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Variations of Farming Systems and Their Impacts on Surface Water Environment in Past 60 Years in Intensive Agricultural Area of Taihu Region, China

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Abstract

Based on agricultural nitrogen (N) balance model and field experiments, the impacts of farming system changes of Taihu Region of China on surface water environment were studied. During past 60 years, farming systems changed greatly in Taihu Region. The traditional method of manure collection and application was replaced by chemical fertilizer utilization gradually. Chemical N fertilization intensity decreased greatly due to the abolition of "3 crops per year" and reduction of cropland area in 1990-2010. Crops depleting soil fertility increased, while those improving soil fertility decreased, leading to an excessive dependence on chemical fertilizer application, which increased the risks of soil N loss to surface water environment in Taihu region. However, field experiments showed that the agricultural N loss with runoff only accounted for 2% of fertilizer N application rate. The majority of N was exported by crop harvesting. Our findings showed that the agricultural N loss might not be the main source of N pollution in Lake Tai after 2000. To control N pollution of Lake Tai, more attention should be paid to industrial and domestic wastewater from urban and rural areas, wastes from livestock and poultry breeding, bait input for aquaculture, etc in the Taihu Region, China.

Keywords

Farming Systems, Agricultural Nitrogen Balance, Fertilization, Environmental Pollution, Taihu

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Region, China

1. Introduction

Land Use and Land Cover Change (LUCC) is the core of global environmental change researches. According to the UN Agenda 21 in 1992, LUCC research will be the focus in the 21 century ([1]-[3]). Land use changes cause variations of geographical processes and surface landscape structures, greatly impacting on N biogeochemical cycles and material transport, and produce a series of water environmental and ecological problems ([4]-[7]). Water issues caused by LUCC and its impacts on N cycles have been a highlight of the world and have caused a significant concern ([8]-[12]). On basin scale, the impacts of LUCC on N cycles are based on three aspects: First is that the land use intensity changes, including farming intensities, multiple-cropping index, returning cropland to forest/grassland/lakes, abandoned cultivation, etc. Second is the impact of LUCC on hydrological process. Hydrological process is a basic driving force for N entering from land to river as well as to groundwater. Third is the impact of LUCC on the N transportation and formation by changing soil erosion process. All these three processes affect water environment of receiving water, and may finally lead to water eutrophication.

Nutrient cycles link agricultural systems to their societies and surroundings. The inputs of nutrients (mainly N and P) are essential for high crop yields [8]. While it is reported that more than 80% of fertilizer N applied in agricultures do not make it into crops worldwide [8]. This indicates a huge imbalance of N in agriculture lands. The surplus N accumulates in soils, goes on to enter rivers, and is transported downriver to lakes, which causes harmful algal blooms, water quality degradation, and reduction of aquatic species ([13]-[16]).

Cropping system is one of the most important factors influencing agricultural nutrient cycle [14]. It is a comprehensive system for crop planting and related technical measures, including tillage, cultivation, fertilization, irrigation, weeding, rotation, soil and water conservation and crop protection, etc [11]. Under different cropping systems, N is redistributed in crops, soil and water, impacting potentially on surface water environment ([12] [14] [16]). Previous studies have pointed out the vitally important impacts of agricultural nonpoint sources pollution on surface water quality ([10] [17] [18]). While great changes have taken place in farming systems of Taihu region during past 60 years. With the urbanization process, the water quality of Lake Tai (China's third largest freshwater lake) has deteriorated since the 1980s [19]. Is agricultural nonpoint sources pollution still the dominant cause leading to water pollution of Lake Tai? In this study, the variations of farming systems in Taihu region, Jiangsu province, China, and their potential impacts on surface water environment over past 60 years are analyzed. The objectives of this study are to re-examine the role of agricultural N imbalance in affecting surface water environment due to the changes of cropping systems in most developed area of China, and to provide a new perspective to study nitrogen sources of water pollution in developed area.

