



Article Development and Assessment of a New Framework for Agricultural Nonpoint Source Pollution Control

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Abstract: The transport of agricultural nonpoint source (NPS) pollutants in water pathways is affected by various factors such as precipitation, terrain, soil erosion, surface and subsurface flows, soil texture, land management, and vegetation coverage. In this study, based on the transmission mechanism of NPS pollutants, we constructed a five-factor model for predicting the path-through rate of NPS pollutants. The five indices of the hydrological processes, namely the precipitation index (α), terrain index (β), runoff index (TI), subsurface runoff index (LI), and buffer strip retention index (RI), are integrated with the pollution source data, including the rural living, livestock and farmland data, obtained from the national pollution source census. The proposed model was applied to the headwater of the Miyun Reservoir watershed for identifying the areas with high path-through rates of agricultural NPS pollutants. The results demonstrated the following. (1) The simulation accuracy of the model is acceptable in mesoscale watersheds. The total nitrogen (TN) and total phosphorus (TP) agriculture loads were determined as 705.11 t and 3.16 t in 2014, with the relative errors of the simulations being 19.62% and 24.45%, respectively. (2) From the spatial distribution of the agricultural NPS, the TN and TP resource loads were mainly distributed among the upstream of Dage and downstream of Taishitun, as well as the towns of Bakshiying and Gaoling. The major source of TN was found to be farmland, accounting for 47.6%, followed by livestock, accounting for 37.4%. However, the path-through rates of TP were different from those of TN; rural living was the main TP source (65%). (3) The path-through rates of agricultural NPS were the highest for the towns of Wudaoying, Dage, Tuchengzi, Anchungoumen, and Huodoushan, where the path-through rate of TN ranged from 0.17 to 0.26. As for TP, it was highest in Wudaoying, Kulongshan, Dage, and Tuchengzi, with values ranging from 0.012 to 0.019. (4) A comprehensive analysis of the distribution of the NPS pollution load and the path-through rate revealed the towns of Dage, Wudaoving, and Tuchengzi as the critical source areas of agricultural NPS pollutants. Therefore, these towns should be seriously considered for effective watershed management. In addition, compared with field monitoring, the export coefficient model, and the physical-based model, the proposed five-factor model, which is based on the path-through rate and the mechanism of agricultural NPS pollutant transfer, cannot only obtain the spatial distribution characteristics of the path-through rate on a field scale but also be applicable to large-scale watersheds for estimating the path-through rates of NPS pollutants.

Keywords: nonpoint source pollution; path-through rate; pollution load; hydrological process

1. Introduction

The second national census of pollution sources indicated that agriculture is the leading cause of nonpoint source (NPS) pollution in China [1]. Sustainable agriculture depends on a delicate balance between economic stability (achieved through the increase in agricultural productivity to meet national food demands), reduction in the use of finite natural resources, and reduction of NPSs, which have detrimental effects on the environment [2,3]. Contrary to point source pollution, NPS pollution is characterized by random

and intermittent occurrences, complex mechanisms and processes, uncertain discharge channels and amounts, variable spatial and temporal pollution loads, and difficulties in monitoring, simulation, and control [4]. These characteristics cause challenges in the simulation and evaluation of the transport processes of NPS pollutants from agricultural sources to sinks [5].

Many studies have indicated that NPS pollution is closely related to watershed runoff and confluence, while it is not evidently related to watershed-scale loading and concentration [6]. In general, the migration of NPS pollutants and their transport process can be divided into two hydrological processes: the generation of pollutants and their transfer to waterbodies [7]. The path-through rate is an all-new concept and a comprehensive parameter that can describe the NPS pollution transfer process, starting from pollutant generation to transfer to waterbodies [8]. Based on this concept, risk assessment can be performed for NPS pollutants from each grid to the receiving waterbodies in the watershed scale by determining the risk grade according to the water criteria of different water environmental function zones. Although some scholars have also conducted similar studies through point monitoring (based on the coefficient of NPSs into the river), their results cannot be extended to the watershed scale. In particular, the pollutants generated and accumulated in watersheds are derived, transmitted, and intercepted by precipitation, slope, and other surface and terrain factors, expressed as a scaling factor, which is obtained by dividing the part of the pollutant load transferred to waterbodies by the total pollutant load.

