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Effects of Land-Use and Land-Cover Change on Nitrogen Transport in Northern Taihu Basin, China during 1990–2017

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Abstract: Different land uses have varying degrees of impact on nitrogen transport in the catchments. In recent decades, rapid urbanization has dramatically changed the Earth's land surface, which may cause excessive nitrogen losses and a negative influence on the environment. In the long-term scale, it is important to explore how the nitrogen transport responds to land use change and its effects on aquatic habitats. In this study, the water and sediment samples were collected from northern Taihu Basin, and nine periods of land use data were obtained using the techniques of supervised classification. Results revealed that the proportion of farmland area decreased from 28.33% to 7.09%, while that of constructed land area increased from 23.85% to 61.72% during 1990–2017. Most of the constructed land originated from farmland, which makes it the dominant land use type due to rapid urbanization. In spatial distribution, high total nitrogen (TN) losses regions remain distributed over constructed land and farmland, which may aggravate the trend of local water quality deterioration. Of these regions, constructed land was the dominant contributor (46.29%–63.62%) of TN losses from surface runoff. In temporal variation, the TN losses of runoff decreased by 47% from 175 t N·a⁻¹ in 1990. However, they increased by 2.91% from 75.28 t N·a⁻¹ after 2013 with rapid population growth and high fertilizer application (>570 kg·ha⁻¹). The nitrogen load in sediments also has a significant response ($t = 2.43$, $p = 0.02$) to the effects of land use change on the overlying water, indicating that the role of nitrogen in the sediment as a source and/or sink to the waterbody may change frequently. Given the increasing accumulation of nitrogen loads in highly urbanized regions, water quality would cause more aggravation in the long-term without reasonable land management measures.

Keywords: Taihu Basin; land use change; nitrogen losses; nitrogen transport; water eutrophication

1. Introduction

Land use and land cover are products of both anthropogenic activities and natural environment [1]. In the 1990s, the International Geosphere-Biosphere Programme and the International Human Dimensions Programme on Global Environmental Change initiated their core project on the land use and land cover change (LUCC) [2,3]. Through the comparative study of regional cases, the driving factors of both the natural and social economy that influenced the land users or managers to change the land use type and the management mode have been explored [4–8]. These results indicate that in the future, the anthropogenic land use activities may play a major role in global environmental

changes [9–11]. Inappropriate land use management is one of the main factors resulting from the increase of nitrogen losses and pollution in the waterbody [12–14]. Farmland and constructed land have been considered as the major sources of aqueous nitrogen, while the forest-grassland could be the sink for the pollutants [15–17]. The accumulation of nutrients between the water and sediment is a long-term process that may be affected by multiple factors [1,13,15]. Thus, the research of land use types, transition and the accuracy of classification impacting water quality in the short-term may have greater uncertainty [18–20].

Nevertheless, most of the existing studies have focused on exploring the effects of land use types on water eutrophication in the short-term scale [6,18,19]. In the long-term scale, we lack comprehensive research on the influence of land use changes in nitrogen transport between the waterbody and sediment combining with anthropogenic activities and the natural environment. These issues were not getting enough attentions in the research of LUCC impacting nitrogen transport and water eutrophication. In addition, the anthropogenic activities varied in form and intensity, influencing the nitrogen flow and habitat in the catchments differently [19,20]. Excessive ammonia and nitrogen oxide emissions from economic activities not only result in large amounts of nutrients being wasted, but also cause serious environmental problems [14,21–23]. However, a great quantity of fertilizer is needed for agricultural production. Due to the low nitrogen fertilizer utilization but high nitrogen losses by runoff and leaching [21], lots of nutrients and pollutants were discharged from domestic sewage, livestock and poultry farming via surface runoff [22,23].

Nowadays, the nitrogen losses of runoff have not been controlled effectively in farmland, constructed land and the processes of their transition [24–26]. With rapid urbanization, the urban surface runoff has increased due to the expansion of the impervious surface area. Moreover, with the limited sewage treatment capacity and the uncompleted system of rain and sewage separation, more nutrients and pollutants were discharged into the water via runoff [22,24,26]. This leads to the following question: where is the nitrogen from and how do we quantify nitrogen losses response to the land use change in long-term scale? As we know, sediment and water are an interactive whole. If the research of land use change impacts the water quality separated from the sediment, the effects may be overestimated or underestimated [25,26]. Therefore, when exploring the effects of land use change on nitrogen transport between water and sediment in the catchment, it is important to clarify the long-term variation and identify the multiple factors. As the anthropogenic land use activities have altered the lakeside zone type and ecological structure more or less, the identification of source and/or sink of nitrogen in different regions is the key task for the eutrophication control [9,10,14].

Since the mid-1980s, the status of water eutrophication in Taihu Lake in China has changed from mild to severe [27–30]. Following the Wuxi water crisis in 2007, a highly publicized drinking water crisis occurred in Wuxi city, Jiangsu Province, the environmental governance has focused more attention on environmental water treatment [31–33]. Although the water quality has been improved overall, water eutrophication and algal blooms still happen frequently. With the intensity of anthropogenic activities especially the land use changes and rapid urbanization [34,35], the nitrogen balance and the habitat in the catchments were damaged year-by-year. Considering the accumulative effects of pollution, a large amount of nutrients (e.g., nitrogen and phosphorus) remained in the water environment which may provide rich nutrients for cyanobacteria growth. Therefore, it is urgent to quantify the contribution of land use changes to the nitrogen load in the catchment for making long-term efficient adjustments and control mechanisms.

The present study was conducted to: (1) understand the spatiotemporal distribution of land use change; (2) calculate the nitrogen losses of runoff from different land use types and its transition; (3) analyze how the land use change impacts nitrogen transport between the water and sediment, and provide some suggestions for environmental water management in the future.