2. Materials and Methods

2.1. Study Area

The Taihu region (119˚13'E - 121˚19'E, 30˚46'N - 32˚14'N) is situated at the heart of Yangtze River Delta. It covers an area of 1.76×10^4 km^2, accounting for 47.7% of the total area of Taihu Basin. Four districts (Suzhou, Wuxi, Changzhou and Zhenjiang) and two counties (Danyang and Jurong County of Zhenjiang district) are included in this region (**Figure 1**). It is one of the most developed areas in China with high population density, urbanization, and economic development [20]. In 2012, it contributes 1.02% of national agricultural production (~8.48×10^9\$), with national 0.53% farmland and 0.36% rural labor force. The number of population exceeded 1.62×10^7. The population density of 920 persons per km^2 was much higher than that of whole nation (141 persons per km^2). The farmland area decreased from 872 kha in 1949 to 509 kha in 2012; while the per capita area of cultivated farmland decreased from 0.13 ha in 1949 to 0.03 ha in 2012. The GDP (Gross Domestic Production) was 3732×10^8\$ (1\$ = 6.31 RMB) in 2012, accounting for 43.6% of that of Jiangsu province and 4.5% of that of China ([21]-[24]).

Low-lying areas from east to west, consists of plain, hilly mountains and water respectively occupying 58.3%, 14.2% and 17.5% of Taihu region area. It belongs to the north subtropics monsoon climate, and is very vulnerable to extreme climate and season fluctuation [12]. The annual average air temperature is 15˚C - 17˚C. The

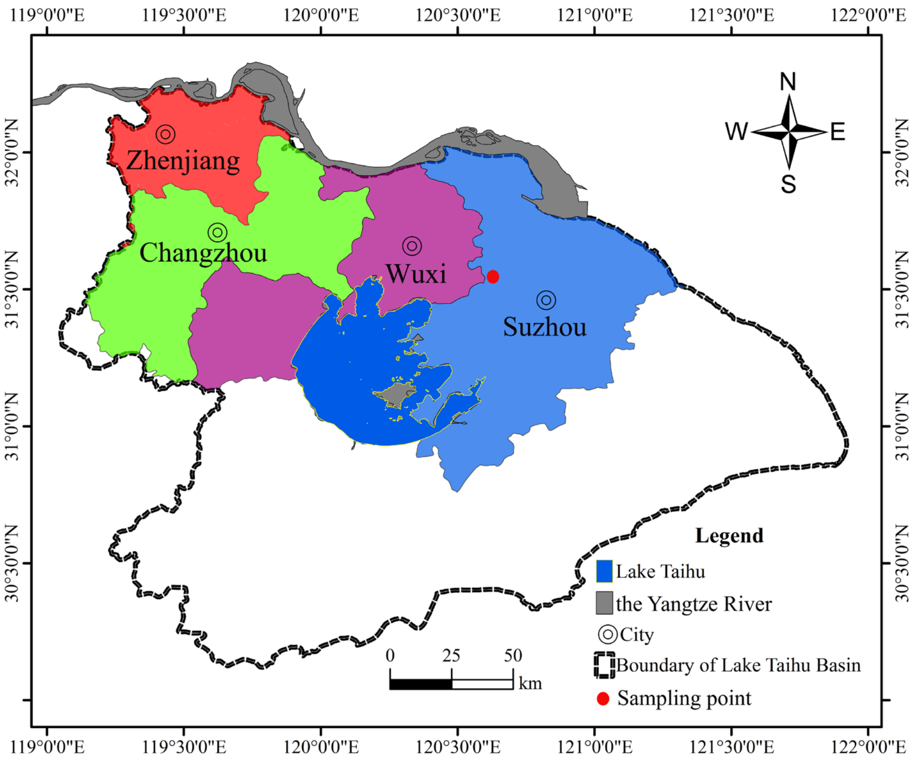

Figure 1. Map of Taihu region, Jiangsu, China.

annual mean precipitation is 1100 - 1400 mm, with rains being unevenly distributed throughout the year. The wet season (May-September) accounts for 60.5% of total rain in a year. Water and heat over the same period and abundant heat resources are beneficial to crop growth in subtropical areas. Paddy rice is the dominant crop, and the other major crops are wheat, rapeseed, and soybeans. Cropping systems is two crops per year with rice-wheat being most popular in this region.

2.2. Nitrogen Budgets

Agricultural nitrogen budgets were calculated based on agricultural nitrogen balance model according to Roy [25].

$$N_{surp} = N_{input} - N_{output} = \left(N_{fe} + N_{ma} + N_{dep} + N_{fix}\right) - \left(N_{hav} + N_{gas}\right) \tag{1}$$

N_{surp} is defined as the ratio of yearly N surplus and crop acreage in Taihu region. N_{input} is nitrogen input from outer sources, including fertilizer application (N_{fe}), manure application (N_{ma}), atmospheric nitrogen deposition (N_{dep}) and nitrogen fixation (N_{fix}); N_{output} is nitrogen output from agricultural system, including N export by harvesting (N_{hav}), N loss to atmosphere through ammonia volatilization and denitrification (N_{gas}). All terms are in unit of kg N ha^{-1} yr^{-1}.

