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Key Points:

- Contribution of different drivers to N runoff loss was compared between hills and plains
- Fertilization contributed more to N loss in hills due to N increment of paddy land
- Rainfall contributed more to N loss in plains due to N increment of urban land

Supporting Information:

Supporting Information may be found in the online version of this article.

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Difference in the Contribution of Driving Factors to Nitrogen Loss With Surface Runoff Between the Hill and Plain Agricultural Watersheds

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Abstract Identifying the factors and quantifying their contributions to nitrogen (N) loss associated with surface runoff is of great significance to the control of non-point source N pollution. However, the distinct geographical units, such as hills and plains, may lead to great differences in the contribution of these driving factors, which has been rarely investigated. This study developed an effective framework, which simulated the N loss with surface runoff in hills and plains by SWAT and NDP, and analyzed their spatial distribution variations by spatial autocorrelation analysis, and distinguished the contribution of their driving factors by multi-scenario simulation and partial redundancy analysis (pRDA). The framework was instantiated in a hill and a plain agricultural watershed, respectively, in the upper Taihu Lake Basin, China. We found the contribution of fertilization to N loss with surface runoff in the hills (10.4%) was greater than that in the plains (6.4%), which may be due to the N increment effect of paddy land. The contribution of rainfall to N loss with surface runoff in the plains (93.2%) was greater than that in the hills (74.3%), which may be due to the N increment effect of urban land. The developed framework could provide a viable way to study the environmental impacts of natural and anthropogenic drivers in different types of agricultural watersheds, thus offer scientific references for nutrient control measures.

Plain Language Summary Geographical units within a watershed, such as hills and plains, exhibit distinct characteristics that give rise to variations in the processes of nitrogen (N) runoff loss and their associated driving factors. These variations pose challenges to the development of effective control measures for non-point source N pollution. Consequently, we have developed an effective framework capable of simulating N loss with surface runoff, analyzing variations in their spatial distribution, and discerning the external (e.g., rainfall) and internal (e.g., land management, fertilization) contributions of driving factors in the hills and plains, respectively. A notable discovery was that fertilization played a more substantial role in N loss in hills due to increased N levels resulting from paddy land, whereas rainfall emerged as the primary driver of N loss in plains due to elevated N levels associated with urban land. This research contributes to our understanding of the complexities surrounding N runoff loss and provides valuable insights for the development of targeted control measures within diverse geographical units.

1. Introduction

With the growth of population and the continuous advancement of agriculture and urbanization, land has been highly utilized and fertilizer has been heavily invested to ensure food production. Excessive nitrogen (N) eventually enters to the surrounding water bodies mainly through surface runoff (including drainage), which poses a hidden danger to the sustainable development of water environment and the safety of agricultural products (Zhang, Hou, et al., 2020). On a global basis, humans have more than doubled the amount of N fixation, with highly industrialized and agriculturalized areas experiencing mineralized N concentration up to 25 times that of pre-development concentrations (Alldred & Baines, 2016). Agricultural non-point source N loss accounts for 80%–90% of the total N pollution in water bodies, which mainly occurs in the rainy season (Law et al., 2022; Zhang, Hou, et al., 2020). Therefore, identifying the spatial-temporal dynamics of N loss with surface runoff and its driving factors and contribution will provide a scientific basis for the fine management of watershed N regulation (Vystavna et al., 2023).

Much research has already been undertaken on the driving factors of N loss. For example, Gabriel et al. (2018) found that extreme rainfall or drought was expected to exacerbate current stresses on N loss from population and economic growth, land use and fertilization. However, land use has a more direct impact on N loss than climate through changing N sources and transport processes. In agricultural watersheds, rainfall affects agricultural production systems and also annual and seasonal variability in hydrologic processes, and thereby influences N runoff loss and its spatial distribution (Li et al., 2022; Wade et al., 2022; Wenng et al., 2020). Agricultural land generally produces the largest N load, followed by tea plantations, citrus groves and woodlands (Zhang, Li, et al., 2020). Although urban land accounts for a small proportion in agricultural watersheds, it increases the proportion of impervious surface, reduces land permeability, leads to higher runoff amount and non-point source N load especially during rainy seasons (Alamdari et al., 2022; Vystavna et al., 2023; Zhu et al., 2022). N fertilization can increase crop yield, but cannot be fully absorbed by crops, and then partially enter rivers and lakes through runoff or seepage. In summary, N runoff loss in agricultural watersheds varies among different land use and cropping patterns based on rainfall, surface vegetation, soil property and agricultural management (Ding et al., 2022).

Owing to the gaps in developmental status of each region, the changing trends in land use and fertilization vary from region to region. Moreover, uncertainties exist since it is not known how management practices may adapt to land use and climate change (Wenng et al., 2020). To assess the response of non-point source N pollution discharge to external (e.g., climate) and internal (e.g., water management, land management), the simulation evaluation method based on scenarios analysis was used in different watersheds, including the recent consideration of detailed N transport processes in small (<50 km^2) watersheds and regional and global-scale assessments of total N (TN) load using empirical and process-based models (Alamdari et al., 2022). Hills and plains are the most common and representative landform types in a region, and their hydrological and N transport processes are significantly different. In the hills, the dry and wet conditions change dramatically, which makes it difficult to study the spatial and temporal pattern of water yield and N loss load (Bales et al., 2006; de Jong et al., 2005). The Soil Water Assessment Tool (SWAT) model is one of the best available tools for simulating the response of hill agricultural watersheds to water quality. Majority of studies conducted by the SWAT model focus on how to reduce N pollution through land use changes, as well as regulating fertilizer application rates (Oduor et al., 2023). However, the use of watershed models such as SWAT to simulate the hydrological and N transport processes in the plains is not ideal. The main reason is that the distributed watershed models divide the space into hydrological response units (HRUs), and determines the flow direction according to the elevation difference between these units, while the flow direction in the plains is more susceptible to water level difference. Moreover, most watershed models cannot track the artificial drainage processes such as pumping and culvert drainage, which control the hydrological and material exchange in the plains (Huang et al., 2018). Therefore, the conclusion of performances of driving factors with different watershed characteristics on N loss is still limited.

Aiming at the above knowledge gaps, we proposed a hypothesis: different from the great impact of fertilization in paddy land in the hill agricultural watershed, the impact of urbanization on N loss with surface runoff would be greater in plain agricultural watershed. To test this hypothesis, we developed a model framework for quantifying the N loss with surface runoff in hill and plain agricultural watersheds, and distinguishing the contribution of driving factors to spatial distribution variations of N loss.

