi Lake Basin. The major sources of NPS TP ranked differently from those of NPS TN: with rural breeding first (43.7%), followed by land use (31.6%), and rural residents (24.7%). Dry land was still identified as the dominant contributor for NPS TP among those seven land use types. And the contribution of rural breeding contributed the largest amount of NPS TN pollution.

For the different rural breeding sources, livestock breeding contributed largest amount of NPS TN pollution with the contributed proportions of 38.7%, and poultry breeding contributed the largest amount of NPS TP pollution with the contributed proportion of 37.3%. For all types of NPS TN pollution sources, rural life, dry land, and livestock breeding were the main pollution sources, and the total output proportion of the three types was 77.8%. For all types of NPS TP sources, dry land, rural life, and poultry breeding were the main pollution sources, and the total output proportion of these three types was 61.9%. The fact that 96% of the villages have no drainage

ditches and sewage treatment system (Liu et al. 2013) may aggravate the NPS pollution load of Nansi Lake Basin. Therefore, the improvement of sewage systems in villages and efficient treatment of sewage systems is significant for NPS pollution management.

Spatial distribution of NPS TN and TP pollution in Nansi Lake Basin

The contribution rates of NPS TN and TP pollution for different districts flowing into rivers were calculated and shown in Figs. 6 and 7. The contribution rates of NPS TN pollution load in descending order were as follows: Heze city (45.1%), Jining city (36.0%), Zaozhuang city (14.8%), and Taian city (4.1%). Furthermore, the contribution rates of NPS TP pollution load were as follows: Heze city (43.5%), Jining city (37.7%), Zaozhuang city (14.6%), and Taian city (4.2%). The range of NPS TN and TP contribution rates in country level were 2.2–8.9% and 2.1–8.8%, respectively. For NPS TN contribution in county level, the contribution rates of Shanxian, Tengzhou, Yuncheng, Caoxian, and Zaozhuang city were over 5%, and the contribution rates of Yutai, Yanzhou, Jining, and Qufu were below 3%.

The load intensities of NPS TN and TP pollution for different districts flowing into rivers were calculated and shown

Fig. 6 Proportions of NPS TN pollution from different sources for different districts

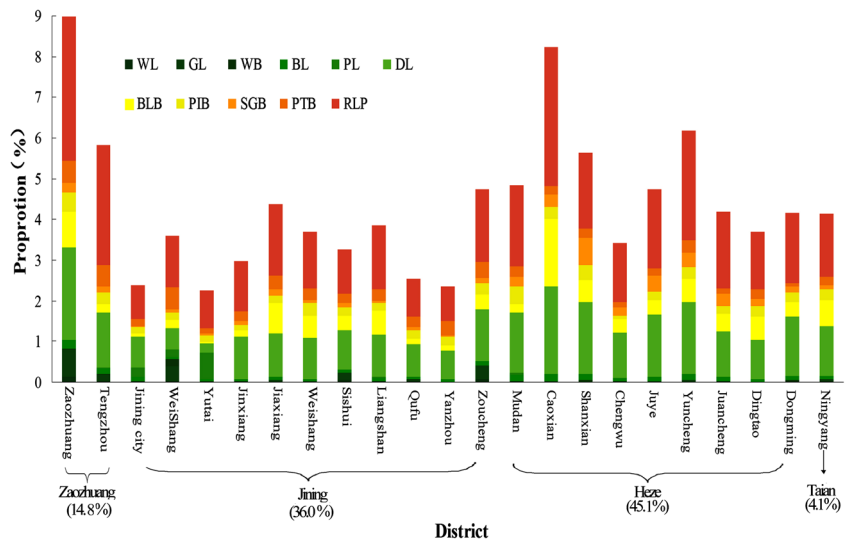
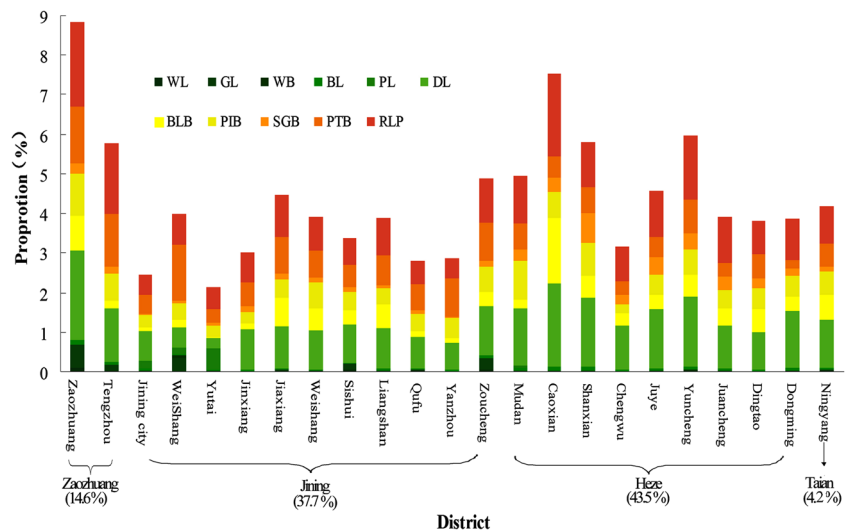