Currently, there are approximately three types of estimation methods for calculating the path-through rate of watershed agricultural NPS pollutants: field monitoring, the traditional empirical model, and physical-based models [9]. Field monitoring has several advantages such as high accuracy and the ability to monitor the entire migration process of NPS pollutants. However, owing to its high cost and time requirement [10], it is more applicable to small-scale watersheds or fields rather than to large-scale watersheds on a national scale.

The traditional empirical model involves the export coefficient mode (ECM), hydrologic curve partitioning, the soil conservation service curve number (SCN-CN), and so on. [11]. This model integrates the transport process as a "black box" or "gray box" model, completely ignoring the migration mechanism and the transformation of NPS pollutants at the watershed scale [12]. In addition, the export coefficients are inherently highly variable and reflect particular site conditions for each study. Physical-based models include SWAT, HSPF, and AnnAGNPS, which are integrated with a large number of parameters and functional formulas for simulating the whole process of NPS pollutant transfer from sources to sinks and are more commonly applied for mesoscale watersheds. However, there are uncertainties with regard to their practical application because of differences in input data, insufficient localization of parameters, and lack of operator experience [13].

To overcome these weaknesses and to fully leverage the advantages of the existing physical-based and empirical models, two hypotheses were proposed. First, it is possible to generalize the major process of transfer of pollutants from sources to sinks, and second, the generalized process can be integrated into a new model on the watershed and administrative scales. In this study, a novel path-through rate model of NPS pollutants is developed using five major factors, namely the precipitation index (α), terrain index (β), runoff index (LI), subsurface runoff index (TI), and buffer strip retention index (RI). Hereafter, the model is referred to as the path-through rate five-factor model (or PTRFFM). The technical framework of the PTRFFM is illustrated in Figure 1. To check the reliability of the PTRFFM, a case study is conducted in a headwater watershed of a prominent drinking water supply in the Beijing-Tianjin-Hebei region. Overall, the objective of this study is to establish the PTRFFM to predict the path-through rate of NPS pollutants jointly on the watershed and administrative scales and estimate the resources, load, and distribution of agricultural NPS pollutants. The study also aims to verify the reliability of the PTRFFM through the assessment of the pollution load, critical source areas, and spatial distribution.



Figure 1. Technical framework of the PTRFFM.

2. Methods and Materials

2.1. Study Area

The Miyun Reservoir is the most important source of drinking water in the Beijing-Tianjin-Hebei region (Figure 2). The Chaohe watershed is the upper watershed of the Miyun Reservoir, and it covers a drainage area of 4888 km², consisting of 16 rainfall stations and 3 hydrology stations. The average annual precipitation is between 350 mm and 630 mm, where approximately 70% of the precipitation occurs from June to September, during which the region frequently receives heavy rainfalls.



Figure 2. Map of the study area.

The watershed governs 24 towns, which are mainly composed of agricultural populations (~390,000). Livestock, poultry farming, and agriculture are more developed in the area compared with other industries, and the main crops are wheat, corn, and soybean. In the recent two decades, agricultural NPS pollution has become the dominant factor affecting the water quality of the Miyun Reservoir, where nitrogen and phosphorus predominantly limit the eutrophication of water bodies. Consequently, forecasting the critical source areas of the upper watershed is highly significant for ensuring the safety of the water environments and promoting sustainable development in this area.

2.2. Construction of the PTRFFM

2.2.1. Conceptualization of Five Major Parameters

Considering the effects of precipitation, terrain, runoff, surface runoff, and buffer strip retention on NPS pollution, the five-factor model is described by Equation (1).

$$\lambda_i = f(\alpha, \beta, TI, LI, RI) \tag{1}$$

where λ_i is the path-through rate, α is the precipitation index, β is the terrain index, TI is the surface index (mm), LI is the subsurface runoff index (mm), and RI is the buffer strip retention index. These five indices are correlated with the spatial unit (grid cell), which was set to be 30 m × 30 m based on the digital elevation model (DEM), precipitation, slope, land use, and vegetation coverage map using ArcGIS 10.2.