2. Framework Development

The framework consists of several major steps as follows: (a) modeling the N loss and runoff processes in the hill agricultural watershed by SWAT model; (b) modeling the N loss and drainage processes in the plain agricultural watershed by NDP model; (c) analyzing the spatial distribution variation of N loss with surface runoff by spatial autocorrelation analysis; (d) quantifying the contribution of rainfall, land use and fertilization to N loss with surface runoff by multi-scenario simulation and partial redundancy analysis (pRDA) (Figure 1).

2.1. Modeling in the Hill Agricultural Watershed

SWAT (Soil and Water Assessment Tool) model was used to simulate N loss with surface runoff in the hill agricultural watershed (Arnold, 1994). The hydrological processes described by the SWAT model were divided into the land surface part (i.e., runoff generation and overland flow) and the water surface part (i.e., flow routing in river channel) of the water cycle. The former determined the amount of water, sand, nutrients and chemicals input

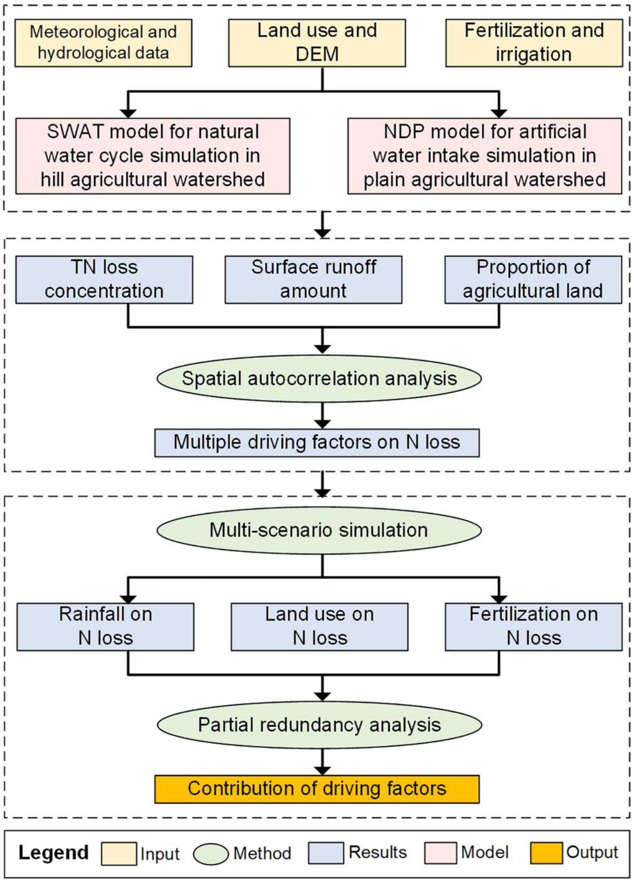

Figure 1. Model framework for identifying the contribution differences of driving factors to N loss with surface runoff between the hill and plain agricultural watersheds.

from each sub-watershed to the main channel; the latter controlled the transport of material from the river network to the outlet of the watershed. Water yield was the total amount of water (i.e., surface runoff, lateral flow, groundwater) leaving the HRUs and entering main channel during the time step. SWAT contained N storage of ammonium N, nitrate N and organic N in the soil. Since land use was reflected at the HRU level, N load of different land use types was extracted from the HRUs. The TN loss load was calculated by multiplying various forms of the N concentration by the water amount in different hydrological pathways (Equation 1, Malago et al., 2017).

$$\Delta TN^T = \Delta TN^T_{Surf} + \Delta TN^T_{Lat} + \Delta TN^T_{GW} \tag{1}$$

where ΔTN^T was the TN loss load (ton); ΔTN^T_{Surf} was the TN loss load due to surface runoff (ton); ΔTN^T_{Lat} was the TN loss load due to lateral flow (ton); ΔTN^T_{GW} was the TN loss load due to groundwater (ton).

2.2. Modeling in the Plain Agricultural Watershed

NDP (Nitrogen Dynamic Polder) model was used to simulate N loss with surface runoff (i.e., drainage) in the plain agricultural watershed (Huang et al., 2018). NDP driven by water level was used to realize the precise characterization of multi-scale and multi-process of non-point source N migration and transformation in the polders. It included five water balance modules and three N dynamic modules using a daily time step. The key augmentations of NDP model involved the representation of artificial drainage, as well as the water interactions among surface water, groundwater, and soil water in farmlands. These two mechanisms were particularly important due to their significant impact on water and N balance in the polders (Equation 2).

$$\Delta TN^T_{Polder} = \Delta TN^T_{Pump} + \Delta TN^T_{Culvert} + \Delta TN^T_{Seep} - \Delta TN^T_{Irr} \tag{2}$$

where ΔTN^T_{Polder} was the TN loss load from the polder (ton); ΔTN^T_{Pump} was the TN loss load due to flood drainage (ton); $\Delta TN^T_{Culvert}$ was the TN loss load due to culvert drainage (ton); ΔTN^T_{Seep} was the TN loss load due to seepage (ton); ΔTN^T_{Irr} was the TN import amount due to irrigation (ton).

2.3. Analyzing the Spatial Distribution Variations

The spatial autocorrelation analysis was used to study the spatial distribution of the classification results by TN concentration and to test whether the attribute value of a unit was related to the attribute value of its adjacent spatial units. The global spatial autocorrelation was performed to summarize the degree of spatial dependence in a total spatial range, which was expressed by Moran's I index; a clearly positive Moran's I indicated that the observed values of factors tend to be spatially aggregated; a clearly negative Moran's I indicated that the observed values of factors tend to be spatially dispersed. Local spatial autocorrelation was thereafter performed to describe the similarity between a spatial unit and its domain. It could indicate the extent to which each local unit followed a global general trend (including direction and magnitude), illustrating how spatial dependence changed with position (Zhao et al., 2019).

The bivariate Local Indicators of Spatial Association (LISA) was used as a straightforward extension of the LISA functionality to two different variables, one for the location and another for the average of its neighbors (Song et al., 2020). In this study, surface runoff amount or the proportion of agricultural land was selected as the x-variable and TN concentration was selected as the y-variable. A clearly positive Moran's I indicated significant positive spatial correlation and a clearly negative Moran's I indicated significant negative spatial correlation. All spatial changes of various factors were analyzed using GeoDa (Anselin, 2004) and ArcGIS 10.7.