Input: *Nitrogenous fertilizer use and manure N input*

N_{fe} is defined as the ratio of annual N fertilizer utilized to cropping land. Data of all N fertilizer forms were converted into element N to estimate N fertilizer input. N_{ma} is calculated as animal manure N input divided by crop acreage of Taihu rigion. Manure N applied (N_{ma}) in 1990s was calculated as 1/9 of N_{fe} [17]. N_{ma} after 2000 was set to 0 because no manure was applied to cropland since 2000 in Taihu region.

Input: *Atmospheric N deposition*

N_{dep} is atmospheric N deposition including dry and wet deposition of nitrogen. According to [26], atmospheric N wet deposition was 30.2 kg N ha^{-1} in 2003-2004 in Tahihu region. And the ratio of atmospheric dry and wet N deposition was 0.26:1 [27]. Thus we calculated the total atmospheric N deposition to be 38.0 kg N ha^{-1} a^{-1}. We use this value to represent N_{dep} in 2000s. N_{dep} in 1980s is set to 1/5 of N_{dep} in 2000s according to [28]. That is

7.6 kg N ha^{-1} a^{-1}. N_{dep} in 1990s was obtained by averaging the atmospheric N deposition in the 2000s and in the 1980s. That is 22.8 kg N ha^{-1} a^{-1}.

Input: *symbiotic N fixation*

N_{fix} is defined as the ratio of symbiotic N fixation in paddy field and crop acreage of Taihu region. Symbiotic N fixation by microorganisms in paddy field was obtained by multiplying rice planting area and N fixation ratio of rice which is set to 45 kg N ha^{-1} according to [29].

Output:

N_{hav} is defined as the ratio of yearly harvested N and crop acreage of Taihu region. It is obtained as follows.

$$N_{hav} = \left(\sum_{i=1}^{2} Y_i \times r_i \right) \Big/ S \tag{2}$$

Y is crop yield of Changzhou, Suzhou and Wuxi County. $i = 1, 2$. 1 represent rice, 2 represent wheat. r is crop N absorb ratio in growing season, it is set to 0.019 and 0.03 ([30] [31]). S is the sum of agricultural acreage of Changzhou, Suzhou and Wuxi County.

N_{gas} is defined as the ratio of N export to atmosphere through ammonia volatilization and denitrification and cropland acreage of Taihu region. N_{gas} is set to 45% of N fertilizer application respectively according to [32].

2.3. Data Sources

Data of N fertilizer intensity, crop planting area, crop yield, arable land area, land area, population size, GDP in 1985-2010 were obtained from Statistical Yearbooks of Changzhou, Suzhou, Wuxi and Zhenjiang ([21]-[24]).

Data of the above parameters in 1949-1984 were obtained from Wuxi County [33].

Data of agricultural N loss through runoff was obtained from field experiment carried out in a typical cropland of the Taihu Region in 2012-2013.

2.4. Field Experiment

The field experiment was established at the Changshu Agroecological Experimental Station, Institute of Soil Science, Chinese Academy of Sciences (31˚33'N, 120˚42'E) in Nov 2012-Nov 2013 (**Figure 1**). The station is on the northeast side of Taihu Lake, within 32 km of the shore. A rice-wheat cropping rotation has been dominantly adopted here.

The experiment utilized one field with an area of 15 by 10 m. The field was divided into 2 sample plots, with 3 replications per plot. One represents farming systems in 1980s. The other represents 2000s. The experiment was started in Nov. 2012, the beginning of the wheat season, and continued for one integrated rice-wheat rotation. In the 2000s sampling plot, chemical fertilizers including 50% urea, 30% ammonium carbonate and 30% compound fertilizer were applied at rates of 300 kg N ha^{-1} in the rice season and 250 kg N ha^{-1} in the wheat season. In the 1980s sampling plot, 70% ammonium carbonate and 30% organic fertilizer (pig manure) were applied at rates of 188 kg N ha^{-1} in the rice season and 157 kg N ha^{-1} in the wheat season. For N application, 30% was basally applied, 40% was topdressed at the tillering stage, and the remaining 30% was topdressed at the ear differentiation stage for each crop. 6 pots were installed near the experimental plots to collect runoff from the field. By doing this, the results of this experiment were expected to appropriately reflect the conventional farming systems in 1980s and 2000s in the Taihu Lake Region. Total nitrogen concentration of runoff samples were analyzed by an ultraviolet spectrophotometer.