2. Materials and Methods

2.1. Study Area

The study area is Taihu Basin ($29^{\circ}55'–32^{\circ}19' N$, $118^{\circ}50'–121^{\circ}55' E$), located in the lower reaches of the Yangtze River Delta, China (Figure 1). It has an area of $36,900 \text{ km}^2$ (2338 km^2 of the lake area) and has a typical subtropical monsoon climate with an annual mean temperature of 16°C and an average 1181 mm of annual precipitation [36]. Due to industrialization and urbanization resulted from dense population and active economies, this area has become a comprehensive base in China [37]. As a typically disc-shaped and shallow lake, Taihu Lake has an average water depth of 1.9 m . Wheat and rice are the main crops in the study area. Wuxi city is a highly urbanized and heavily polluted region, in the northern Taihu Basin with low terrain and dense river networks. A high nitrogen fertilizer application level ($>570 \text{ kg N ha}^{-1}$) and low utilization rate (30%–35%) have been the characteristics of agricultural production in this area. In addition, the largescale livestock production and aquaculture enterprises have decreased in recent years. However, many scatter-feeds of aquaculture enterprises are near the lake shore or in the lake. Due to the limited sewage treatment capacity and rapid urbanization in this region, more domestic sewage is discharged directly into the rivers and lake [28,30]. Currently, the water quality in the lake is seriously aggravated.

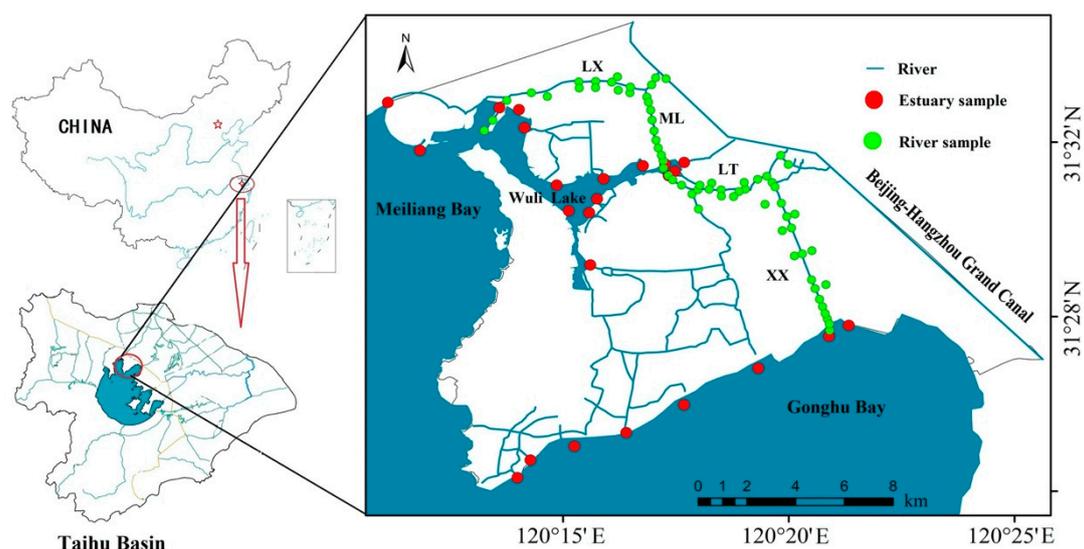


Figure 1. Location of Sampling Site in the Northern Taihu Basin. Estuary Sampling Sites Including Water and Sediments Samples; LX, ML, LT, XX Represent Liangxi River, Mali River, Liangtang River and Xiaoxi River, Respectively.

In the study area, dams or gates have been constructed on more than 85% of the estuarine rivers, destroying the natural hydrological connectivity between the rivers and the lake. The flow direction of these rivers is almost controlled by the gates or dams (Figure S1). Thus, three types of rivers (inflow river, outflow river and outflow-inflow river) were formed due to the regulation of these gates. The inflow river was defined by the flow direction from the river to the lake. For the same reason, the flow direction from the lake to the river was the outflow river. The outflow-inflow river was defined as the flow direction usually changed by the regulation of the gates. In recent years, a prosperous economy and rapid urbanization have caused more nitrogen to be discharged into the lake as a result of land use change and the associated anthropogenic activities. The construction of dams and gates may accelerate the nitrogen accumulated in the waterbody. All of these reasons have had an increasing impact on the lake's water quality.

2.2. Sample Collection and Laboratory Experiments

The surface water samples and sediment samples were collected from the different land use types. All surface water samples were collected in triplicate at a depth of 1 m using a layered collector with three compartments. The three discrete water samples were mixed to form a composite sample and placed in a polyethylene bottle. For each sediment sample, the sediment in the 0–10 cm-depth of the benthic surface was collected using a Peterson grab bucket. After the rubble and litter had been removed, the samples were transferred to a sealable plastic bag.

All the samples were transported to the laboratory immediately after collection. The water samples were stored at 4 °C until physicochemical analyses had been completed. One half of each sediment sample was stored at –20 °C. The other half was freeze dried (at 20 Pa and –50 °C). After freeze drying, the sediment sample was ground for further analysis.

Samples were analyzed for several physicochemical parameters according to the standard methods [38]: total nitrogen (TN), total phosphorus (TP), ammonia nitrogen (NH₃-N), nitrate nitrogen (NO₃⁻-N), total inorganic nitrogen (TIN = NH₃-N + NO₃⁻-N), total organic nitrogen (TON = TN-TIN) and total organic carbon (TOC). Specifically, the concentrations of TN, TP, NH₃-N and NO₃⁻-N were determined by ultraviolet spectrophotometry (UV-3600, Shimadzu, Japan). The TOC concentration was measured using a TOC analyzer (SSM-500A, Shimadzu, Japan).