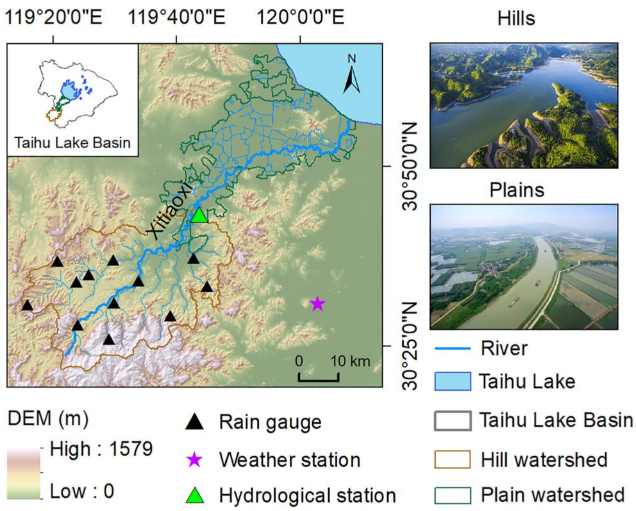

Figure 2. Location of the study area (Xitiaoxi River Basin) and hydrological and weather stations.

2.4. Quantifying the Contribution of Driving Factors

To explore whether land use/management changes have a positive or negative impact on N loss, we examined three land use conversion scenarios (from 20% of paddy land to dry land, urban land and ponds, respectively) and two land management scenarios (reducing 20% and 40% of N fertilizer amount). The land management scenarios setting was based on some studies that proved reducing 20%–40% of N fertilizer amount in the rainy season did not influence the crop production (Xue et al., 2011).

To assess how much of the N loss can be explained by the variations of rainfall, land use and fertilization, the partial redundancy analysis (pRDA) performed by CANOCO 5.0 (ter Braak & Šmilauer, 2012) was used to quantify the unique and shared contributions of above three groups of environmental factors to the total variance. Annual accumulated rainfall, fertilizer amount and the proportion of agricultural land/urban land were selected as the *x*-variables, annual TN load was selected as the *y*-variable. The shared contribution was obtained by subtracting the unique variation of each explanatory variable from the total variation explained by all explanatory variables (Dalu et al., 2017).

3. Framework Application

3.1. Study Area

The Xitiaoxi River Basin is located in the upper reaches of the Taihu Lake with advanced economy and dense population (Figure 2). The average fertilization amount is 353 kg·ha^{-1}, exceeding the international ecological security standard with 225 kg·ha^{-1}. As one of the most important tributaries of Taihu Lake, the Xitiaoxi River is 159 km long, suppling 26.8×10^8 m^3 (27.7%) of water volume to Taihu Lake, furthermore contributing greatly to the nutrient load (Li et al., 2017; Zhou et al., 2013). The terrain is high in the southwest and low in the northeast, with the elevation between 0 and 1,576 m. The area of the hill agricultural watershed (119°14′–119°48′E, 30°23′–30°45′N) is 1,367 km^2. Paddy land, dry land, pond and urban land account for 12%, 13%, 0.1% and 3% of the total area, respectively. The area of the plain agricultural watershed (119°48′–120°9′E, 30°45′–31°5′N) is 869 km^2, which is composed by basic geographical units of 141 polders. The main land use types are paddy land (69%), followed by dry land (14%), urban land (11%) and pond (6%).

The study area belongs to subtropical monsoon climate, with an average annual temperature of 15.5°C and an average annual precipitation of 1,465.8 mm. More than 75% of rainfall occurs in the rainy season (from April to October), which is easily coincident with fertilizer, causing a large amount of farmland N runoff loss (Yuan et al., 2023). In the hill agricultural watershed, runoff is characterized by high intensity and short duration due to their topography. In the plain agricultural watershed, the pumps and ditches reduce the flood peak flow and prolong the hydraulic retention time, providing convenient conditions for the diffusion of N in the agricultural land (Huang et al., 2018).

According to the "Huzhou Water Resources Bulletin", the average annual precipitation in 2013 was 1,220.2 mm, which belonged to the dry year (12.7% less than the normal year); from July to August, there was continuous high temperature and little rain; residents in hills had difficulty in drinking water, meanwhile large areas of crops and freshwater aquaculture in plains suffered from drought. In 2015, the average annual precipitation was 1,675.1 mm, which belonged to the wet year (20% more than the normal year); the Mei-yu period lasted for 35 days, with the rainfall was 55.7% more than the annual; the average water yield coefficient of the watershed was 0.62. The average precipitation in 2017 was 1,334.1 mm, which belonged to the normal year; the average water yield coefficient was 0.48. Therefore, this study selected 2013 (dry year), 2015 (wet year) and 2017 (normal year) as the typical hydrological years.

3.2. Datasets

(1) Spatial data (i.e., DEM, land use and soil) for model construction and scenario setting (Table 1).

Table 1
Data List of Hydrological and N Transport Models for the Hill and Plain Agricultural Watersheds

Type	Indicator	Time series	Spatial and temporal resolution	Source	Model	Usage
Land use	Land use type	2015	30 × 30 m	GLC_FCS30-2015	SWATNDP	Input data
DEM	–	–	90 × 90 m	SRTM DEM	SWAT	Input data
Soil	Soil type	–	1:100 million	HWSD	SWAT	Input data
Meteorology	*Pr*, T_{Max}, T_{Min}, T_{Ave}, *Wet*, *WS* and H_{Sun}	2009–2017	Daily	CMDC	SWATNDP	Forcing data
Hydrology	*Q*	2010–2012	Daily	Hydrological station (Hengtangcun)	SWAT	Calibration data
	WL	2015–2016	Daily	Water level logger	NDP	
	Irrigation and flood drainage	2009–2017	Daily	Surveying	NDP	Forcing data
Water quality	*TN, TDN, NO, NH*	2010–2012	Monthly	Water quality monitoring station (Hengtangcun)	SWAT	Calibration data
		2014–2016	Monthly	Water sampling	NDP	Input data, calibration data
Fertilization	Fertilization amount, Fertilization date	–	–	Surveying	SWATNDP	Input data

Note. Pr: daily precipitation (mm); T_{Max}, T_{Min} and T_{Ave}: daily maximum, minimum and average air temperature, respectively (°C); *Wet*: daily average humidity (%); *WS*: daily average wind speed (m s^{-1}); H_{Sun}: daily sunshine hours (h); *WL*: water level (mm); *TN*: total nitrogen concentration (mg L^{-1}); *TDN*: total dissolved nitrogen concentration (mg L^{-1}); *NO*: nitrate nitrogen concentration (mg L^{-1}); *NH*: ammonia nitrogen concentration (mg L^{-1}).

