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Abstract: In recent years, freshwater resource contamination by non-point source pollution has become particularly prominent in China. To control non-point source (NPS) pollution, it is important to estimate NPS pollution exports, identify sources of pollution, and analyze the pollution characteristics. As such, in this study, we established the modified export coefficient model based on rainfall and terrain to investigate the pollution sources and characteristics of non-point source total nitrogen (TN) and total phosphorus (TP) throughout the Huangqian Reservoir watershed—which serves as an important potable water source for the main tributary of the lower Yellow River. The results showed that: (1) In 2018, the non-point source total nitrogen (TN) and total phosphorus (TP) loads in the Huangqian Reservoir basin were 707.09 t and 114.42 t, respectively. The contribution ratios to TN export were, from high to low, rural life (33.58%), farmland (32.68%), other land use types (20.08%), and livestock and poultry breeding (13.67%). The contribution ratios to TP export were, from high to low, rural life (61.19%), livestock and poultry breeding (21.65%), farmland (12.79%), and other land use types (4.38%). The non-point source pollution primarily originated from the rural life of the water source protection zone. (2) Non-point source TN and TP pollution loads and load intensities showed significantly different spatial distribution patterns throughout the water source protection area. Specifically, their load intensities and loads were the largest in the second-class protected zone, which is the key source area of non-point source pollution. (3) When considering whether to invest in agricultural land fertilizer control or rural domestic sewage, waste, and livestock manure pollution control, the latter is demonstrably more effective. Thus, in addition to putting low-grade control on agricultural fertilizer loss, to rapidly and effectively improve potable water quality, non-point source pollution should, to a larger extent, also be controlled through measures such as establishing household biogas digesters, introducing village sewage treatment plants, and improving the recovery rate of rural domestic garbage. The research results discussed herein provide a theoretical basis for formulating a reasonable and effective protection plan for the Huangqian Reservoir water source and can potentially be used to do the same for other similar freshwater resources.

**Keywords:** drinking water source; total nitrogen; total phosphorus; non-point source pollution; temporal and spatial distribution; control plan

## 1. Introduction

Rapid population growth and industrial development have created a serious pollution problem that has drastically compromised the safety of potable water sources in China. In particular, eutrophication—which results from excessive nitrogen and phosphorus in the reservoir's water source—is a significant problem [1–3]. Recently, eutrophication has destroyed the ecological environment and deteriorated the water quality within the water



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). source areas, thereby threatening the health of an extremely large populace [4]. Given that extensive efforts go toward continuous point source pollution control, potable water sources in China are much more affected by non-point source pollution (NPSP)—which has become a major issue that needs to be urgently addressed [5,6]. Currently, more than 60% of China's lakes have eutrophication problems, and more than 50% of the lakes' nitrogen and phosphorus originate from non-point source pollution [7,8]. For example, Lakes Chaohu [9], Taihu [10], and Dianchi [11] are typical reservoirs in China that demonstrate the water eutrophication problems caused by non-point source pollution. They are also important potable water sources; thus, contamination by non-point source pollution seriously threatens the health of the communities they supply. As such, prevention and management of non-point source pollution for the purpose of protecting potable water sources have garnered significant attention from the scientific community; characterizing non-point source pollution is the foundation of non-point source pollution management [12].