2.3. Land Use Detection and Analysis

Digital images (30 m grid resolution) for nine periods from 1990 to 2017 were analyzed to determine the land use in the study area. Nine remote sensing measurements (1990, 2000, 2005, 2009, 2010, 2011, 2013, 2015, 2017) of Landsat 4-5 TM and Landsat 8 OLI products were obtained from the Geospatial Data Cloud, and the Yangtze River Delta Science Data Center (National Earth System Science Data Center, National Science and Technology Infrastructure of China) [39,40]. To better classify the land use types, all the images from June to September, an exuberant vegetation growth state, were selected once a year [1,16,19]. In order to improve the accuracy of the interpretation, the cloud coverage ratio of each image was lower than 10%. Then, the methods of radiometric calibration and boundary extraction were used to preprocess the images [16,19].

All the land use data were interpreted based on the supervised classification by ENVI 5.3 software (ESRI, USA). Combining with the vector data and several field investigations, there has a total of ten land use types. Each land use type has 10 regions of interest (ROI) as training samples for further analyze. 90% of the training sample data for supervised classification were selected randomly by the maximum likelihood classifier (MLC) and 10% of training samples were used for accuracy evaluation (Table S4). Based on the calculation of the confusion matrix, the accuracy of the image classification met the research requirements (88% < kappa < 95%). The areas of patch which ranged from 900–1800 m² were dissolved into the surrounding land use types. According to the Chinese National Standard of Current Land Use Classification System [18], all land use types were reclassified into five categories (farmland, water area, forest, grassland and constructed land). The spatiotemporal distribution of land use and the reclassification were accomplished using the spatial analyst tool of ArcGIS 10.0 (ESRI, USA).

2.4. Calculation of the TN Losses in Runoff

The contribution of each pollution source to the water nitrogen load was assessed from land use types and the associated anthropogenic activities based on the point and nonpoint-source pollution. Point-source pollution included industrial sewage and urban sewage. Nonpoint-source pollution is the pollutants in runoff from fertilizer application, livestock, aquaculture, urban surface and other land use types [41]. The TN (except for NO_y and NH₃) losses in runoff were calculated using Equation (1) [42,43]:

$$N_T = \sum_{i=1}^n N_i \times F \quad (1)$$

where, N_T is the amount of TN losses in the runoff (t), N_i are the pollutant generated coefficients of eight sources (i including industrial sewage, urban sewage, rural sewage, farmland runoff, constructed land runoff, forest-grassland runoff, livestock and aquaculture) [44–46], and F_i are the coefficients of pollutants discharged into the water for sources i [41]. All the coefficients are shown in Table 1 and Table S1. Literatures indicate that the F_i values have more uncertainty than the N_i values when calculating TN losses, because the N_i values have significant difference in spatial distribution but no regular variation in temporal [44–46]. In addition, due to the rapid economic growth and improvement of sewage systems (e.g., the rate of sewage treatment has increased from 30% to 88%), the F_i values have changed markedly in the temporal distribution [41]. Therefore, the average value of N_i was selected in this paper would reduce the uncertainly. Meanwhile, considering the improvement of sewage treatment capacity, different values of F_i were selected when calculating TN losses in different periods.

Table 1. Coefficients of Various Pollutants Generated and Discharged into the Rivers.

	Industrial Effluents	Urban Sewage	Rural-Domestic Sewage	Farmland Runoff	Aquaculture
N_i	–	2.92 kg N a ⁻¹ cap ⁻¹	2.19 kg N a ⁻¹ cap ⁻¹	–	1800 kg N·(ha a) ⁻¹
F_i (%)	50–65	12–70	10	5–6.36	100

2.5. Statistical Analysis

All the experimental data and Yearbook data (1990–2018) were analyzed using the Statistical Product and Service Solutions software (IBM, USA) and Microsoft Excel 2016 (Microsoft, USA). Using ENVI 5.3 and ArcGIS 10.0 (ESRI, USA) software, the land use types and physical-chemical parameters were interpreted for further study. In addition, the attribute data of land use types were processed by reclassification. The redundancy analysis (RDA) was analyzed based on R v.3.5.1 [47]. Pearson's correlation analysis and paired-sample t -tests were used to explore the effects of dam construction, land use change, estuary shape and monsoons on the water quality. Two significance levels of $p < 0.05$ and $p < 0.01$ were used to define statistical significance.

3. Results and Discussion

3.1. Characteristics of Land Use Changes

Each land use type exhibited obvious temporal distribution characteristics and periodic differences during 1990–2017 (Figure 2). The areas of farmland decreased constantly while those of constructed land increased rapidly. After 2005, constructed land became the dominant land use type in the region and its proportion had been increased from 23.85% in 1990 to 61.72% in 2017. The area of forest-grassland and water had a decreased trend on the whole. In recent years in China, the emphasis by the government has been placed on environmental protection, and so the area of forest-grassland has been increasing based on the expansion of city afforestation [33]. Between 1990 and 2010, there was a strong transition among the land use types. Then, the transition became relatively stable after 2010. Before 2010, with the reform of administrative divisions in the study area [41], the government strengthened the construction of the Binhu district. Therefore, with the high-frequency exchange among the land use types in the different functional areas of the city, the area of constructed land rapidly increased while the area of farmland gradually decreased. However, since 2010, the growth rate of constructed area has slowed as the structure of each functional area of the city gradually improved when urbanization reached a high level. Thus, the exchange frequency among land use types has decreased gradually.