DEM reflected the terrain characteristics of hills (CGIAR-CSI, 2013), therefore it was an important basis for SWAT model to extract water system and to divide sub-watersheds. The land use data was extracted from the GLC_FCS30-2015 global land-cover products (Liu et al., 2020), which was divided into forest land, grass land, paddy land, dry land, pond and urban land. The soil data was constructed using Harmonized World Soil Database (HWSD, FAO and International, 2019), which were generalized into 6 categories and 9 races, including high active strong acid soil (*Plinthic Alisols*, *Haplic Alisols*, *Ferric Alisols*), bottom active strong acid soil (*Humic Acrisols*), embryonic soil (*Ferralic Cambisols*, *Dystric Cambisols*), high active leaching soil (*Haplic Luvisols*), loose lithologic soil (*Dystric Regosols*) and artificial soil (*Cumulic Anthrosols*).

(2) Meteorological data for model input.

The temperature, solar radiation, wind speed, relative humidity and precipitation were derived from the China Meteorological Data Service Centre (CMDC, 2024). Precipitation had a great impact on runoff, and improving the density of precipitation data was very important for simulation accuracy. Therefore precipitation data of 13 rainfall stations in Tianjintang, Fendai, Hanggai, Shuangshe, Fushi Reservoir, Zhangli, Bingkeng, Laoshikan Reservoir, Xiaofeng, Yinkeng, Licun, Dipu and Hengtangcun were used (Figure 2).

(3) Hydrologic and water quality data for model boundary and model calibration.

The SWAT model was verified by the daily runoff from 2009 to 2012 and the monthly TN load from 2010 to 2012 of Hengtangcun Hydrological Station. The NDP model was verified by the daily water level from 2015 to 2016 measured by the water level recorder (HOBO U20), and the monthly TN concentration from 2014 to 2016 by field sampling and analysis. The drainage and irrigation data of the polders were obtained from water volume and pumping time recorded by the pumping stations.

(4) Agricultural management data for model input.

The agricultural land in our study area was mainly paddy land, and the rest was less than 1/6 of the paddy land. Therefore, only the agricultural management information of paddy land was edited in the models (Table SI-1 in

Supporting Information S1). According to the survey results, the average fertilization rate was set to 50 kg urea/acre, equivalent to pure N 345 kg/ha (Zhu, 2014).

3.3. Configuration

To develop the SWAT model, the hill agricultural watershed was divided into 45 sub-watersheds (minimum watershed area threshold: 2,000 ha) according to the river network and the outlet; the river network was generated based on DEM; the Hengtangcun Station was defined as the total outlet of the watershed. The 45 sub-watersheds were further divided into 4,993 HRUs according to the consistency of soil type and surface coverage. The information of point source, reservoir, agricultural management (sowing, irrigation, fertilization and crop harvesting) was input according to the actual situation. The water yield, sediment yield and nutrient load on each HRU were calculated separately, furthermore the water balance and total nutrient load of the whole watershed were obtained. The global sensitivity analysis was used to screen the most influential runoff and N-related parameters. Nash-Sutcliffe efficiency coefficient (*NSE*), percentage bias (*PBIAS*) and coefficient of determination (R^2) were used to evaluate the goodness of fit of SWAT model for runoff and TN simulation. SWAT model can well capture the seasonal variation trend of discharge and flood peak flow in heavy rainfall events, with the *NSE* between simulated and measured discharge and TN loss were 0.81 and 0.67, respectively. The detailed information of sensitivity parameter analysis processes as well as model calibration and validation processes of SWAT model were given inSection 1 in Supporting Information S1.

The initial conditions of the NDP model included the area, water level, nutrient concentration, population, and number of livestock and poultry of the four land use types in 141 polders of the plain agricultural watershed. The boundary conditions included time series meteorological and irrigation data. NDP model contained five water balance modules composed of 22 parameters and three N dynamic modules composed of 25 parameters. All the parameter values were obtained from previous studies in a typical polder located not far (about 38 km) from the study area (Huang et al., 2018). Since the hydrological and hydraulic conditions of these polders were consistent under the unified management of the Taihu Lake Basin Authority of the Ministry of Water Resources, the simulation results of NDP model were reliable. Compared with the case of watershed simulation using 257 models (Wellen et al., 2015), the simulation results of NDP model were acceptable, with the *NSE* between simulated and measured water level and TN concentration were 0.73 and 0.53, respectively; compared with the traditional method, the simulation accuracy by NDP model in the plains was improved by 63%. The detailed information of sensitivity parameter analysis processes as well as model calibration and validation processes of NDP model were given in Section 2 in Supporting Information S1.

4. Results

4.1. N Loss With Surface Runoff

Among different runoff components, in the hills, surface runoff was the main pathway of TN loss, accounting for 61%. Especially during the rainy season, the dissolved N cannot be fully absorbed by the plant and soil and be partly washed out with the surface runoff. In the dry season, the proportion of TN loss from deep soil water (i.e., interflow or groundwater) was higher (70%) than that from surface runoff (Figure 3a). In the plains, the TN loss from surface runoff was higher (78%) than that in the hills, which should also be concerned in the dry season. Because in the rainy season, the drainage gates were closed to keep the soil water saturation for rice planting. During this period, the water pressure difference between the polders and the surrounding rivers led to an increase in TN loss though seepage. In the dry season, after rice harvest, in order to continue wheat planting, the excess water and N of the paddy land was discharged through drainage. Moreover, due to the slow growth of crops in winter, N fertilization could not be completely absorbed, resulting in excess N being transported to the surrounding rivers through surface runoff (Figure 3b).