Huangqian Reservoir is located in the Dawen River, which serves as the main tributary of the lower Yellow River. It is an important potable water source, as it supplies drinking water to one million people in Tai'an City, Shandong Province, China. The Huangqian Reservoir Basin is home to 850,000 people. Instead of being safely collected and disposed of, most of the residential garbage, sewage, and waste residue in the basin has been directly dumped or abandoned on both sides of the river. Furthermore, relative to surrounding areas, the basin supports a higher percentage of livestock/poultry breeding, but has a smaller number of biogas digesters; thus, animal manure is not effectively treated. In addition, the fruit and crop planting areas account for more than 40% of the total watershed area. Thus, annual chemical fertilizer and pesticide application is 8000 t and 300 t, respectively, while the utilization rate of both is only 30–40%. Compounding the problem is the fact that 55.3% of the basin area has serious soil erosion. Forests, fruit and crop planting areas, and rural residential land experience the most serious soil and water loss. As a result, most of the rural domestic pollution, livestock/poultry waste, and chemical fertilizers and pesticides enter the water source via rainfall erosion and acutely pollute the receiving water body. The monitoring section of Huangqian Reservoir implemented the "Surface Water Environmental Quality Standards" (GB3838-2002) III standard. According to the water quality monitoring data from 2010 to 2019, the detection of relevant units during the rainy season has shown that the TN and TP in the Huanggian Reservoir's water source drastically exceed the standard, and the annual average values of total nitrogen and total phosphorus exceeded this by 5.67- and 3.25-times, respectively. Furthermore, eutrophication was also observed. For all these reasons, non-point source pollution in the Huangqian Reservoir water source has attracted significant attention.

Model simulation is an important research tool for analyzing non-point source pollution characteristics on a river basin scale [13]. Currently, the simulation models can be divided into two categories: the mechanical model and the empirical model. The mechanical models—such as SWAT [14,15], AnnAGNPS [16,17], and HSPF [18,19]—and the empirical models—such as the export coefficient model [20]—are widely used in the study of non-point source pollution. Cai et al. [21] adopted an improved export coefficient model considering rainfall and pollutant migration loss to estimate the total nitrogen pollution load of non-point sources in the Weihe River Basin. Compared to the traditional export coefficient model, it is more realistic. Cheng et al. [22] used the improved export coefficient model to estimate non-point source phosphorus pollution risks under complex precipitation and terrain conditions, and the simulation results were satisfactory. However, the mechanical model requires a very large quantity of complex data for accurate results [23]. Thus, from a practicality perspective, the empirical model, and particularly the export coefficient model, is preferred, as it is not affected by the lower quantity or simplicity of the data and has been widely used due to its high simulation accuracy [24].

Therefore, in this study, the modified output coefficient model was used to consider the rainfall and topographic factors, in order to analyze the non-point source TN and TP pollution characteristics in the Huangqian Reservoir watershed. The objectives of this investigation were to: (1) establish an environmental database of the Huangqian Reservoir's potable water source region for non-point source pollution (NPSP) load estimation; (2) estimate the Huangqian Reservoir watershed's non-point source (NPS) TN and TP load loss in the potable water source protection region using the modified export coefficient method; (3) determine the pollution sources and spatial distribution characteristics of non-point source (NPS) TN and TP throughout the Huangqian Reservoir watershed; (4) analyze the effect of implementing the "China's Battle Plan for Agricultural and Rural Pollution Control in Shandong Province (2018–2020)" on reducing non-point source (NPS) TN and TP in the Huangqian Reservoir watershed.

# 2. Materials and Methods

## 2.1. Study Area

Huangqian Reservoir Basin is comprised of four river systems: the Mata, Xiagang, Shiwuzhi, and Honghe Rivers. The Huangqian Reservoir's water source is the main tributary of the lower Yellow River (i.e., the Dawen River), which is located at 117°4′–117°22′ E and 36°16′–36°28′ N and covers a total control basin area of 292 km<sup>2</sup> (Figure 1a,b).The elevation throughout the water source shows significant variability, with an average altitude of 530 m. The region is characterized by a warm temperate, semi-humid, continental monsoon climate, with an annual average temperature of 13 °C. The average annual precipitation is 758 mm, most of which is concentrated in summer. The Huangqian Reservoir Basin has a total population of 850,000, with an industrial sector comprising primarily agriculture, forestry, and livestock farming (Figure 1d). Huangqian Reservoir is an important potable water source for Tai'an City, as it supplies 1.2 million m<sup>3</sup> of water to the city each day and irrigates 65 km<sup>2</sup> annually.