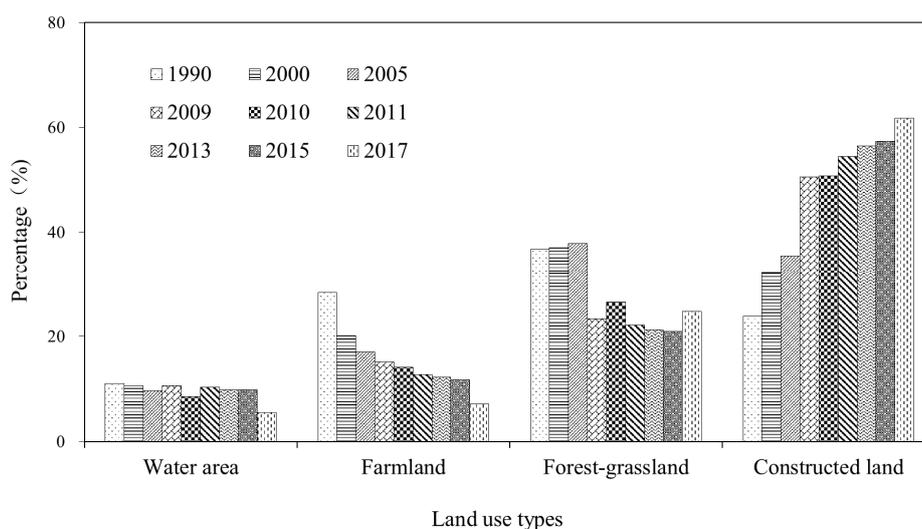


Figure 2. Temporal Variation of Land Use Types from 1990 to 2017.

Meanwhile, the spatial distribution of land use types was profoundly affected by its transition (Figure S2). Almost all the forest-grassland was concentrated in the western mountainous part of the study area and remained stable on the whole. The constructed land was concentrated in the central Binhu district and the northern old city district which has an expanding trend. Except for the areas of river network that were dispersed through the whole study area, the other water areas were concentrated in Wuli lake (Figure 1 and Figure S2). The transition from farmland to constructed land was the dominant form of land use conversion, concentrated in the central region. This may be caused by both expansions of the old city and construction of the new district. Furthermore, with changes in landscape planning and the land policy, the transition of water area to the constructed land, farmland and forest land mainly occurred in the northwest region. Similarly, the transition from the farmland to forest-grassland was found in the east and south of Gonghu Bay wetland park (Figure S3). Therefore, we deduced that urbanization and policy adjustment were the main forces driving spatiotemporal transition among the land use types [39].

Based on the spatiotemporal variation of land use change in this area, the areas of constructed land were preliminary estimated to be increased by 12.93% in 2018 compared to 2017, while the area of farmland was decreased by 26.64%. Spatially, the high-frequency transition of land use types was still concentrated in the central zone, we need to consider the dramatic changes of land use types in this study area, how the nitrogen losses in runoff and transport between the waterbody and sediment responds.

3.2. Spatiotemporal Variation of the TN Losses

To better unlock the spatiotemporal distribution of land use and its transition impacting on nitrogen transport and losses at the drainage basin scale, the TN losses in runoff from different land use types and their transitions were interpolated with the spatial analysis of ArcGIS 10.0 software (Figures 3 and 4). In the period of 1990–2017, constructed land was the dominant contributor (41%–62%) to the TN losses in runoff (Figure 3). In comparison, the dry land, the paddy land and the forest-grassland contributed 8%–17%, 13%–19%, and 11%–22%, respectively. The TN losses in runoff from different land use types decreased during 1990–2017, although an increasing trend has been exhibited since 2013.

Due to the improvement of urban sewage treatment in the study area, the amount of TN losses from runoff into the water bodies has been reduced on the whole [35]. Consequently, the TN losses from the constructed land exhibited a downward trend over time, which decreased by 63% in 2017 compared with 1990. Nevertheless, since 2011 the TN losses in runoff from the land use types has increased with the rapid development of urbanization and population growth [19,48], as well as the

transition from farmland to the constructed land. Therefore, the TN losses from the dry land and paddy land exhibited a downward trend overall. The TN losses from the forest-grassland show a fluctuated and downward trend mainly due to the deforestation and land reclamation in the early stages. However, in recent years, environmental protection and construction of the ecological garden city have obviously increased the forest-grassland areas [33]. As plants have the good effective function of absorbing and intercepting nutrients, the risk of water pollution has been reduced gradually, e.g., in Gonghu Bay wetland park (Figure S3), the river water quality (located in wetland park) is better than that in the lake (almost no plants).

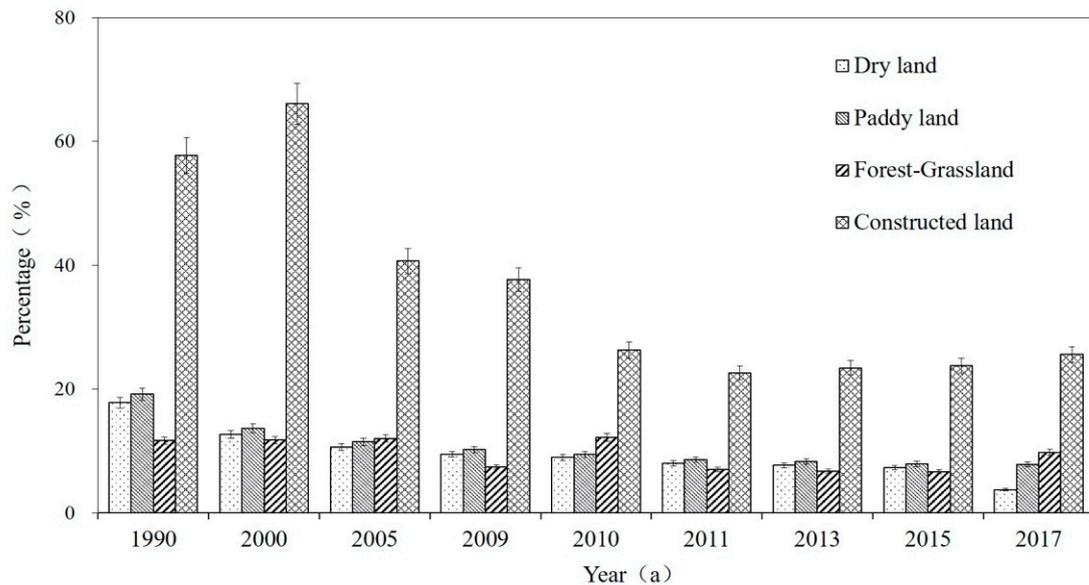


Figure 3. Variation of the total nitrogen (TN) Losses in Runoff from Different Land Use Types during 1990–2017.