4.2. Driving Factors of N Loss With Surface Runoff

Runoff amount were the dominant factor controlling the TN loss. The TN loss in the hills was 3,583.42, 2,209.54 and 1,730.53 ton in the wet year, normal year and dry year, respectively; and the TN loss in the plains was 4,062.55, 2,650.08 and 1,935.57 ton in the wet year, normal year and dry year, respectively. The change trend of TN loss and runoff amount was consistent, showing the characteristics of wet year > normal year > dry year,

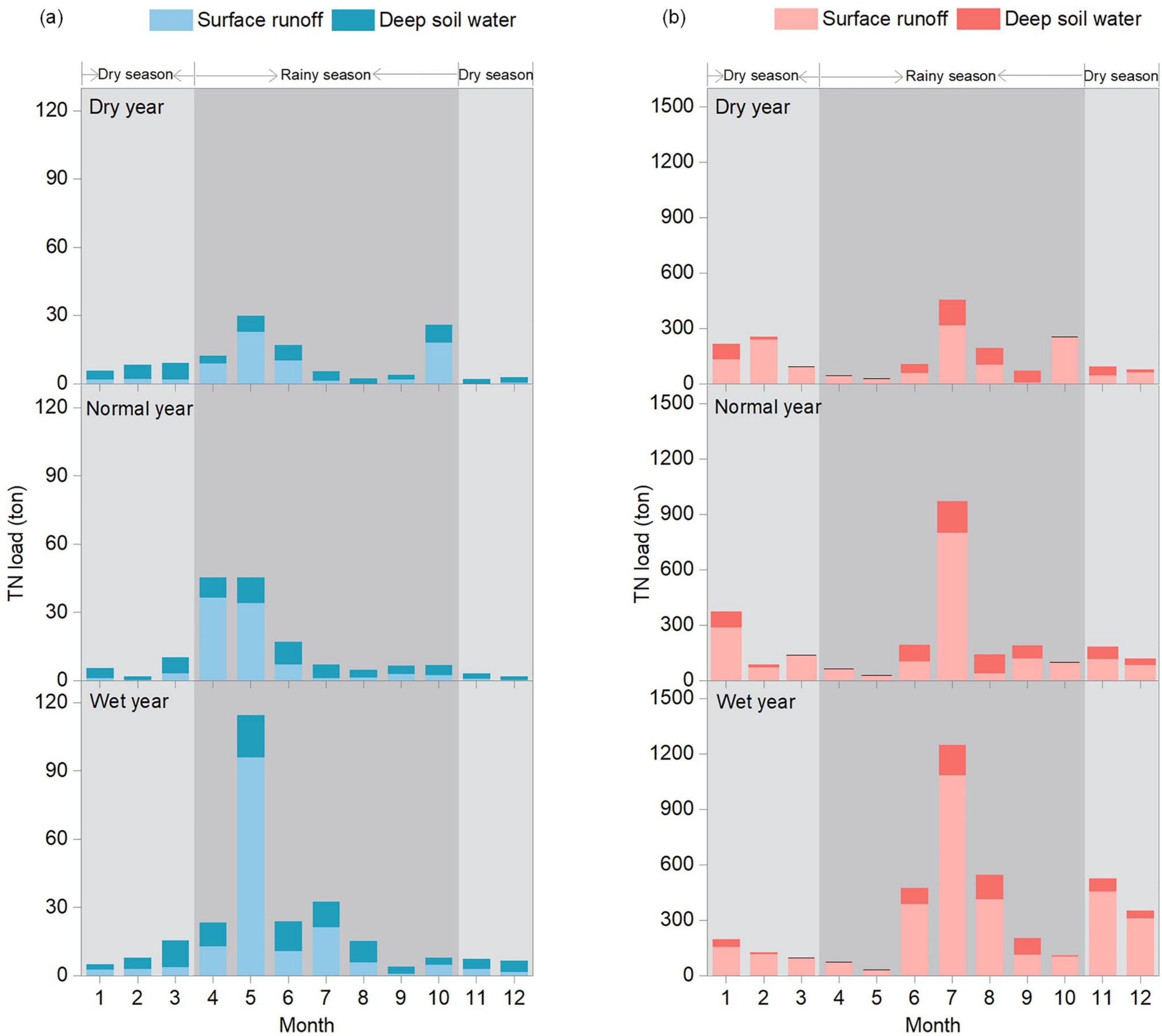

Figure 3. TN loss with surface runoff and deep soil water in the hill (a) and plain (b) agricultural watersheds.

properly because N was easy to be accumulated in dry years, and easily to be washed out in wet years (Kaushal et al., 2011).

However, the change trend of TN concentration and runoff amount was not consistent. According to the results of bivariate LISA, in the hills, TN concentration showed a weak negative correlation with runoff amount (Moran's I = −0.293), indicating that due to the dilution of runoff, TN concentration was decreasing. In the plains, there was no significant correlation between the TN concentration and runoff amount (Moran's I = 0.078), that was, the TN concentration did not decrease when the runoff amount was low, which may due to the effect of fertilization at low flow periods (Figures 4b and 4e). The above proved that when the runoff amount was reduced to a certain threshold, the TN concentration was no longer controlled by the runoff amount, but was affected by other factors such as land use and fertilization.

Due to the large amount of N fertilization applied on the agricultural land (i.e., paddy land, dry land and aquaculture pond), it may be the hot spots of TN runoff loss. As shown in Figure 4, the TN concentration increased with the proportion of agricultural land in the hills (Moran's I = 0.356), while the opposite trend was observed in