**Figure 1.** Geographical location of Huangqian Reservoir's potable water source. (**a**) Location of the potable water source in China; (**b**) Water system and digital elevation of the water source; (**c**) The division map of water source protection zone;(**d**) Land use map of water source areas in 2018.

To better protect the water quality of potable water sources, the Huangqian Reservoir Basin is divided into the first-class protected zone, second-class protected zone, and quasi-protected zone (Figure 1c). Herein, non-point source pollution characteristics in the Huangqian Reservoir Basin are systematically analyzed by using sub-basins and water source protection zones as control units.

## 2.2. Data Sources

Land use, meteorological, water quality and quantity, digital elevation, and social and economic data associated with the Huangqian Reservoir water source were obtained and used throughout the course of this investigation. Each data type's uses and sources are listed in Table 1. As shown, land use and socioeconomic data were mainly used for pollution source analysis. Rainfall and digital elevation data were employed to analyze the model's rainfall and terrain influence factors, respectively. Water quality and quantity data were primarily used to correct the modified export coefficient model.

Data Type	Data Type Resolution /Coverage Range		Data Sources	
Land use data	$30 \times 30 \text{ m}$	Land use types and pollution source analysis	Chinese Academy of Sciences Resource and Environment Data Center	
Meteorological data	Daily rainfall from five rainfall stations in Pengjiayu, Ximata, Qincun, Huangqian Reservoir, and Xiagang—2010 to 2018	Rainfall influence analysis	Tai'an Hydrological Center; China Meteorological Data Network	
Water quality and quantity data	Runoff and water quality data for 2018	Model calibration	Tai'an Hydrological Center; Huangqian Reservoir Management Bureau	
Digital elevation data (DEM)	30 m	Terrain impact analysis	China Geospatial Data Cloud	
Socioeconomic data	Huangqian reservoir water source area	Livestock breeding and rural life pollution analysis	Daiyue District Statistical Yearbook of Tai'an City, Shandong Province (2018)	

Table 1. Data and source.

#### 2.3. The Modified Export Coefficient Model and Export Coefficient Determination

2.3.1. The Modified Export Coefficient

The modified export coefficient model has been widely used in estimating non-point source pollution loads [23]. In recent years, experts and scholars have improved the traditional export coefficient model by introducing the rainfall influence coefficient, watershed loss coefficient, and the rainfall and slope index [21,25–27]. The results showed that the modified export coefficient model, which was based on regional topography and hydrological conditions, was more representative of actual non-point source pollution. The precipitation influence and the terrain influence factors were introduced to improve the traditional export coefficient model. The model is mathematically summarized as follows:

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

where *L* is the loss of nutrients; the parameter  $\alpha$  is the rainfall impact factor;  $\beta$  is the topographic impact factor;  $E_i$  is the export coefficient (i.e., annual load per unit area) for the nutrient source *i*;  $A_i$  is the area of land use type *i* or the number and population of livestock type *i*;  $I_i$  is the first nutrient input; *P* is the nutrient input of rainfall:

## (1) Precipitation impact factor $\alpha$

The rainfall is the driving force behind non-point source pollution. Differences in rainfall duration, intensity, and spatial distribution can affect non-point source pollution by changing the watershed's runoff behavior. However, the traditional export coefficient model does not sufficiently represent the rainfall factors' influence on non-point source pollution [23]. Thus, in this study, the influence of rainfall on the dissolved pollutant load was considered from the perspectives of rainfall interannual differences  $\alpha_t$  and spatial distribution  $\alpha_s$ .

$$\alpha = \alpha_t \bullet \alpha_s = \frac{L(r)}{L(r_{ave})} \bullet \frac{r_j}{r_{ave}}$$
(2)

where L(r) is the non-point source pollution storage of annual rainfall r;  $L(r_{ave})$  is the non-point source pollution storage of multi-year average rainfall  $r_{ave}$ ;  $r_{ave}$  is the average annual rainfall in the basin;  $r_i$  is the annual rainfall for space unit j.