3. Results

3.1. Variations of Farming System in Taihu Region

As is shown in **Table 1**, cropping system in Taihu region changed from 2 crops (winter wheat-summer rice) per year to 3 crops (winter wheat-summer rice-autumn rice) per year in 1949-1978. Rice-wheat rotation was the dominant cropping system in this region in 1950s. Planting area of rice and wheat accounted for 42.6% and 46.4% of the total cropping acreage, respectively. In the middle of 1960s, 3 crops per year expanded to meet people's increasing demand for food. Until the middle and late of 1970s, 3 crops per year were very popular in the Taihu region. This becomes evident from the fact that almost 100% of the cropland in Wuxi County in 1975-78 was used for 3 crops per year and 86% in Suzhou County in 1976. The planting area of "3 crops per year" decreased and even disappeared during 1979-1985. Rice-wheat and rice-rape were most popular in 1980s, with the proportion of 80% and 13% of the total cropland, respectively. In 1990-2012, rice and wheat occupied over 90% of the total planting area of food crops in the Taihu region. The planting area of food crops (mainly rice and wheat) decreased significantly due to the large amount of cropland was transformed into construction land from 1990 to 2003. With the implementation of farmland protection policy, it stabilized in 2003-2010 (**Figure 2**, **Figure 3**).

The ratio of food crops and cash crops was 9:1 in 1949-1978. Cash crops include green manure crops, vegetables, rapes, potatoes, melons, beans, etc. Among the cash crops, green manure crops were conductive to maintain soil fertility and to guarantee grain yield due to strong ability of symbiotic N fixation. Therefore, the planting area of green manure crops expanded significantly year by year, occupying 35.2% - 69.7% of total in 1949-1978. After 1978, the ratio of food crops and cash crops decreased gradually. The ratio was 8:2, 7:3 and 6:4 in 1980s, 1990s and 2000s, respectively (**Table 1**). Compared with food crops, the planting area of cash crops changed relatively slowly in 1990-2010 (**Figure 2**). The proportion of vegetable lands and total planting area increased from 5.2% in 1990 to 22.2% in 2010. The rape area changed stably in 1990s, but decreased significantly in 2000s (**Figure 3**). The rape area only occupied 4.7% of total planting area in 2010. Cotton and green manure crops area decreased while vegetables increased during 1990-2012 (**Figure 3**). Farmers prefer planting cash crops to planting food crops because cash crops have higher commercial value.

Fertilization intensity in this study refers to the number of N fertilizer utilized to cropland in unit of kg N ha^{-1}. As shown in **Figure 4**, fertilization intensity increased from about zero in 1949 to above 700 kg N ha^{-1} in 1978, showing an increasing trend. After the implementation of family contracted responsibility system in 1978, fertilization intensity in Taihu region increased sharply to 437 kg N ha^{-1} in 1985 due to the abolition of "3 crops per

Table 1. Farming system in different periods in Taihu region, Jiangsu Province, China.

Periods	Cropping system	Food crops/Cash crops	Fertilization Intensity	Fertilization Structure	
				Organic/Inorganic fertilizer ratio	N/P/K
1949-1978	2 to 3	9:1	98-above 700 kg N ha^{-1} a^{-1}	9:1	From 1:0:0 to 1:0.29:0.09
1979-1989	3 to 2	8:2	738 - 437 kg N ha^{-1} a^{-1}	3:7	1:0.23:0.005
1990-now	2	from 7:3 to 6:4	578 - 445 kg N ha^{-1} a^{-1}	1:9 to 0:1	1:0.08:0.13