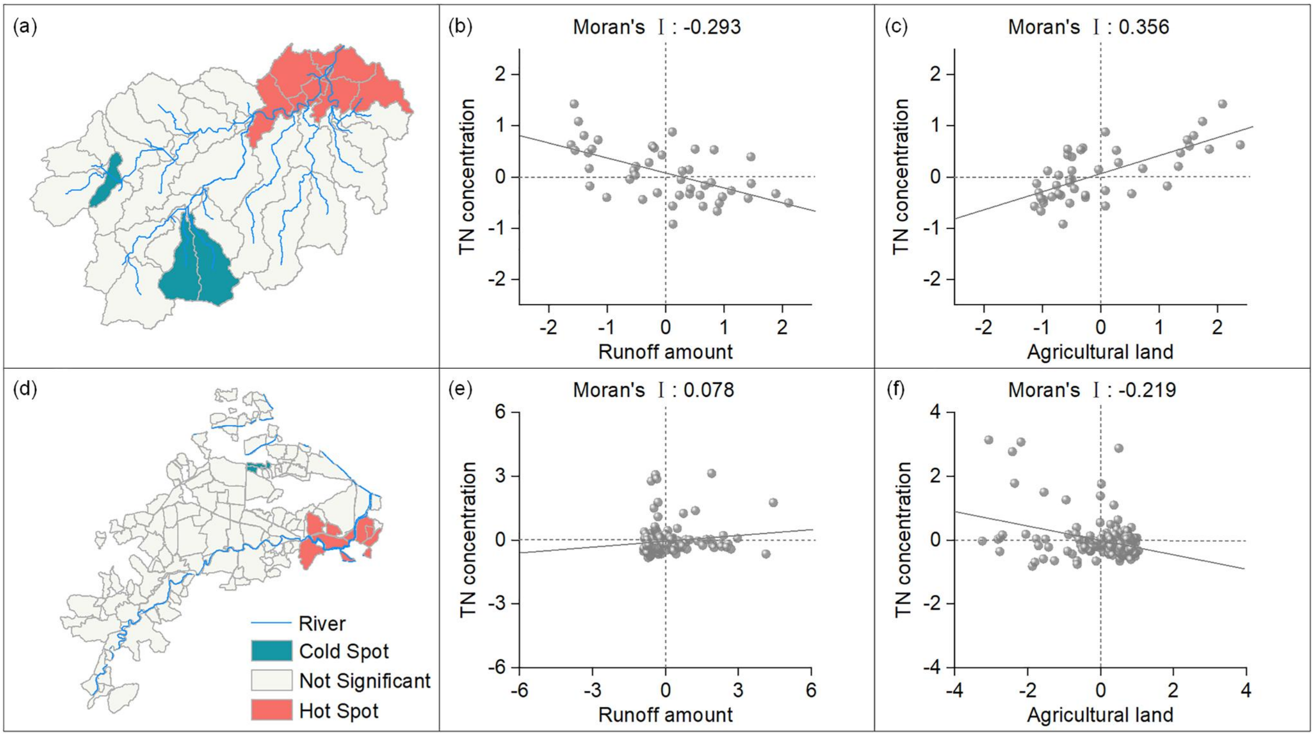

Figure 4. The Spatial autocorrelation analysis of TN loss concentration with surface runoff in the hill (a) and plain (d) agricultural watersheds; the bivariate LISA analysis between TN loss concentration with surface runoff and runoff amount in the hill (b) and plain (e) agricultural watersheds; the bivariate LISA analysis between TN loss concentration with surface runoff and the proportion of agricultural land in the hill (c) and plain (f) agricultural watersheds.

the plains (Moran's I = −0.219). Therefore, agricultural land may be the main N source landscape in the hills, not in the plains. Further analysis showed that the TN loss of paddy land and dry land increased with the runoff amount especially in the hills (Figure 5). Moreover, the TN sink of pond in the plains was much higher than that in the hills, which can compete with the amount of TN source in paddy land and dry land. Therefore, we speculated that the paddy land and dry land may be the main N source in the hills, rather than in the plains.

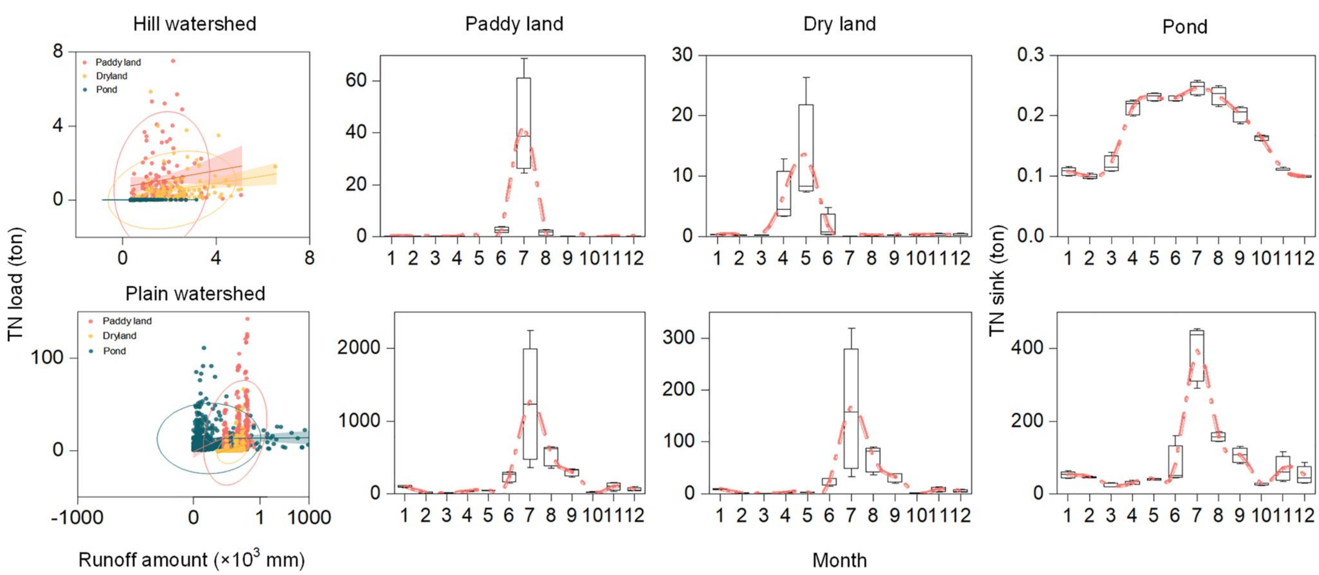

Figure 5. The linear correlation analysis between TN loss load with surface runoff and runoff amount and the time variation analysis of TN load/sink under different agricultural land types in the hill and plain agricultural watersheds.

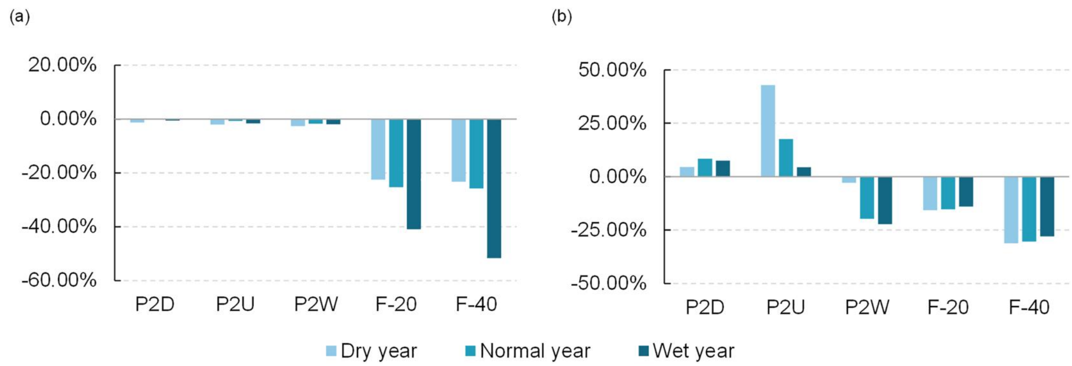

Figure 6. TN reduction rate under different land and fertilization management scenarios in the hill (a) and plain (b) agricultural watersheds; P2D means from paddy land to dry land; P2U means from paddy land to urban land; P2W means from paddy land to pond; F-20 means 20% reduction in N fertilizer; F-40 means 40% reduction in N fertilizer.