Precipitation index. The precipitation index (α) is determined by multiplying the impact factors of the spatial and temporal changes in annual precipitation, denoted by α_s and α_t , respectively. In this paper, the spatial changes in precipitation are obtained from the rainfall data of 16 stations based on the inverse distance weighting interpolate method. The NPS pollution load delivered to a river is positively correlated with precipitation; therefore, the NPS pollutant loss can be identified as a function of rainfall [14].

$$L = f(r) \tag{2}$$

$$\alpha = \alpha_s \times \alpha_t = \frac{f(r)}{f(r)} \times \frac{R_j}{\overline{R}}$$
(3)

where *L* is the annual loss of NPS pollutants that drain into rivers with runoff (kg), which can be obtained by observing the outlets, and *r* is the annual rainfall in the entire study watershed for a given year (mm). According to hydrology and water quality data, monitored by 16 hydrological stations near the Chaohe River watershed from 1991 to 2018, the correlation equation between the watershed annual rainfall and the load of the NPS pollutants discharged into rivers can be realized as follows:

$$L_N = 0.0402r^2 - 27.81r + 530.14 R^2 = 0.8631$$
(4)

$$L_P = 0.0007r^2 - 0.4538r + 77.939 R^2 = 0.7374$$
⁽⁵⁾

In addition, the spatial variation impact factor α_s , is obtained as, where R_j is the annual rainfall in a grid cell for a given year (mm) and \overline{R} is the average rainfall of the entire watershed for a given year (mm).

Terrain index. The terrain index (β) is used to describe the terrain heterogeneity effect on NPS pollutant loads. Many studies have demonstrated that the slope has a profound impact on parameters such as runoff flux and velocity. Since runoff is the primary carrier of nutrients, nutrient loss is also greatly impacted by the slope [15]. The relationship between the runoff and slope can be established as shown below [16]:

$$\beta = \frac{\theta_j^{\ d}}{\overline{\theta}^d} \tag{6}$$

where θ_j is the slope for grid *j* (30 m × 30 m), and $\overline{\theta}$ is the average slope of the entire study watershed. The temporal variation of the terrain is not remarkable in short time periods; therefore, the β value varies for different grid cells. In a specific grid cell, the β value does not change over the year.

The runoff index (TI) was used for the calculation of the runoff index using the Soil Conservation Services Curve Number method (Soil conservation service curve number, SCS-CN) [17].

$$S = \left(\frac{25,400}{CN}\right) - 254\tag{7}$$

$$TI = \frac{(P - 0.3S)^2}{p + 0.7s} \tag{8}$$

where TI is the runoff depth (mm); CN is the curve number, which is a function of land use, soil, management, and hydrologic condition; P is the rainfall of the watershed (mm/a); S is the potential maximum retention (mm/a). In SCS models, soils are defined by groups based on their runoff potential-low, moderately low, moderately high, and high.

Subsurface flow index. In China, NPS pollution by nitrate leaching is not exclusively limited to intensively used agricultural ecosystems, but it has also been reported for forest ecosystems exposed to high loads of atmospheric N deposition. *LI* is a widespread method for evaluating the soil water infiltration capacity with regard to the impact on the losses of nitrogen and phosphorus, through which hydrological soil groups are based on the soil type, land use, and previous soil moisture condition. *LI* is determined by multiplying the rainfall spatial distribution index (*PI*) and seasonal distribution index (*SI*), where the *PI* represents the maximum theoretical rainfall that can be used for the infiltration of watershed units and *SI* represents the effect of seasonal rainfall changes on the soil moisture infiltration [18,19]. The specific calculation formula is as follows:

$$LI = PI \times SI = \frac{(prec - 0.4 \times S)^2}{prce + 0.6 \times S} \times \sqrt[3]{2 \times \frac{prec(ls)}{prec}}$$
(9)

where *prec* is the annual precipitation (mm/a), *S* is the annual potential maximum retention (mm/a), and *prec*(*ls*) is the total precipitation in non-flood seasons (November to May of the following year).

Buffer strip retention. Runoff rates have spatial patterns controlled by loading and filtering along with the flow paths from the upslope contributing area and downslope dispersal area [20]. *RI* values are a relative estimate of high to low buffering, and they represent a relative buffering likelihood. *RI* can be expressed as follows:

$$RI = ln\left(\frac{\sum_{DA=1}^{N} T_{DA_i}}{tan_{B_{DA_i}}}\right)$$
(10)

where $\sum T_{DAi}$ is the cumulative trapping efficiency within a dispersal area, and B_{DAi} is the average slope of a dispersal area.

Soil erosion index. Phosphorus is lost in the form of particles in water reservoirs owing to soil erosion. The universal soil loss equation is applied to evaluate the path-through rate of total phosphorus in this study [21].

$$A = R \times K \times L \times S \times C \times P \tag{11}$$

where A is the estimated average soil loss in tons per acre per year, R is the rainfall-runoff erosivity factor, K is the soil erodibility factor, L is the slope length factor, S is the slope steepness factor, C is the cover management factor, and P is the support practice factor.