Fig. 7 Proportions of NPS TP pollution from different sources for different districts



in Figs. 8 and 9. The ranges for the load intensity of NPS TN and TP pollution were 11.3–22.9 and 2.1–3.9 kg/ha, respectively. The average NPS TN and TP load intensities of Nansi Lake Basin were 17.7 and 2.9 kg/ha, respectively. Both NPS TN and TP loads in district level had the similar trend that the load intensities of the districts in the eastern part of Nansi Lake Basin were relative lower than those in the western part. Jining city, Zoucheng county, Sishui county, Weishan county, and Zaozhuang city in eastern part of Nansi Lake Basin had the

lower load intensities for both NPS TN and TP pollution. Dingtao county, Juancheng county, and Caoxian county in the western part had the higher load intensities for both NPS TN and TP pollution. It is of great significance to note that the counties nearby the main channel of ERSNWTP, such as Wenshang, Yanzhou, Liangshan, and Jiaxiang also had the higher load intensities for both NPS TN and TP pollution. Therefore, these four counties should be the key areas for the NPS TN and TP pollution.

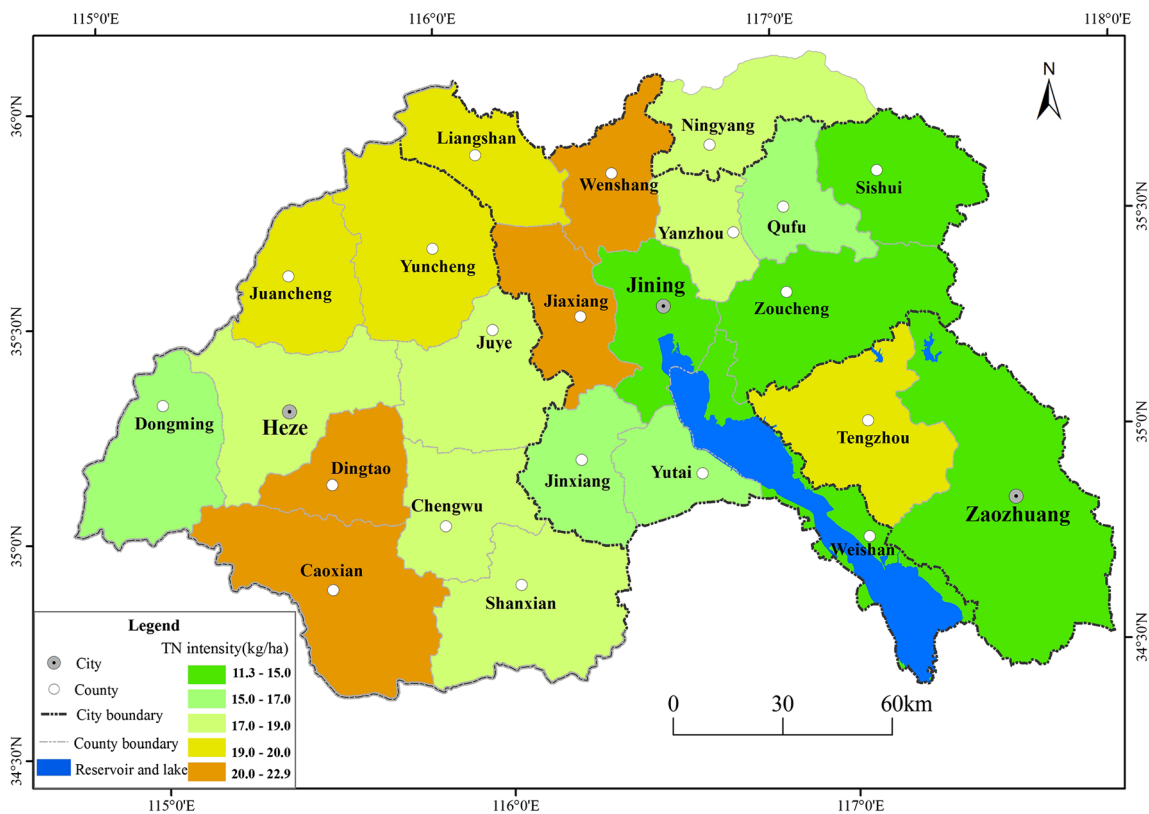


Fig. 8 Load intensity of NPS TN pollution for different districts

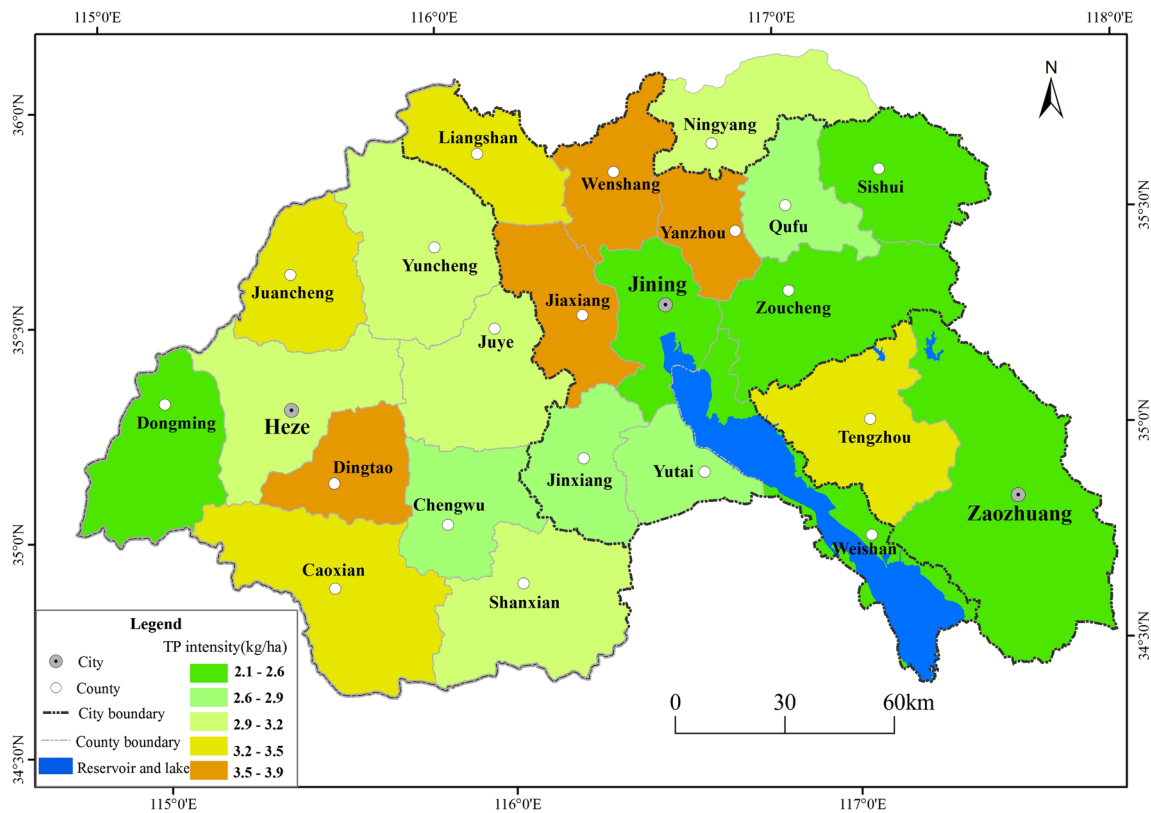


Fig. 9 Load intensity of NPS TP pollution for different districts

Conclusions

This research improved Johnes export coefficient model (ECM) by developing a method to determine the export coefficients of different land use types based on the hydrological and water quality data. The case study in the Nansi Lake Basin provided reliable export coefficients of different land use types, which showed this method is convenient, feasible, and is suitable for the places where NPS pollution monitoring data are scarce. It should also be note that the ECM and improved ECM are empirical models and the export coefficients are affected by rainfall, terrain, soil types etc. Therefore, the relationships between export coefficients and their impact factors need to be evaluated in a future study.

The NPS TN and TP pollution had become the main pollutants affecting water quality in Nansi Lake Basin, and the total proportion of NPS TN and TP pollution was 66.8 and 66.0%, respectively.

Rural living was found as the most important factor in contributing NPS TN pollution, while rural breeding contributed the most of NPS TP pollution in Nansi Lake Basin. Therefore, the improvement of sewage systems in villages and efficient treatment of sewage systems of rural breeding are significant for NPS pollution management in Nansi Lake Basin. Dry land was the main source for both NPS TN and TP pollution. This was primarily because of the overuse of fertilizers and inappropriate cultivation techniques. Finally, this

study analyzed the spatial distribution and contribution of NPS TN and TP pollution, and highlighted that the counties nearby the main channel of ERSNWTP such as Wenshang, Yanzhou, Liangshan, and Jiaxiang were the key areas for the control of NPS TN and TP pollution.

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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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