According to the 2010–2018 hydrological and water quality data for the Huangqian Reservoir Basin, the Huangqian Reservoir showed a correlation between the annual average rainfall (*r*) and the storage capacity of dissolved non-point source pollutants (*L*) (Figure 2). The TN and TP correlation coefficients were 0.7511 and 0.9110, respectively. The annual average rainfall in the basin from 2010 to 2018 was 721.51 mm. Based on the correlation between rainfall and TN/TP storage, the annual average storage loads of TN and TP were 307.89 t and 1.76 t, respectively. Based on the average rainfall and its spatial distribution, the spatial Kriging interpolation method was employed to obtain the rainfall influence factors on the TN and TP load in 2018. As shown in Figure 3, the values were 1.083~1.242 and 1.216~1.393, respectively. These results showed that the overall spatial distribution of TN and TP rainfall influencing factors decreased from northeast to southwest;



**Figure 2.** Relationship between the rainfall and the inflow of total nitrogen (TN) and total phosphorus (TP) from non-point source pollution.



Figure 3. Spatial distribution of rainfall influencing factors.

(2) Terrain influence factor  $\beta$ 

Non-point source pollutants primarily migrate as rainfall runoff. Thus, the watershed slope is the main influencing factor. Changes in slope can impact the quantity of slope runoff and, in turn, the loss of non-point source pollutants. Studies have shown that runoff can be expressed as the product of the slope and a constant power function. As such, the

influence of slope on TN and TP can be expressed as the relationship between slope and runoff [28,29] as follows:

$$\beta = \frac{L(\theta_j)}{L(\theta_{\text{ave}})} = \frac{c \cdot \theta_j^a}{c \cdot \theta_{\text{ave}}^d}$$
(3)

where  $\theta_j$  is the slope of sub-watershed *j*;  $\theta_{ave}$  is the average basin slope; *c* and *d* are constants.

In this study, the values of *c* and *d* were 0.1881 and 0.6104 [30], respectively, and  $\theta_{ave}$  was 13.17°. The topographic impact factor of the water source protection area of Huangqian Reservoir was determined to be 0.579~1.222 by spatial Kriging interpolation (Figure 4).



**Figure 4.** Spatial distribution of terrain impact factor  $\beta$ .

#### 2.3.2. Determination of Export Coefficient Values

The pollution sources were divided into three types: land use, livestock breeding, and rural domestic life. Because of differences in rainfall, terrain, and land use type, the export coefficient for each pollution source type will be unique based on the local conditions [31–34], and therefore differ from one region to another. In this work, the export coefficient values associated with farmland, forest land, grassland, residential land, and other land uses were calculated by averaging the corresponding values from six regions: Miyun Reservoir [12], Jiaozhou Bay [35], Zhangjiakou Qingshui River upstream [36], Shandong Nansi Lake Basin [37], Liaoning Hun River Basin [38], and Yimeng Mountain Yunmeng Lake [39].

In this study, livestock and poultry were divided into four types: bovine, pig, sheep, and poultry. The export coefficient for livestock and poultry breeding was determined by calculating the excretion coefficient of livestock and poultry manure, the average content of pollutants in live livestock and poultry manure, and the transport rate of livestock and poultry manure to water [40].