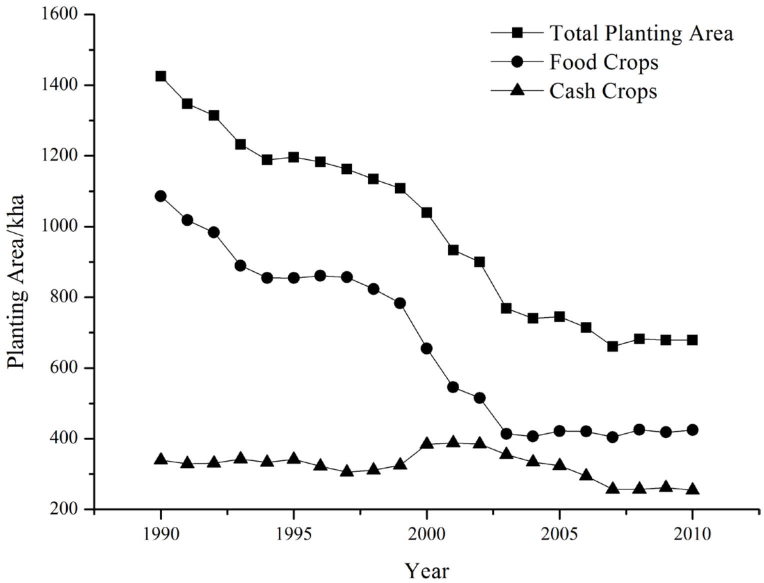

Figure 2. Yearly changes of planting area of food crops and cash crops in Taihu region in 1990-2010.

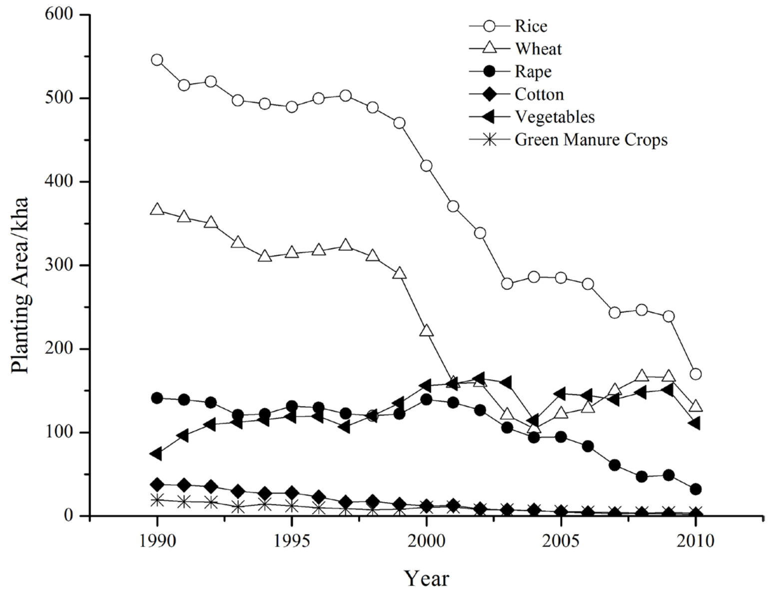

Figure 3. Yearly changes of planting area of different crops in Taihu region in 1990-2010.

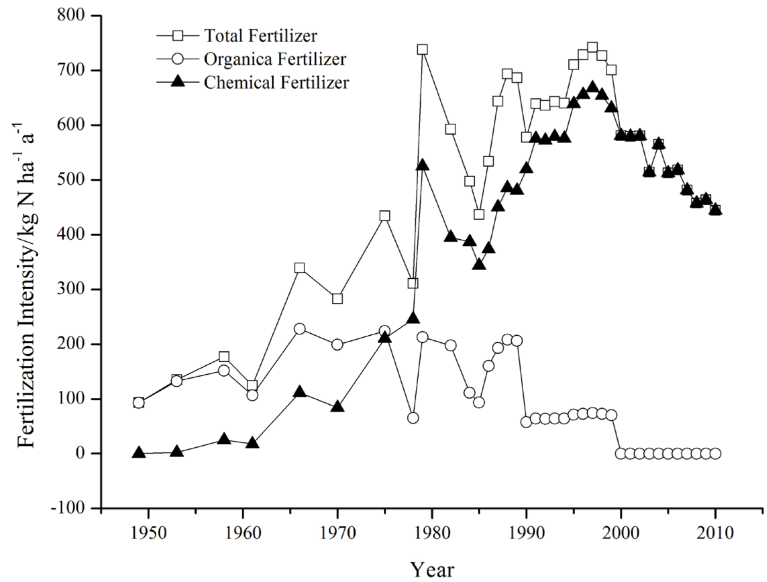

Figure 4. Yearly Changes of fertilizer use intensity in Taihu region, Jiangsu Province, China in1949-2010.

year" [34]. While it increased to 687 kg N ha^{-1} in 1989 due to the increase utilization of chemical N fertilizer. Fertilizer utilization represents an increasing trend in 1990s with the highest value of 742 kg N ha^{-1} in 1997. While it shows a decreasing trend in 2000s with the lowest value of 445 kg N ha^{-1} in 2010, which is still far exceeding the local optimum amount of N fertilizer application ([34] [35]). It has become a common problem to utilize excessive chemical fertilizer in agricultural area of Taihu region.