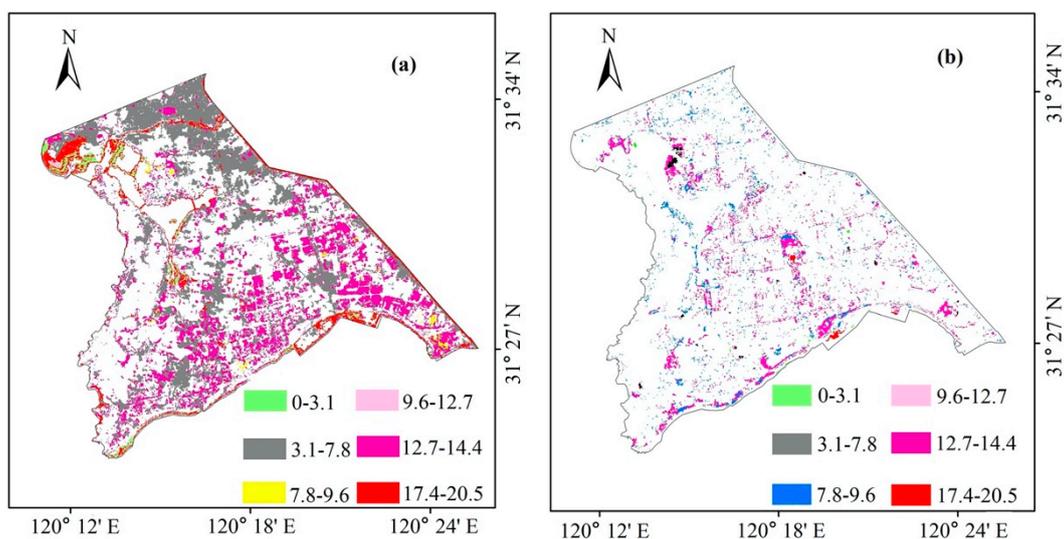


Figure 4. Spatial Variation of the TN Losses Amount in Runoff from Land Use Change during 1990–2017. (a) the Areas with Increased TN Losses; (b) the Areas with Decreased TN Losses. Unit: $\text{kg N}\cdot\text{ha}^{-1}$.

As shown in Figure 4, the TN losses of the increased and decreased regions have a significant spatial distribution difference. For example, the regions with increased TN losses were mainly distributed in the southern and northern areas with a form of radial pattern (Figure 4a and Figure

S2). This indicated that the transition of southern forest-grassland and northern farmland into the constructed land is accompanied by rapid urbanization. The distribution of decreased TN loss areas was relatively dispersed (Figure 4b). Except for in the Lihu wetland park (Figure S3) where the areas of decreased TN losses were concentrated uniformly, the form of the distribution in other areas was discrete points or ribbons. The TN losses in runoff due to the land use changes increased by 42.86% (87.23 t) in 2017 compared to those in 1990. With the rapid increase of constructed land and impervious surfaces, the TN losses may have an upward trend in the future with no effective management.

As the transition of constructed land into forest-grassland was distributed typically at the roadside, the areas of decreased TN losses in runoff showed a zonal distribution [15,17]. However, the characteristics of the distribution in the southern region were mainly influenced by government policy. The local government responded to the national policy of returning the farming areas to forestry and actively constructed a wetland park around Taihu Lake. Consequently, the forest-grassland areas around the lake show a ribbon distribution. Furthermore, forest and grassland effectively absorb and intercept nutrients. Thus, with the area of forest-grassland increased, the TN losses in runoff also decreased, which greatly improved the river water quality in the area the wetland park. Based on the regulation of the land use changes from 1990 to 2017, with no effective control measures, the amount of TN losses in runoff in 2018 was predicted to increase by approximately 26.74% compared to that of 2017. Considering the nitrogen loads have an enrichment effect in the sediment, the contribution of TN from land use types and changes to water cannot be ignored in a long-term. Based on the results above, we calculated where the nitrogen was discharged from and how the land use change impacted nitrogen transport in spatiotemporal variation.

3.3. Response of Physicochemical Parameters to Land Use Change

To better reveal the TN loads and its distribution in the water and sediment response to the different land uses types, the water and surface sediment samples were collected and analyzed for several physicochemical parameters (Figure 5). The average concentrations of TN, TP, NH₃-N and TOC in water were 2.58 ± 1.16 , 0.09 ± 0.10 , 1.14 ± 0.80 and 11.30 ± 2.34 mg·L⁻¹, respectively. Meanwhile, the average values of TN, TP, and TOC in the sediment were 1.81 ± 0.28 , 0.19 ± 0.02 and 4.99 ± 0.93 g·kg⁻¹, respectively (Table 2 and Table S2). The TN concentrations, of which the NH₃-N contributed for 40.18%, exhibited obvious characteristics of spatial distribution. In Wuli lake, the water quality near the lake was better than that near the river while the opposite trend was found in Gonghu Bay. In addition, the water quality in the western Wuli lake (Figure S2) was better than that in the eastern region that is close to the downtown district [48], while the western Wuli lake is located near the Yuantouzhu wetland park and waterworks (Figure S3). The ratios of N: P and C: N ranged between 11–25 and 2.7–8.4, respectively. On one hand, this indicates a habitat for algae growth [49,50]. On the other hand, high ratios reflect the excessive external pollutant inputs, which may be caused by the high transition of the farmland and forest-grassland to the constructed land [49–51]. Thus, the anthropogenic land use activities varied in form and intensity, influencing the hydrodynamic force and nitrogen flow in the catchment differently.