According to the 2019 China Water Resources Bulletin and the actual situation of Huangqian Reservoir, the per capita domestic water consumption of rural residents in Huangqian Reservoir Basin was 85 L/d, and the sewage production rate was 30%. The monthly average concentrations of TN and TP in domestic sewage were 55 mg/L and 7 mg/L, respectively, and the loss coefficient was 90%. Therefore, the export coefficients of TN and TP in domestic sewage were 0.4607 kg/a and 0.0587 kg/a, respectively. The garbage generated by each rural resident in Huangqian Reservoir Basin was 144 kg/a,

and the TN and TP in each kilogram of garbage were 34.05 g and 11.7 g, respectively. According to the actual survey, the waste loss rate in this region was 20%. Therefore, the export coefficients of TN and TP in rural domestic waste were 0.9806 kg·person<sup>-1</sup>·a<sup>-1</sup> and 0.3370 kg·person<sup>-1</sup>·a<sup>-1</sup>, respectively. According to the survey, the annual feces and urine output per person in the region were 91.25 kg and 365 kg, respectively. The nitrogen and phosphorus contents in feces were 1.16% and 0.53%, respectively, and the nitrogen and the phosphorus contents in urine were 0.5% and 0.1%, respectively. In this study, the loss coefficient of feces was 20%, and the loss coefficient of urine was 50%. Therefore, the export coefficients of domestic feces and urine in the region were 1.1242 kg·person<sup>-1·a<sup>-1</sup></sup> and 0.2792 kg·person<sup>-1·a<sup>-1</sup>, respectively. The export coefficient of rural life in Huangqian Reservoir Basin was basically consistent with the research results in certain parts of the country [41–45] (Table 2).</sup>

Pollution Source	Unit	Category	TN Export Coefficient	TP Export Coefficient
	t∙km <sup>-2</sup> •a <sup>-1</sup>	Farmland	2.302	0.13
Land use		Forestland	0.434	0.015
		Grassland	1.152	0.035
		Residential land	1.296	0.032
		Water area	1.500	0.036
Livestock/ poultry breeding	kg∙head (only) <sup>-1</sup> ∙a <sup>-1</sup>	Bovine	16.412	1.204
		Pig	1.208	0.243
		Sheep	0.37	0.132
		Poultry	0.023	0.01
Rural life -	$kg \cdot a^{-1}$	Domestic sewage	0.4607	0.0587
	kg·person <sup>-1</sup> ·a <sup>-1</sup>	Domestic waste Feces, urine	0.9806 1.1242	0.337 0.2792

**Table 2.** Export coefficients of non-point source pollutants in the Huangqian Reservoir's potable water source area.

#### 2.4. Non-Point Source Pollution Load Estimation and Model Validation

The calculated value of the export coefficient model represents the number of pollutants lost to the surrounding environment. However, due to vegetation interception and natural sedimentation, only a fraction of the pollutants lost to the surrounding environment enter the water body. The inflow coefficient is a ratio that relates the concentration of pollutants entering the water body to the concentration of lost pollutants. The inflow coefficient mainly depends on the underlying surface conditions and the distance of pollutants to the nearby river. Determining a reasonable inflow coefficient is key to accurately estimating the non-point source dissolved pollutant inflow [45]. The inflow coefficient value in this work refers to the relevant research results— $\lambda_{TN} = 0.3$  and  $\lambda_{TP} = 0.2$ . In 2018, the non-point source TN simulation value in the Huangqian Reservoir was 212.13 t, while the measured value was 238.80 t—a relative error of 11.17%. The non-point source TP simulation value was 22.884 t, while the measured value was 19.35 t—a relative error of 18.26%. The results showed that when based on the Huangqian Reservoir's rainfall terrain, the modified export coefficient model is superior to that of the traditional model and can be used to analyze the non-point source pollution characteristics in the Huangqian Reservoir water source.