Fertilization structure refers to the ratio of organic fertilizer and chemical fertilizer or the proportion of net nitrogen, phosphorus and Potassium in chemical fertilizer applied to cropland. Organic fertilizer includes manure from livestock, poultry and human being, waterlogged compost, green manure, methane fermentations waste etc. Chemical fertilizer includes ammonium bicarbonate, urea, compound fertilizer, ammonia water, phosphorus fertilizer and potassium fertilizer etc. In 1949-1978, organic fertilizer was collected and applied to cropland from many sources such as straw compost, human waste, rubbish, mud mixed compost and biogas fertilizer etc. It occupied about 90% of agricultural area of Taihu region in 1949-1978. After the 1978 reform, a large number of farmers moved to township enterprises to work. Moreover, it's hard and insanitary to collect and ret organic manure. Therefore, organic fertilizer was replaced by chemical fertilizer gradually. The ratio of organic fertilizer and chemical fertilizer was 3:7, 1:9 and zero in 1980s, 1990s and 2000s. Chemical N fertilizer was received adequate attention while P and K fertilizer were lack of attention during 1949-1978. N and P were excessive and K was lack in soil during 1979-1990. With more and more attention paid to water environment caused by excess N input, P and K fertilizer application have been gradually appreciated recently. Chemical N, P and K fertilizer accounted for about 84.6%, 6.0% and 9.4% of the total chemical fertilizer respectively.

3.2. Agricultural N Balance in Taihu Region

Changes of agricultural N input, output and surplus in the Taihu region since 1949 were shown in **Table 2**.

Manure N has ever been the biggest contributor of agricultural N in 1949-1978. But the contribution decreased after 1978 until organic fertilizer was fully replaced by chemical N fertilizer in 2000 (**Table 2**). The increase of atmospheric N deposition indicates that air pollution became worse in 2000s. Biological N fixation decreased due to the reduction of cropland. Among the N exports, crop N export by harvesting was the main way for N exports in 1949-1978. But the ratio decreased evidently to 38% in 1990-2010. However, the ratio of gas N loss increased from 24.6% in 1949-1978 to 47.6% in 1979-1989. It varied stably in the recent 20 years with the average value of 53.5%. The ratio of N loss by runoff and leaching showed a slightly increasing trend during the period (**Table 2**). Agricultural N budget of the Taihu region has been in surplus since 1949. The agricultural N surplus was highest in 1979-1989 and lowest in 2000-2010.

3.3. Agricultural N Loss through Runoff from Field Experiments

Runoff samples were collected for 7 times separately from 1980s and 2000s sample plots. 3 times were in the wheat season and 4 times in the rice season. Results of agricultural N loss with runoff for the 1980s and 2000s sample plots are shown in **Table 3**. Agricultural N loss with runoff in 2000s plot was evidently higher than that in 1980s plot in both the wheat season and rice season due to the increase of fertilization intensity. The N loss with runoff was much lower than N fertilization application intensity (1980s: 345 kg N ha^{-1}; 2000s: 550 kg N ha^{-1}) for both fields. It only accounted for about 2% of chemical N fertilization intensity in one wheat-rice rotation, playing a minor role in agricultural N balance. The main pathway of N export is crop harvest. Field experiments showed that N export by wheat harvesting was 117 kg N ha^{-1} and N export by rice harvesting was 147 kg N ha^{-1} for 1980s plot, accounting for about 74.5% and 78.2% of fertilizer N input, respectively. For 2000s

Table 2. Nitrogen balance sheet of the Taihu region, Jiangsu province, China.