2.2.2. Data Standardization of the PTRFFM

The five-factor raster data are standardized to unify the calculation unit. After the normality test, the standardization of the normal and skewed distribution data is calculated as follows:

Normal distribution
$$X = \frac{X_i - min(x)}{max(x) - min(x)}$$

Skewed distribution
$$NX = \frac{\Phi X_i}{X_i} \Phi X_i < X_i$$

 $NX = \frac{X_i}{\Phi X_i} \Phi X_i > X_i$

where *X* and *NX* are standardization factors, X_i is the value of each factor for grid *i*, min(x) is the minimum grid value for each factor, max(x) is the maximum grid value for each factor, X_i is the maximum value for factor *i*, and ΦX_i is the median for factor *i*.

The precipitation and terrain indices are both dimensionless quantities; therefore, α and β do not need to be standardized. *TI*, *LI*, *RI*, and *A* have dimensions to ensure that the NPS pollutant load to water bodies is less than the pollutant source load; therefore, they need to be standardized, and then a spatial overlay operation needs to be performed for each of the standardized results. The standardization method of total nitrogen (TN) and total phosphorus (TP) is as follows:

$$\lambda_{TN} = \alpha \times \beta \times NormTI \times NormLI \times NormRI$$
(12)

$$\lambda_{TP} = \alpha \times \beta \times Norm(A + TI) \times NormRI \times NormRI$$
(13)

2.3. Data Source

The input data include a DEM, land use, soil type, vegetation map, weather data, hydrological data, and socioeconomic data of the study area (Table 1). The land-use types are classified into five types: farmland, grassland, forest, residential land, and unused land. The NPS of the study watershed is categorized into three types: rural living, livestock, and land use.

Туре	Scale	Resolution	Description	Source
DEM	1:1,000,000	$30 \times 30 \text{ m}$	Elevation, overland and channel slopes and lengths	National Geomatics Center of China
Land use	1:50,000	$30 \times 30 \text{ m}$	Land use classifications	Institute of Geographical and Natural Resources Research, Chinese
Livestock	_	Village	Breeding number of cow, pig, sheep, and poultry	National pollution source census
Soil type	1:1,000,000	30 imes 30 m	Soil type classifications	Institute of Soil Science, Chinese Academy of Sciences
Administrative division	1:1,000,000	$1 imes 1 \ {\rm km}$	Towns and municipalities of the study area	National Geomatics Center of China
Vegetation coverage		$1 imes 1 \ { m km}$		
Precipitation	2001–2014	Daily step	16 stations in the study area	China Meteorological Administration
Hydrological data	2001–2014	Monthly step	Calibration and validation of the five-factor model	China Meteorological Administration
Water quality data	2001–2014	Monthly step	Calibration and validation of the five-factor model	Miyun Reservoir Management Office Xiahui Hydrological Station
Social economics	24 towns	Township level	Population, livestock rearing, fertilizer application	Field investigation; Statistics year-book
Export coefficient	Different sources		Losses of various nutrient sources	Statistical data and field monitoring

Table 1. Source and	l description	of basic data.
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2.4. Estimation of the Agricultural NPS Pollutant Loads

The ECM is used to estimate the NPS loads when the path-through rate is obtained using the PTRFFM. The calculation method is as follows:

$$L = \sum_{i=1}^{n} PE_i[A_i(I_i)]$$
(14)

where *L* is the loss of nutrients (kg), E_i is the export coefficient of the nutrient source *i* (kg·ca⁻¹·a⁻¹ or kg·km⁻²·a⁻¹) (see Table 2 for details), A_i is the area of the watershed occupied by the land use type *i* (km²), or the number of livestock type *I*, or of people, I_i is the nutrient input to the source *i* (kg), and *p* is the path-through rate of each grid.