#### 3. Results and Discussion

# 3.1. Source Analysis of Non-Point Source Total Nitrogen and Total Phosphorus Pollution in the Watershed

The modified export coefficient model was used to determine the sources of nonpoint TN and TP pollution loads in the Huangqian Reservoir watershed, and the results are shown in Table 3. Obviously, there were significant differences in non-point source pollution load and load intensity among different land use types. The non-point source TN and TP load (231.08 t and 14.63 t, respectively) and load intensity (2.53 t  $(km^2 \cdot a)^{-1}$  and 0.16 t  $(km^2 \cdot a)^{-1}$ ) of farmland were significantly higher than those of other land use types. With respect to livestock/poultry breeding, non-point source TN pollution load was mainly produced by cattle (39.28 t) and sheep (28.20 t), while TP pollution load was mainly produced by sheep (11.28 t) and poultry (6.87 t). Interestingly, the proportion of rural domestic life TN and TP pollution sources in the watershed was relatively large and originated from feces and urine (104.04 t and 28.96 t, respectively), as well as domestic waste (90.75 t and 34.96 t, respectively).

**Table 3.** Main sources of non-point source pollutants in the Huangqian Reservoir's potable water source area.

Pollution	Category	TN		ТР			
Source		Load/t∙a <sup>-1</sup>	Load Percentage/%	Load Intensity /t·(km <sup>2</sup> ·a) <sup>-1</sup>	Load/t∙a <sup>-1</sup>	Load Percentage/%	Load Intensity /t·(km <sup>2</sup> ·a) <sup>-1</sup>
	Farmland	231.08	61.94	2.53	14.63	74.47	0.16
	Forestland	76.83	20.60	0.52	3.01	15.33	0.02
T 1	Grassland	32.16	8.62	1.36	1.10	5.62	0.05
Land use	Waters	8.17	2.19	1.26	0.22	1.12	0.03
	Residential land	24.81	6.65	1.40	0.68	3.47	0.04
	Total	373.05	100.00	7.07	19.64	100.00	0.30
	Cattle	39.28	40.65	-	3.23	13.04	-
Livestock	Pig	15.05	15.57	-	3.39	13.69	-
and poultry breeding Rural life	Sheep	28.20	29.18	-	11.28	45.54	-
	Poultry	14.10	14.59	-	6.87	27.74	-
	Total	96.63	100.00	-	24.77	100.00	-
	Domestic sewage	42.64	17.96	-	6.09	8.70	-
	Domestic garbage	90.75	38.22	-	34.96	49.94	-
	Stool and urine	104.04	43.82	-	28.96	41.37	-
	Total	237.43	100.00	-	70.01	100.00	-

In 2018, the non-point source TN and TP pollution loads in the Huangqian Reservoir water source were 707.09 t and 114.42 t, respectively. The non-point source TN pollution load generated by rural life was the highest (237.43 t), accounting for 33.58%. Non-point source TN pollution load of farmland ranked second, reaching 231.08 t, accounting for 32.68%. Livestock/poultry breeding produced the lowest amount of the non-point source TN pollution load (96.63 t) and accounted for 13.67%. The maximum non-point source TP pollution load originated from rural domestic life (70.01 t) and accounted for 61.19%. Livestock/poultry breeding came in second, with a non-point source TP pollution load of 24.77 t, which accounted for 21.65%. Farmland produced the lowest amount of non-point source TP pollution load (14.63 t) and accounted for 12.79%. Other land use types contributing to non-point sources TP pollution accounted for 4.38% (Figure 5).

In summary, rural life was the largest contributor to non-point pollution within the water source protection area.



**Figure 5.** Contribution of non-point source total nitrogen (TN) and total phosphorus (TP) load source in the Huangqian Reservoir's potable water source area.