Table 2. Nutrient Concentrations in the Waterbody.

	All Study Area	Meiliang Bay	Gonghu Bay
TN	^a 2.80 ± 1.97/2.36 ± 1.94 ^b	^a 3.03 ± 4.28/ ^b 2.28 ± 2.09 ^b	^a 1.16 ± 0.55/2.51 ± 1.75 ^b
TP	^a 0.08 ± 0.12/0.09 ± 0.06 ^b	^a 0.11 ± 0.14/ ^b 0.07 ± 0.05 ^b	^a 0.04 ± 0.04/0.11 ± 0.05 ^b
NH ₃ -N	^a 1.17 ± 1.90/1.10 ± 0.87 ^b	^a 1.48 ± 2.31/ ^b 1.15 ± 1.05 ^b	^a 0.58 ± 0.36/0.74 ± 0.26 ^b
COD _{Mn}	^a 9.33 ± 2.59/13.27 ± 8.04 ^b	^a 9.39 ± 3.05/ ^b 9.86 ± 4.67 ^b	^a 9.22 ± 2.96/19.23 ± 9.47 ^b

Note: a and b are the concentrations of river samples and lake samples.

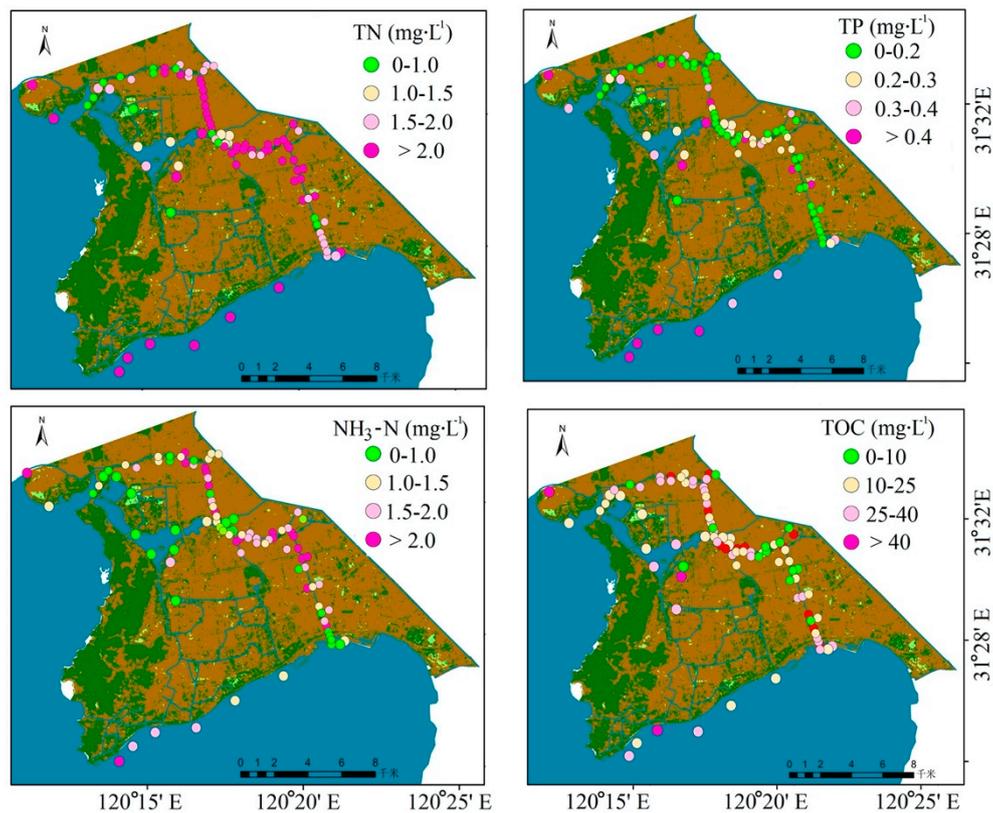


Figure 5. Spatial Distribution of Physicochemical Parameter.

Nitrogen transport and distribution in rivers may also be affected by the hydrodynamic force. In this area, almost all the rivers were controlled by the dams or gates. Thus, four typical inflow-outflow rivers (Liangxi River, Mali River, Liangtang River and Xiaoxi River) were collected to explore the effects of TN losses from different land use types on the nitrogen distribution based on different river types. Liangxi River (opened dam) and Xiaoxi River are the typical outflow rivers, where the water quality deteriorated gradually from upstream to downstream. In addition, the nitrogen concentrations in the tributary streams were higher than those in the trunk stream. These phenomena indicated that external pollution sources were the dominant contributor for water environmental degradation [30]. Clean water from the lake regions may reduce the pollution risk of river water when the gate is opened. Mali River and Liangtang River are the typical inflow rivers with a weak hydrodynamic force due to the dams being closed all year round. As a result, the self-purification capacity of these rivers has declined gradually. Moreover, the external nitrogen still continuously discharged into the rivers with runoff that resulted in a large amount of nitrogen accumulating in the river water environment [35]. Thus, the water quality of inflow rivers was obviously worse than that of out-flow rivers.