Table 2. Export coefficients	s for	different	pollution	sources.
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	Pollution Source	TN	TP
	Farm land	2.97	0.041
	Forest	0.24	0.0071
Land use (kg·ha ^{-1} ·a ^{-1})	Grassland	1.57	0.013
	Unused land	5.00	0.008
	Residential land	2.00	0.003
Rural living (kg·ca ^{-1} ·a ^{-1}) rural resident		2.83	0.89
	Big livestock	7.36	0.31
T • (1	Pig	0.41	0.15
Livestock	Sheep	1.4	0.045
	Poultry	0.071	0.004

3. Results and Discussion

3.1. Major Parameters of the Spatial Distribution from the PTRFFM

The results show that α_{TN} and α_{TP} were 1.28–8.18 and 0.29–7.42, respectively. β , LI, and TI were the same for TN and TP, that is, 0–2.57, 301.99–663.32 mm, and 182.49–483.12 mm, respectively. In addition, RI_{TN} and RI_{TP} were 0.21–17.5 and 0–17.81, respectively, and A_{TP} was 0.00166–852.63 (t/(hm²·a)).

A comprehensive analysis of the spatial distribution of the five factors (Figures 3 and 4), TN, TP, α , LI, and TI was performed; the results show a high similarity of the spatial distribution characteristics with the rainfall map. Specifically, the areas with high rainfall (upstream Wudaoying, Tuchengzi, Dage, Anchungoumen, and downstream Tuchengzi) also had high α , LI, and TI because these three indices were mainly calculated on the basis of rainfall. The terrain index β is proportional to the watershed slope, showing a high trend in some parts northwest and a low trend in the cultivated land in the valley. From the perspective of the impact degree of the land use, the cultivated land area was characterized by high LI and TI and low β and RI. The likely reason for this trend is that the CN value of the cultivated land was larger than those of other types of land use, which promoted the generation of surface and subsurface runoffs. In addition, β of the cultivated land in the valley was lowest because of the gentle slope in the area. Low RI is possibly because of interception by forest grass and the water surface buffer system. Thus, cultivated land has no intercepting effect on NPS pollution.



Figure 3. Spatial distribution of the five factors of NPS with respect to TN.

3.2. Spatial Distribution Characteristics of the Path-through Rate

For a field of scale (Figure 5), the path-through rate of TN was mainly distributed in the farmland of the towns of Wudaoying, Dage, Tuchengzi, Anchungoumen, and Huodoushan; this is similar to the spatial distribution characteristics for TP.

For the town scale (Figure 5), the primary spatial distribution of the path-through rate of TN was in towns such as the upper reaches of Wudaoying, Dage, and Tuchengzi and the lower reaches of Anchungou Gate and Huodoushan; the rates ranged from 0.17 to 0.26. However, the upper reaches of the basin and the west side of the middle reaches, including Wudaoying, Kulongshan, Dage, and Tuchengzi, were the primary distribution areas for the path–through rate of TP, with the rates ranging from 0.012 to 0.019.



Figure 4. Spatial distribution of the five factors of NPS with respect to TP.

3.3. NPS Pollution Load and Source Analysis

The TN and TP loads of the research area in 2014 were obtained according to the ECM and coefficient models. The simulation results indicate that the TN load was 13.87 times that of the TP load, which were 7505.0 t and 540.95 t, respectively.

As shown in Table 3, for different pollution sources, the pollution loads of TN were 1118.2 t, 2815.3 t, and 3616.5 t, for rural living, livestock, and land use, correspondingly accounting for 15%, 37.4%, and 47.6% of the TN loads, respectively. Among all the pollution sources, farmland was the major contributor (2258.9 t·a^{-1.}), mainly owing to the unreasonable fertilizer rate and long-term conventional tillage [22]. Among the four types of livestock, big livestock was the primary contributor (1404.9 t), followed by poultry (1187.7 t), because of the improper management of animal feed and livestock manure. The major TP pollution sources differed from the major TN pollution sources. The main contributors of TP loads were rural living (351.65 t·a⁻¹), followed by livestock (156.6 t·a⁻¹) and land use (32.7 t·a⁻¹), which account for 65%, 28.95%, and 6.05% of the TP loads, respectively. In recent years, the urbanization level in most parts of the studied watershed gradually increased, and the living standards of people continued to increase, resulting in rural living becoming the major contributor to the TP load.



Figure 5. Spatial distribution of path-through rate for the NPS pollution of field and town scales.