## 3.2. Spatial Distribution Characteristics of Non-Point Source Pollution in the Watershed

In this work, the spatial distribution maps of TN, TP load, and load intensity were obtained by the ARC GIS 10.6 software. The TN and TP pollution loads differed significantly between the sub-basins. In 2018, the non-point source TN and TP pollution loads in the basin were 2.73–82.76 t $\cdot a^{-1}$  and 0.11–13.91 t $\cdot a^{-1}$ , respectively (Figure 6). Pollution loads were largest in the second-class protected zone. The order of the non-point source TN and TP loads from large to small were as follows: the second-class protected zone, the quasi-protected zone, and the first-class protected zone. The non-point source pollution load intensity represents the pollutant load per unit land area—an important indicator for analyzing land pollution. Non-point source pollution load intensity is related to various factors—such as basin area, population density, and industrial structure. The analysis of non-point source pollution in the water source area showed a TN and TP load intensity of  $0.69 \sim 4.99$  t·(a·km<sup>2</sup>)<sup>-1</sup> and  $0.04 \sim 1.10$  t·(a·km<sup>2</sup>)<sup>-1</sup>, respectively. As depicted in Figure 7, the largest load intensities of NPS total nitrogen (TN) and total phosphorus (TP) were mainly distributed in the second-class protected zone, which is the middle of the basin. Non-point source TN and TP pollution load intensities in the second-class protected zone were 2.83 t  $(a \cdot km^2)^{-1}$  and 0.46 t  $(a \cdot km^2)^{-1}$ , respectively. The coverage of forest grassland reached more than 60% in the upper reaches of the Mata River and the Xiagang River, while there were few rural residents, and the region was the quasi-protected zone of the potable water source. Thus, non-point source total nitrogen and total phosphorus pollution loads and load intensities were small.



Figure 6. Spatial distribution of total nitrogen (TN) and total phosphorus (TP) pollution load.



Figure 7. Spatial distribution of total nitrogen (TN) and total phosphorus (TP) pollution load intensity.

Furthermore, the Huangqian Reservoir's water source second-class protected zone was the key non-point source pollution source.

## 3.3. Analysis of Watershed Non-Point Source Pollution Load Control Scheme

Based on the above analysis, the main factors contributing to high non-point source pollution in the Huangqian Reservoir water source included a low recovery rate of rural domestic sewage and garbage, domestic feces and urine, and large-quantity application of chemical fertilizers and pesticides on agricultural land.

According to the requirements of "China's Battle Plan for Agricultural and Rural Pollution Control in Shandong Province (2018–2020)", prior to 2020, the province's rural livestock/poultry manure treatment and utilization rate was 90%, rural domestic sewage treatment utilization rate was 63%, and rural domestic waste and manure pollutants comprehensive utilization rate was 81%. Furthermore, the province implemented the fertilizer reduction and efficiency project to control pollution from farmland fertilizer loss. The project entailed promoting in-depth soil testing and formula fertilization, comprehensively implementing cultivated land quality monitoring, improving the fertilization formula accuracy, and increasing the fertilizer utilization rate to more than 40%. Studies have shown that a 30% reduction of the fertilizer application will not significantly impact crop yields for wheat, maize, and other crops, but it will reduce yields for chestnut, cherry, walnut, and other fruit trees to a certain extent [46]. In the Huangqian reservoir water source protection area, the coverage area of fruit trees is about three-times that of crops. As such, a 15% reduction in chemical fertilizer was selected as a realistic control for agricultural land. Thus, to determine the most effective control scheme for the basin in its entirety, three non-point source pollution control schemes were evaluated: Scheme 1---only animal breeding and domestic rural life pollution were controlled; Scheme 2-only agricultural pollution was controlled; Scheme 3-animal breeding, domestic rural life, and agricultural pollution were controlled (Table 4).

Table 4. Evaluated non-point source pollution control schemes.

Scheme	Fertilizer Application	Rural Livestock and Poultry Manure Utilization Rate	Rural Domestic Sewage Utilization Rate	Domestic Waste and Fecal Pollutant Utilization Rate
Present Situation (S0)	Unchanged	Unchanged	Unchanged	Unchanged
Scheme 1 (S1)	Unchanged	Reached 90%	Reached 63%	Reached 81%
Scheme 2 (S2)	Reduced 15%	Unchanged	Unchanged	Unchanged
Scheme 3 (S3)	Reduced 15%	Reached 90%	Reached 63%	Reached 81%