As we know, sediment and water have the characteristic of mutual influence and interaction. In order to reduce study errors, both of them should be considered when exploring the effects of land use changes on water quality [25,26,52]. Sediment has a katabatic or aggravated effect on water pollution when it receives the nitrogen losses from different land use types [52–55]. The role of sediment as a sink and/or source for the nitrogen in the waterbody is usually changed by the multiple factors [13,15,30,52]. In this study, the spatial distribution of nutrient loads in the sediment has a similar trend to that in the water (Table S2). For example, in Gonghu Bay, the nitrogen contents in sediments of the lake were higher than in rivers. Due to the construction of the wetland park, the TN load was reduced in the waterbody and sediment [41]. Under a stable hydrodynamic condition, the nitrogen discharged from runoff would be gradually self-purified or deposited into the sediment [56–58]. Under this condition, the sediment acted as the nitrogen sink to the waterbody. Thus, the water quality of the river was

better than that of the lake ($t = 2.43, p = 0.02$). For the long period of accumulation without further management measures, the nutrients load in the sediment and waterbody may increase gradually due to the input of exogenous pollutants.

In the lake region, the hydrodynamic conditions were usually changed by the wind-induced waves and anthropogenic activities [41,52,55]. Large amounts of nitrogen released from the sediment resulted in water quality deterioration. The results of laboratory experiments showed that the level of disturbance has a positive effect on the water quality (Table S3). Under the stirring speed of $120 \text{ r}\cdot\text{min}^{-1}$, the TN concentrations were increased to approximately 4 times those in static release. Thus, in the lake region, the sediment was regarded as the nitrogen source for the waterbody [52,55]. Consequently, we should identify the role of the sediment as nitrogen source and/or sink for the waterbody when exploring the effects of land use change on the water quality. Otherwise, the effects may be unclarified.

3.4. Driving Factors of Nitrogen Balance

Results of RAD (Figure 6) show that the TN concentration was significantly affected by the farmland and constructed land ($F = 4.6, p = 0.006$). In view of this, the constructed land and farmland as a nitrogen source aggravated the nitrogen load in the water environment and degraded the water quality. Contrarily, the forest-grassland may absorb nitrogen and purify the waterbody (Figure 6a). As shown in Figure 6b, the sampling sites of clean water were concentrated in green regions, while the polluted water was concentrated in red regions. In this study area, the samples in the green region were distributed in suburbs or wetland parks, the water quality was purified by the biological interception and absorption. The samples in the red region were distributed near the downtown and residential quarters. More nitrogen was discharged into the water environment in runoff due to the increased sewage and inadequate sewage treatment system. Gradually, in the long-term, the nitrogen pollutants were stored in the water and sediment environment, possibly aggravating the water eutrophication under special conditions.

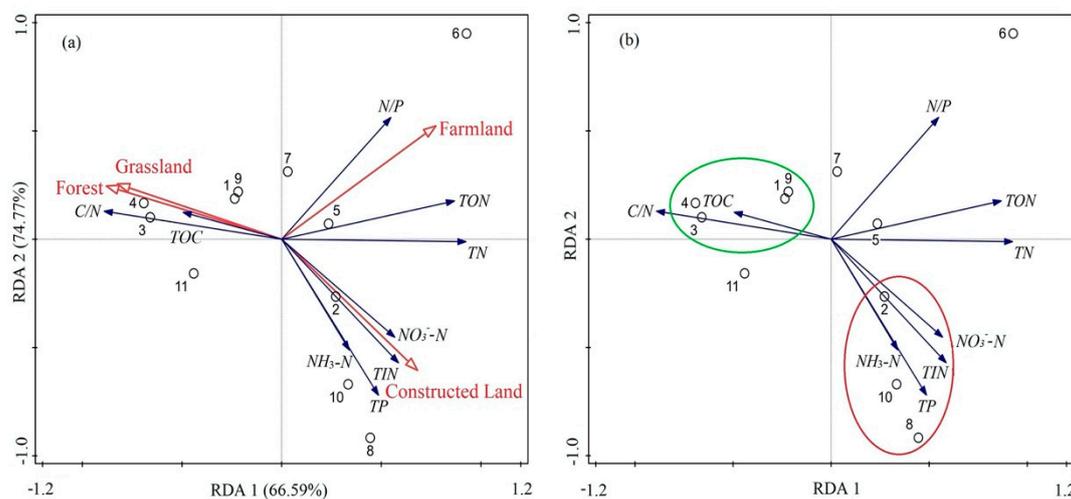


Figure 6. Redundancy analysis (RDA) Biplot of Water Quality with Land Use Types (a), Sampling Site (b).

In spatial scale, land use may have dual influences in the water environment. Then, in the long-term scale, we question how the land use change combining with the anthropogenic activities impacts the nitrogen load between the water and sediment. To better deal with this, long-term monitoring data of TN concentrations in Taliyu lake and Liangxi River were obtained from the Taihu Basin Authority of Ministry of Water Resources and Taihu Laboratory for Lake Ecosystem Research [37,53]. The annual variations of TN losses have a good consistency with the TN concentrations between the rivers and lake (Figure 7). A downward trend of water nitrogen loads was observed, indicating that the government's water environment treatment measures achieved a good result [29,31]. But after 2014,

the TN concentrations have been increasing in Liangxi River as the hydrodynamic force of this river was mainly controlled by the dam. Larger amounts of pollutants input from the land use were blocked and accumulated in the river. When the dam was open, the clean water flowed through the urban area, the pollutants discharged from the land use were finally transported into the Beijing-Hangzhou Grand Canal (Figure S3). Then, the polluted degree of Liangxi River was alleviated to some extent. This is why the water quality of Liangxi River is better than that of Mali River and Liangtang River on the whole.