Pollution Source		Total NPS Pollution Load of 2014					
		TN (t/a)	Percent	Total	TP (t/a)	Percent	Total
Rural living		1118.2	15.0	15.0	351.65	65.00	65.00
	Big stock	1404.9	18.7	37.4	59.17	10.94	28.95
Livestock	Pig	70.0	0.9		25.61	4.73	
LIVESTOCK	Sheep	152.7	2.0		4.91	0.09	
	Poultry	1187.7	15.8		66.91	12.37	
	Farmland	2258.9	30.1	47.6	22	4.07	6.05
	Forest	412.8	5.5		3.1	0.57	
Land use	Grassland	878.1	11.7		6.4	1.18	
-	Unused land	43.3	0.6		0.7	0.13	
	Residential land	23.4	0.03		0.5	0.09	
Total		7505.0	100%	100%	540.95	100%	100%

Table 3. NPS	pollution	load from	n various	sources in 2014.
	ponution	1000 1101	ii vuiious	50urces in 2011.

3.4. Validation of the Results from the PTRFFM

Comparison with the monitoring data. The water quality monthly data of TN and TP, *C* (mg/L), as well as the monthly average runoff data, *Q* (mg/l), monitored by the Xiahui hydrological station at the watershed outlet, were utilized to estimate the TN and TP fluxes of the watershed L_0 . The pollutant flux data obtained from the Xiahui hydrological station were calibrated and validated by multiplying with water quality and runoff, based on the following calculation formula: $L_0 = \sum_{i=1}^{12} 10^{-6} \times C_i \times Q_i$. The relative error *Re* was analyzed by comparing the simulated value L with the monitored value L_0 to assess the accuracy of the PTRFFM.

$$Re = \frac{L - L_o}{L_o} \times 100\% \tag{15}$$

The results demonstrated the effectiveness of the five-factor model based on the mechanism of the transport pathways of agricultural NPS pollutants (Table 4). In 2014, the fluxes of the TN and TP of the studied watershed were 570.86 t and 2.54 t, and the simulated values were 705.11 t and 3.16 t, respectively. The relative errors were less than 25%, 19.62% and 24.45%. The simulated values for TN and TP were both overestimated, probably because the interception, retardation, and attenuation effects of the pollutants after entering the watercourse were not considered in the PTRFFM.

Table 4. Validation results of the five-factor model.

NPS Pollutants	NPS PollutantsSimulated Value L (t)		Re %	
TN	705.11	570.86	19.62	
TP	2.54	3.16	24.45	

Comparison with other relevant studies. To further verify the reliability of the model simulation results, some typical results from the literature on the NPS pollution paththrough rate of China were reviewed (Table 5). For instance, according to Zhou et al., the path-through rates of N and P in the Haihe watershed were 0.2 and 0.1, respectively. A field monitoring evaluation conducted in the Xinanjiang River Basin showed that the point source path-through rate was approximately 2–3 times that of the NPS path-through rate; the TN and TP path-through rates were 0.2 and 0.21, respectively [23]. Deng et al. studied the path-through rates of different NPS pollution sources in the Changle River Basin using the SWAT model and demonstrated that the atmospheric deposition of the NPS path-through rate of TN was 0.17, followed by nitrogen fertilizers (0.15), rural living (0.12), and livestock (0.03) [24]. Ma et al. utilized the pollutant load estimation method in the Laizhou Bay of the Yellow River Basin and discovered that the NPS path-through rate of TN for rural living was 0.17, followed by farmland (0.1) and livestock (0.07) [25]. Based on the analyses mentioned above, the results of the NPS path-through rate of TN in this study are similar to those of other studies. The NPS path-through rate of TP is lower compared with those of the other studies. This is probably because the studied watershed is in the North of China, which is characterized by inadequate annual rainfall. Moreover, phosphorus is mainly lost in the particulate form by soil erosion, and in the North of China, scarce rainfall leads to less soil erosion compared with other research areas with abundant rainfall.

Watershed	Study Area	Scale	Method	λ_{TN}	λ_{TP}	Reference
Haihe Basin	Chaohe River watershed	Mesoscale	PTRFFM	0.17-0.26	0.012-0.019	This study
Haihe Basin	Haihe Basin	Large scale	ECM	0.20	0.10	Zhou et al., 2004
Yangtze River Basin	Xinanjiang River Basin	Large scale	Investigation and monitoring	0.20	0.21	Yang et al., 2006
Songhua River Basin	Harbin Section of Songhua River	Middle scale	Pollutant load estimation method	0.10	_	Fu et al., 2010
Yangtze River Basin	Changle River Basin	Middle scale	SWAT	0.17 (Atmospheric deposition) 0.12 (Rural living) 0.03 (Livestock) 0.15 (nitrogen fertilizer)	- - -	Deng et al., 2012
Yellow River Basin	Laizhou Bay	Large scale	Pollutant load estimation method	0.1 (Farmland) 0.25 (Rural living) 0.07 (Livestock)	0.1 (Farmland) 0.25 (Rural living) 0.07 (Livestock)	Ma et al., 2012

Table 5. Results of the other studies on the path-through rate of NPS pollutants in China.