	Period	1949-1978 kg N ha^{-1}	1979-1989 kg N ha^{-1}	1990-1999 kg N ha^{-1}	2000-2010 kg N ha^{-1}
Input (A)	N_{fe}	67.8 (23.4)	333.9 (59.1)	492.5 (79)	374.3 (86.1)
	N_{ma}	169.2 (58.4)	178.0 (31.5)	73.2 (11.8)	0 (0)
	N_{dep}	7.6 (2.6)	7.6 (1.3)	22.8 (3.7)	38 (8.7)
	N_{fix}	45 (15.5)	45 (8)	33.5 (5.4)	22.4 (5.2)
	Subtotal	290 (100)	564 (100)	622 (100)	435 (100)
Output (B)	N_{harv}	88.7 (71.6)	142.3 (45)	157 (38)	120 (38.1)
	N_{gas}	30.5 (24.6)	150.2 (47.6)	221.6 (53.6)	168.4 (53.5)
	N_{loss}	4.7 (3.8)	23.4 (7.4)	34.5 (8.3)	26.2 (8.3)
	Subtotal	123.9 (100)	315.9 (100)	413 (100)	314.6 (100)
Balance (A-B) Nsurp		165.7	245.8	208.9	120.1

Contribution percentage shown in parentheses.

Table 3. Agricultural N loss through runoff in Changshu Station.

Season	Sampling Date	A TN Concentration mg/L	B Runoff L	C = A*B/1600 N Loss through runoff kg/ha
1980s wheat season	2012.11.23	24.7	141.9	2.2
	2012.12.30	19.9	141.0	1.8
	2013.02.26	24.3	138.6	2.1
	sum			6.1
1980s rice season	2013/7/6	2.3	176.0	0.3
	2013/7/21	3.8	125.0	0.3
	2013/8/19	3.1	143.0	0.3
	2013/8/29	2.9	162.0	0.3
	sum			1.2
2000s wheat season	2012.11.23	27.0	141.9	2.4
	2012.12.30	21.6	142.6	1.9
	2013.02.26	50.5	142.6	4.5
	sum			8.8
2000s rice season	2013/7/6	2.7	176.0	0.3
	2013/7/21	4.2	166.0	0.4
	2013/8/19	17.1	166.0	1.8
	2013/8/29	7.5	168.0	0.8
	sum			3.3

plot, the N export was 157 kg N ha^{-1} in wheat season and 176 kg N ha^{-1} in rice season, accounting for about 62.8% and 58.7% of fertilizer N input, respectively. Field experiments showed that N loss through runoff plays a minor role in agricultural N balance and crop harvesting is the main pathway of N export in both 1980s and 2000s.

4. Discussion

With the process of urbanization in the Taihu region, large quantity of cropland was turned into construction land [36]. Township enterprises developed quickly in Taihu region. A large number of farmers entered township enterprises to work, leading to the lack of labor force in rural area. To obtain optimum grain yield and higher income, farmers changed planting systems, fertilization intensity and structure. But the negative impacts of cropping system changes on surface water environment were ignored.

4.1. The Impacts of Farming System Changes on Surface Water Quality

Green manure crops have ever playing an important role in cash crops in Taihu region. It is found that planting green manure crops such as *Astragalus sinicus L* can not only replenish nutrients to agricultural system, but also reduce environmental pollution ([37]-[39]). But comparing with chemical fertilizer application, more labor forces are needed for green manure crops cultivation. Vegetables are another one of the most important cash crops in Taihu region. The planting area of vegetables expanded significantly year by year to meet requirement of people's lives. It was very common to utilize 600 - 1300 kg N ha^{-1} yr^{-1} to obtain 2 - 3 seasons of vegetables yielding in a year, thus consuming plenty of chemical fertilizers ([40] [41]). But only 21% - 36% of chemical fertilizer N was exported by vegetables harvesting. Because the roots of vegetables are very shallow, it is easy for soil nitrate to loss with runoff and to leach with interflow when encountered with rain storm or excessive irrigation. It's found that the amount of N loss from vegetable land is much higher than that in grain land which is more harmful to water environment ([42] [43]). Therefore, change of crop planting structure is one of the main reasons causing excessive chemical fertilizer application in Taihu region.

With the decrease of cropland area and the understanding of agricultural N pollution, less chemical fertilizer was applied to croplands in 2000s. Present fertilization intensity (rice season: 300 kg N ha^{-1}; wheat season: 250 kg N ha^{-1}) are far exceeding the optimum demands of crops for nitrogen (rice season: 150 kg N ha^{-1}; wheat season: 225 kg N ha^{-1}) in Taihu Region ([28] [44] [45]). The increase of N fertilization intensity did not lead to higher crop yields, but caused serious N loss to environment two times that optimum N fertilization [46]. It is found that significant positive linear correlation relationships exist between agricultural N loss and fertilization intensity at the Taihu basin ([27] [47]). Under different N fertilization level, total N concentration in soil solution in −20 - −40 cm top soil layer correlates with chemical N fertilization intensity significantly in paddy field of Lake Tai basin. Long term excessive application of chemical fertilizer increases the risks of soil N loss to surface water environment ([38] [46] [49]. Field experiments also show that TN loss with runoff increased evidently with the increase of fertilization intensity in both wheat season and rice season (**Table 3**).