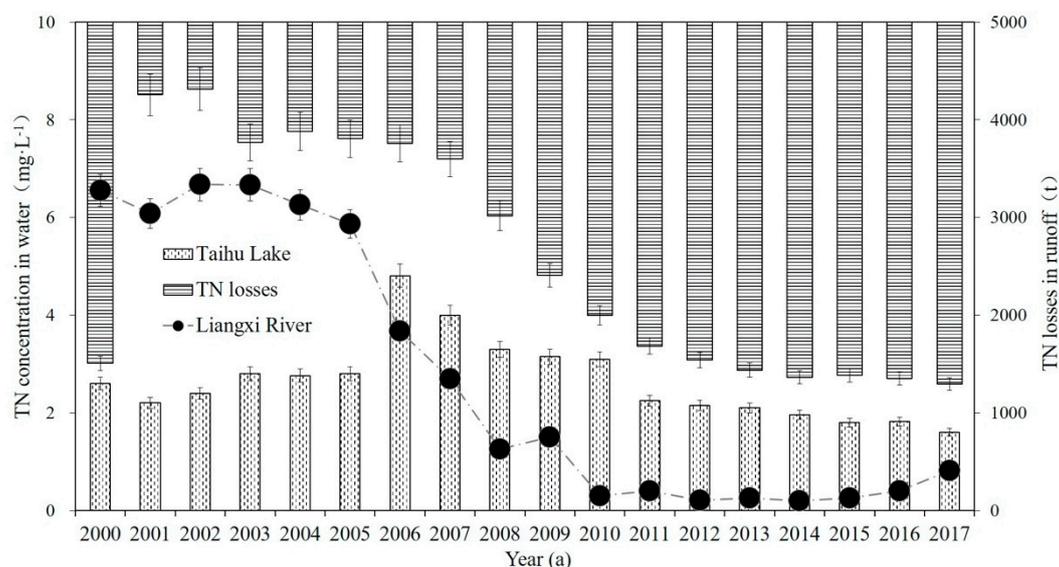


Figure 7. Comparison Between the TN Losses in Runoff Due to Multiple Factors and the TN concentrations in water.

Furthermore, around the Liangxi River, the dominant land use type (Figure 4 and Figure S2) was constructed land (63.6%), which mainly originated from farmland (43%) and water areas (39%). The constructed land and farmland were regarded as the nitrogen source for the waterbody, aggravating the water pollution. The forest-grassland acted as the nitrogen sink, improving the water quality. The amount of TN losses in runoff from the land use change exhibited an increasing trend in Liangxi River basin due to the rapid increase of constructed land area. The amount of TN losses discharged from land use in this basin also increased with multiple-factors (e.g., domestic sewage, runoff of the constructed land and agricultural activities). In addition, the water quality of Liangxi River deteriorated gradually from upstream (TN = 1.44 mg·L⁻¹) to downstream (TN = 2.00 mg·L⁻¹), while the nitrogen and phosphorus concentrations in the tributary were significantly higher than those in the trunk stream ($t = 3.27$, $p = 0.007$). These all indicated that the water quality of Liangxi River was profoundly affected by the discharge of exogenous nitrogen in the long term [16,17]. Although there has a dense river network, the hydrological connectivity of rivers and lakes were completely controlled by dams. Thus, the TN accumulated near the closed dam in the estuary, further aggravating the water pollution. Consequently, long term monitored data combined with the analysis of multiple factors should be given more attention in future land use management and water environmental treatment.

4. Conclusions

In this study, long-term land use data were used to analyze the land use change effects on nitrogen transport between the waterbody and sediment. Some obviously spatial-temporal distribution characteristics between the land use types and their transitions were observed during 1990–2017. On the whole, the areas of farmland decreased constantly while those of constructed land have increased rapidly. Most of the constructed land originated from farmland, making the constructed land become

the dominant land use type, and its proportion has increased from 23.85% in 1990 to 61.72% in 2017. Rapid urbanization and environmental policy adjustments were regarded as the main driving factors of land use change in this area.

The TN losses in runoff have been significantly affected by the land use types and their transitions. Of these, the forest and grassland, as sinks of nitrogen in the waterbody, are effective at absorbing and intercepting nutrients. The constructed land and farmland, as the source of nitrogen in the waterbody, were the dominant contributors (62%–98%) of TN losses from surface runoff. With the coupling of dam construction, hydrodynamic force and anthropogenic activities, the TN losses discharged from different land use types affected the nitrogen transport, resulting in the aquatic habitat degeneration. The nitrogen balance between the water and the sediment were broken by the land use change and the associated anthropogenic activities. Meanwhile, the role of sediment as a source and/or sink for nitrogen in the waterbody has been changed and the dominant contribution of endogenous and exogenous pollution may also be alternate.

Considering the high nitrogen losses from surface runoff, improving the sewage treatment rate and rainfall-wastewater distributary system may be the primary measure in future water environmental management. In addition, we also suggest that more green space (e.g., wetland parks and vegetative buffer strips) should be arranged on the river band, lakeside and estuarine regions. Meanwhile, the water cycle should be accelerated to improve the water self-purification ability in the polluted river through the dam regulation. These suggestions and practices are not only suitable for the treatment of the local environment, but they should also provide a scientific reference for the management of land use change and nitrogen transport internationally.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2071-1050/12/9/3895/s1>, Figure S1: Different Types of Gates and Gams, Figure S2: Spatial Variation of Land Use Types from 1990 to 2017, Figure S3: Location of Sampling Site, Table S1: Coefficients of Various Pollutants Generated and Discharged into Rivers, Table S2: TN Concentrations of Water Samples ($\text{mg}\cdot\text{L}^{-1}$) and Sediment Samples ($\text{g}\cdot\text{kg}^{-1}$) in the Windward Lakeshore Estuary, Table S3: Variation of TN and TP Concentrations under Different Stirring Speed by Laboratory Simulated Experiments, Table S4: Classification Accuracy of Land Use in each Years.

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