3.5. Identification of the Critical Source Area of NPS Pollution

A comprehensive analysis of the distribution of the NPS pollution load and the paththrough rate was performed, and the critical source areas (CSA) with high risks of NPS pollution and high path-through rates were identified (Figure 6). The results demonstrated that Dage, Wudaoying, and Tuchengzi were critical TN source areas with path-through load intensities of 4.31 kg/ha, 4.23 kg/ha, and 2.95 kg/ha, respectively. From the perspective of the path-through pollutant load, Dage, Wudaoying, and Tuchengzi only accounted for 22.09% of the studied area. Nevertheless, they contributed 57.16% of the TN amount discharged into water bodies.



Figure 6. Spatial distribution characteristics of the path-through rate load of NPS pollutants.

The upper and middle reaches of the studied watershed, namely Dage, Wudaoying, Heishanzui, and Tuchengzi, were CSAs of TP, where the path-through load intensity was 0.03–0.008 kg/ha. These four towns contributed 1.37 t, 0.24 t, 0.25 t, and 0.28 t in 2014, accounting for 67.69% of the path-through pollutant load of TP.

In summary, Dage, Wudaoying, and Tuchengzi should be considered for the placement of conservation practices for the effective and efficient implementation of watershed management.

4. Conclusions

(1) The simulation accuracy of the PTRFFM model was acceptable. On the watershed scale, the TN and TP NPS pollutant loads from agriculture were 705.11 t and 3.16 t, respectively, in 2014. Meanwhile, the relative errors of the simulations were 19.62% and 24.45%, respectively. From the spatial distribution of the agricultural NPS, the TN and TP resource loads were mainly distributed among the upper reach towns of Dage, and lower reaches of Taishitun, Bakshiying, and Gaoling. The major source of TN load was farmland, which accounted for 47.6% of the TN load, followed by livestock (37.4%). As for the TP load, rural living was the primary source (65%). The path-through rate of the agricultural NPS pollutants was primarily distributed in Wudaoying, Dage, Tuchengzi, Anchungoumen, and Huodoushan, where the path-through rate of TN ranged from 0.17 to 0.26. However, the priority areas for the path-through rate of TP were distributed among Wudaoying, Kulongshan, Dage, and Tuchengzi, ranging from 0.012 to 0.019. A comprehensive analysis of the distribution of the NPS pollution load and the path-through rate was performed. The results indicated that Dage, Wudaoying, and Tuchengzi were the CSA of agricultural NPS pollution; therefore, they should be considered for effective watershed management.

(2) The spatial distribution characteristics of the path-through rate in the field scale could be obtained. Compared with other methods of estimating the path-through rate for NPS pollutants, such as field monitoring, the ECM, and the physical-based model, the PTRFFM could assess the path-through rates of NPS pollutants in the whole field, watershed, or sub-watershed. It was also able to accurately estimate the watershed path-through rate per unit area of spatial coordinates in the field scale.

(3) The results can be scaled to watersheds with larger scales. Unlike the "black box" or "gray box" models, which facilitate the increase or decrease of the measurement parameters to reflect regional differences when applied to different regions, the five-factor model regards the transport pathways of NPS pollutants by obtaining a functional relationship between the corresponding typical, natural, and geographic parameters of the locations. Thus, the results of the five-factor model can be scaled to obtain the path-through rates for larger watersheds.

In this study, the PTRFFM was developed by considering the pollutant production, surface runoff generation, leaching potential of soil moisture, and landscape interception in the given watershed. The model can be used as an effective tool to identify critical paths for NPS contamination transportation. In the future, to expand this method further, supplementary field monitoring should be performed. For example, the saturated water holding capacity of different soil types should be monitored to decrease the uncertainty of soil infiltration capacity differences among different areas.

Future research is also required to establish new criteria and more efficient optimization techniques to identify the key parameters of NPS pollutants, such as the time lag index [26], and incorporate them into the PTRFFM to realize a better representation of the whole process of NPS pollutant transfer.

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