4.2. Changes of N Pollutant Sources of Lake Tai Water

Agricultural N balance in Taihu region has been in surplus since 1949 (**Table 2**). Although the average agricultural N surplus in 1949-1978 was high (~165.7 kg N ha^{-1}) (**Table 1**), soil N loss was very low due to the high capacity of soil to retain fertilizer N. In this period, organic fertilizer occupied most of total fertilizer N input. The retention rate of organic fertilizer in soil is high. For example, almost 60% - 64% of total nitrogen in livestock manure is retained in soil of paddy field [50]. The nutrients were absorbed by crops for growth. Therefore, agricultural N loss was so weak that it did not impact surface water quality negatively. The water quality of Lake Tai was at Grade I or Grade II in 1949-1978, belonging to clean water.

In late 1978, the implementation of rural household contract system mobilized farmers' enthusiasm for production. More chemical N fertilizer was applied to croplands. Although the fertilization intensity decreased, chemical N fertilizer loss increased because of its soluble characteristics, causing negative effect on surface water environment. Water quality of Lake Tai deteriorated from Grade III in 1980s to Grade IV in 1990s. While in 2000s, chemical N fertilization intensity decreased significantly, the water quality of Lake Tai became worse to Grade V. It indicates that N pollutant sources of Lake Tai may have changed since 2000.

There are many other pollutant sources of Lake Tai, such as industrial and domestic waste water discharge from urban and rural areas, pollutant from intensive feeding of livestock and poultry. While strict measures were implemented to reduce urban and industrial wastewater discharge to Lake Tai after 1998. More than one thousand heavy pollution enterprises in Taihu region were forced to reach drainage standards before discharge. But the Water quality of Lake Tai still deteriorated in 2000s. On one hand, it is probably because the total discharge of urban and industrial wastewater increased along with population growth and economic development. Comparing with 1990, the total population in Taihu Region in 2012 increased 180 million, and the GDP increased 50 times. On the other hand, wastewater discharge from livestock and poultry feeding, excretion from human and animals, and bait input for aquaculture in rural areas increased N pollution in Lake Tai. Along with the improvement of people's living standard in Taihu region, the demands for meat and eggs increased sharply, which promoted the development of intensive livestock and poultry feeding. But only a small amount of manure was processed into organic fertilizer and applied to croplands. A large quantity of manure was stacking randomly which is easily to loss with rainstorm runoff. By using the method of N isotope, Xing *et al.* [51] found that the ratio of nitrogen isotope in rivers of Taihu Region was much higher than that in chemical N fertilizer but close to that of N in the human and animal wastes. They suggested that river N pollutants come mainly from domestic sewage, rural human and animal wastes and industrial wastewater. In this study, based on agricultural N balance, the calculated agricultural N loss (N_{loss}) accounted for only 8.3% of N export in 1990s-2000s. Based on field experiments carried out in Changshu in 2012-2013, calculation shows that the net amount of agricultural N loss with runoff (12.1 kg N ha^{-1} a^{-1}) accounted for only 2% of the N application rate (550 kg N ha^{-1} a^{-1}). While the net amount of N export by harvesting (333 kg N ha^{-1} a^{-1}) accounted for about 60.5% of the N application rate (550 kg N ha^{-1} a^{-1}) for the 2000s sample plot. It indicates that agricultural N loss may not be the main source of

Lake Tai in 2000s. The other N sources of Lake Tai, such as industrial and domestic wastewater in urban and rural areas, waste from human and animals and aquatic breeding industry pollution should be paid more attention.

5. Conclusion

Calculated agricultural N surplus decreased significantly from 1990s to 2000s, while the water quality of Lake Tai deteriorated year by year. Field experiments showed that agricultural N loss with runoff was very low, being only about 2% of fertilizer N application rate. A large quantity of N was exported by crop harvesting. It indicated that agricultural N loss might not be the main source of N pollution in Lake Tai during 1990s-2000s. It was suggested that in highly industrialized and urbanized areas, the N pollutant sources such as industrial and domestic wastewater from urban and rural areas, wastes from livestock and poultry breeding, bait input for aquaculture, etc. should be paid more attention.

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