The results of each scheme, i.e., the pollution load reduction in the Huangqian Reservoir water source protection area, were analyzed and compared (Figure 8). As shown in Figure 8, pollution reduction due to the implementation of Scheme 3 (S3) was the most remarkable. The overall TN and TP storage pollution load decreased by 42.92% and 68.97%, respectively. Specifically, the TN and TP storage pollution load in the first-class protected area decreased by 38.21% and 72.41%, respectively, in the second-class protected area by 45.77% and 69.80%, respectively. The TN and TP inflow pollution load in the quasi-protected areas decreased by 40.41% and 68.13%, respectively.



**Figure 8.** Quantity of non-point source total nitrogen (TN) and total phosphorus (TP) pollution for different control schemes.

Controlling the rural domestic life exclusively and livestock/poultry breeding pollutants (S1) can reduce the TN and TP inflow by 37.86% and 66.95%, respectively, while only controlling chemical fertilizer loss in agricultural land (S2) can reduce the TN and TP inflow by 5.07% and 2.01%, respectively. Clearly, rural domestic life and livestock pollution control are far more effective than land use and soil erosion control. Although chemical fertilizer loss from agricultural land accounts for a large proportion of the pollution load within the water source area, it is difficult to control chemical fertilizer pollution for the following reasons: (1) the planting structure within the study area is difficult to change; (2) reducing the amount of chemical fertilizer use may impact the agricultural yield; (3) the basin is prone to acute soil erosion; (4) chemical fertilizer utilization efficiency is low due to the uniqueness of the geographical environment.

In recent years, ecological villages have been constructed in the Huangqian Reservoir water source protection area. Within these villages, the concept of "changing water, changing toilets, changing circles" has been comprehensively promoted; household biogas pools have been established; scientific composting studies have been conducted; resource utilization efficiency has been improved. Part of the village domestic sewage was treated by biogas digesters, and the rest was collected and treated by other means. In addition, domestic waste management within the water source protection area has gradually increased, resulting in an improved waste treatment system that includes "household collection, village collection, town transportation, and county treatment." Thus, when considering whether to invest in agricultural land fertilizer control or rural domestic sewage, waste, and livestock manure pollution control, the latter is demonstrably more effective.

# 4. Conclusions

Non-point source pollution has become an important source of pollution in drinking water, and it is one of the major environmental problems facing the world. Excessive N, P, and other non-point source pollutants entering drinking water sources are one of the important reasons for eutrophication and harmful algal blooms in water bodies, which seriously threaten the safety of the drinking water environment. In this study, the modified export coefficient method was used to simulate and evaluate NPS TN and TP pollution loads

entering the Huangqian Reservoir potable water source. In summary, the main sources of non-point source pollution in the Huangqian Reservoir potable water source are as follows: (1) rural life (domestic garbage and sewage); (2) agricultural land (large application of chemical fertilizers and pesticides); (3) livestock and poultry breeding (feces and urine of livestock and poultry); (4) other land use types (loss of soil nutrients caused by rainfall erosion). According to the analysis of the non-point source pollution characteristics of the watershed, the non-point source pollution load of the water source is mainly concentrated in the secondary protection area with a large population density and a large proportion of agricultural land. In order to ensure the safety of drinking water, the environmental protection department has launched the "China's Battle Plan for Agricultural and Rural Pollution Control in Shandong Province (2018–2020)". The plan proposes a series of cleaner production measures, such as soil testing, precise fertilization, the establishment of household biogas digesters, township garbage treatment plants, and sewage treatment plants. The paper studied the non-point source pollution control plan of the water source protection area. The results showed that the joint implementation of reducing the application of agricultural and chemical fertilizers and increasing the treatment rate and utilization rate of domestic sewage, garbage, and livestock manure is more effective in the control of non-point source pollution in water sources. This achievement has important practical significance and theoretical value for the comprehensive treatment of non-point source pollution in drinking water source areas.

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