4.3. Contributions of Driving Factors

Multi-scenario simulation results showed that in the hills, the TN loss increased in case of other land use types were converted into paddy land, indicating that paddy land was the main N source landscape and therefore the effect of reducing N fertilization was obvious (Figure 6a). In the plains, the TN loss increased significantly after paddy land converted into urban land, indicating that urban land was the main N source landscape. Moreover, the role of N sink in rural ponds was equivalent to a 20%–40% reduction in N fertilization, with nearly 25% of the watershed TN reduction rate (Figure 6b).

Moreover, it found that the effects of land use transposition and N fertilizer application on N loss were different in wet and dry years. Compared with the normal year and dry year, the N increment effect was doubled in the wet year in the hills (Figure 6a). As the proportion of paddy land of sub-watersheds was increased, the annual TN loss increased faster during high flow periods (Figure 7a). Different from the hills, the N increment effect in the dry year should also be noticed in the plains (Figure 6b). As the proportion of urban land of polders increased, the annual TN loss increased faster during low flow periods (Figure 7b). According to the above results, it can be speculated that natural watersheds were susceptible to agricultural land use and fertilization, especially in high flow periods; complex watersheds (i.e., lowland watershed) had a higher proportion of urban land and more point sources, which can contribute more TN loss in low flow periods.

Further, the pRDA analysis was used to quantify the single and joint contribution of rainfall, land use and N fertilizer application to the total variance of TN loss (Figure 8). TN loss was more sensitive to rainfall than to land

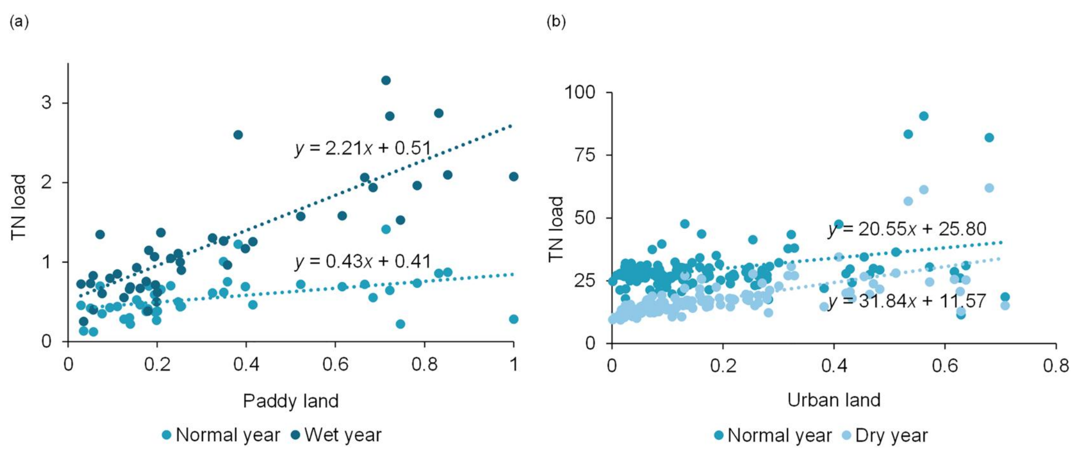

Figure 7. The linear correlation analysis between TN loss load with surface runoff and the proportion of paddy land of sub-watersheds in the hills (a); the linear correlation analysis between TN loss load with surface runoff and the proportion of urban land of polders in the plains (b).

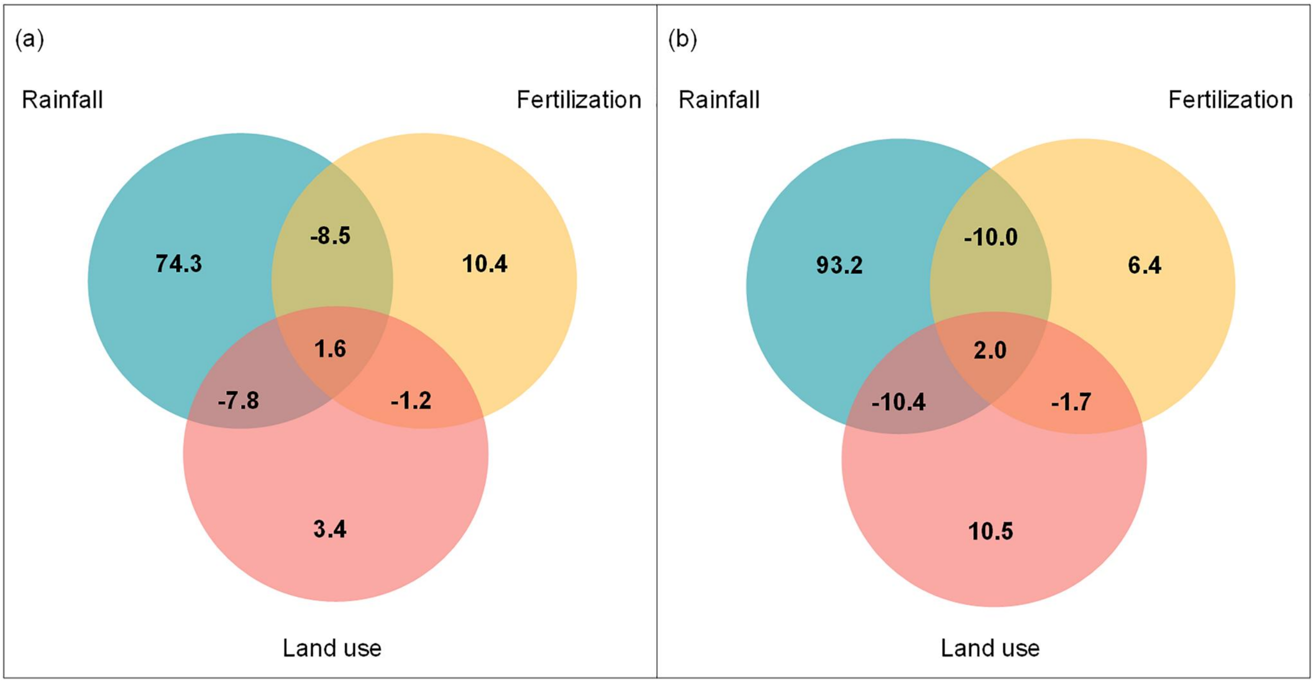

Figure 8. The pRDA results for relative variation fractions of the N loss with surface runoff explained by rainfall, land use and fertilization in the hill (a) and plain (b) agricultural watersheds.

use and N fertilizer application both in hills and plains. Moreover, the contribution of rainfall in the plains was greater than that in the hills; the contribution of land use in the plains was greater than that in the hills; the contribution of N fertilizer application in the hills was greater than that in the plains.

5. Discussion

5.1. The Response of N Loss to Rainfall, Land Use and Fertilization

Overall, the response of N loss to rainfall was more sensitive than that to land use and fertilization. Rainfall striped N from the soil surface, and runoff provided energy for N to transport downstream. The rainfall intensity may determine the amount of N loss entering the water body, and the rainfall frequency may affect the degree of dilution of the N concentration. Therefore, the N load in the rainy season was higher than that in the dry season, while the N concentration in the dry season was higher than that in the rainy season.

Slope affected the intensity and rate of N loss movement. The generation time of N loss with surface runoff was shorter in the hills than that in the plains after rainfall. Land use directly affected the interaction between runoff and soil, and its changes ultimately caused N loss to a large extent (Tan et al., 2023). The proportion of urban land in the plains increased significantly, and the impervious surface caused serious N loss after rainfall. Moreover, the contribution of land use to N loss in the plains was greater than that in the hills, may because the impact of land use change on surface runoff in the hills was relatively small. The main sources of surface runoff in the hills include rainfall, groundwater and a small proportion of soil water. The groundwater was relatively stable and was mainly affected by climate change (i.e., rainfall) rather than human activities (i.e., land use) in the long-term process (Mani et al., 2016). The soil water saturation of the hills in our study area was high, so the N loss with surface runoff occurred and stopped continuously with the continuation and regression of rainfall, which may to some extent cover up the effects of land use (Chelsea Nagy et al., 2012; Li, Shi, et al., 2020).

The contribution of N fertilization to N loss in the hills was greater than that in the plains. Because paddy land was the most important N source landscape in the hills, fertilization had a greater impact on N loss.

5.2. Implications for N Control

Due to the different N fixation and release capacity in each land use type, there are huge differences in N loss to the surrounding water bodies by different combinations of land use types (Kändler et al., 2017). Hou et al. (2019) found that a decrease of nearly 10% in the N load in surface runoff could be achieved by redistributing land use within the experimental watershed without any changes in any types of land management practices. Therefore, N pollution can be reduced by adjusting land use patterns.

In our study, sub-watersheds with high proportion of paddy land in the hills and polders with high proportion of urban land in the plains are considered to be the key source areas of N loss. Therefore, in the hills, returning agricultural land especially paddy land to forest is necessary. Rapid land cover change as a result of agricultural encroachment onto natural land often lead to long-lasting impairment of soil and water conservation and other crucial ecosystem services. Cebecauer and Hofierka (2008) also found that deforestation and conversion of pastures to agricultural land in the hills was the main factors exacerbating the risk of soil erosion and N loss. In the plains, urban development has accelerated the N loss, so it is necessary to control urban expansion. Roberts et al. (2009) also found that urbanization and increased imperviousness in the future could increase nutrient load to the Chesapeake Bay by 56%, leading to eutrophication and harmful algal blooms. The use of pond systems to reduce watershed N pollution has shown increasing advantages. Most of ponds in the hills are natural ponds with high transparency and few aquatic plants, and their N removal is mainly based on sedimentation. Most of the ponds in the plains are aquaculture ponds, receiving and storing runoff and nutrients from paddy land and urban land (Wang et al., 2019), and their N removal is mainly based on the absorption of aquatic plants and other biochemical reactions. It is obvious that the N sink capacity of ponds in the plains is much higher than that in the hills, with TN reduction of 1,169.87 and 19.23 $kg \cdot ha^{-1}\ yr^{-1}$, respectively. Internationally, pond systems have been considered as one of the best management practices (BMPs) for watershed N control (Li, Liu, et al., 2020).

The N loss entering the water body is promoted by N-fertilization time and rainfall. Rainfall is uncontrollable, while reasonably reducing N-fertilization dose and post-rain fertilization could be implemented based on modern agricultural technology (Ding et al., 2022; Guo et al., 2023; Sun et al., 2012; Zhang, Hou, et al., 2020). Unfortunately, due to higher evapotranspiration and extreme rainfall, higher drought or flood risks would be observed in different regions, which may amplify the effects of land use and fertilization (Aksoy et al., 2021; Amiri & Gocić, 2021; Law et al., 2022). Therefore, more N loss load would be observed in most agricultural land-dominated watersheds. Based on the future climate change, more stringent land management and fertilization control would be needed.

6. Conclusion

This study is comparing the response of N loss with surface runoff to rainfall, land use and fertilization between hill and plain agricultural watersheds using two process-based models. The results showed that among different runoff components, surface runoff was the main pathway of TN loss especially in the plains. The variation trend of TN loss load was consistent with that of runoff amount, showing the characteristics of wet year > normal year > dry year. However, the TN loss concentration was also high during low flow periods, may due to the effects of land use and fertilization. In the hills, paddy land was the main N source landscape compared to the urban land with low population density, whose N increment effect was more obvious in high flow periods. In the plains, urban land with high population density was the main N source landscape compared to the agriculture land with relative natural structure, whose N increment effect was more obvious in low flow periods. The contribution of fertilization to N loss in the hills was greater than that in the plains, because the high proportion of paddy land with abundant N fertilizer demand. Moreover, the contribution of rainfall to N loss in the plains was greater than that in the hills, because the high proportion of urban land with more impervious surface led to serious N runoff loss.

Data Availability Statement

Land use data in 2015 in Xitiaoxi River Basin can be obtained from https://doi.org/10.5281/zenodo.3986872. DEM in Xitiaoxi River Basin can be obtained from https://data.tpdc.ac.cn/zh-hans/data/acb49ce8-2bfe-4ab4-97ff-e6e727110703. Soil data in Xitiaoxi River Basin can be obtained from https://www.tpdc.ac.cn/zh-hans/data/acb49ce8-2bfe-4ab4-97ff-e6e727110703. Observed daily meteorological data from 2009 to 2017 in Xitiaoxi River Basin were collected from https://doi.ora/10.11888/AtmosPhys,tpe.00000049.file. Model configuration, calibration and validation, input and output files in the present study are available online at https://doi.org/10.6084/m9.figshare.25138847.v1. The NDP model source code is available to download from https://doi.org/10.6084/m9.figshare.25138838.v1.

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