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Excess nutrient loads to Lake Taihu: Opportunities for nutrient reduction

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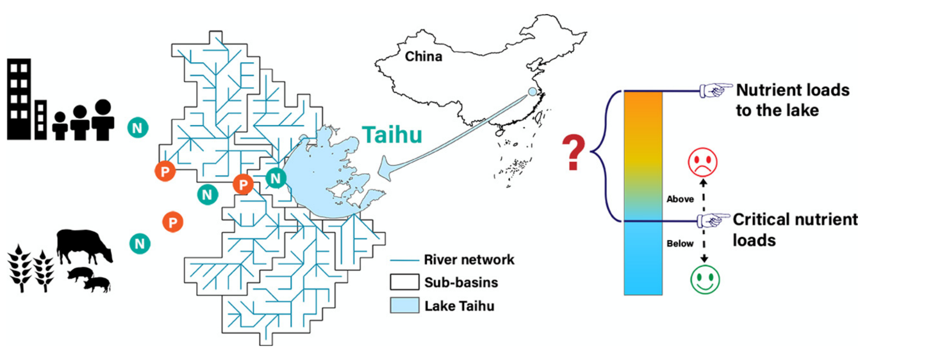
HIGHLIGHTS

- Rivers discharged around 61 kton of TDN and 2 kton of TDP to Lake Taihu in 2012.
- Over half of TDN and TDP loads were from Sub-basins I (north) and IV (south).
- Diffuse sources contributed 90% to TDN and point sources 52% to TDP to Lake Taihu.
- To meet critical loads, river export of TDN and TDP needs to be reduced by 46–92%.
- Opportunities are reducing synthetic fertilizer and improving wastewater treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

Intensive agriculture and rapid urbanization have increased nutrient inputs to Lake Taihu in recent decades. This resulted in eutrophication. We aim to better understand the sources of river export of total dissolved nitrogen (TDN) and phosphorus (TDP) to Lake Taihu in relation to critical nutrient loads. We implemented the MARINA-Lake (Model to Assess River Inputs of Nutrients to seAs) model for Lake Taihu. The MARINA-Lake model quantifies river export of dissolved inorganic and organic N and P to the lake by source from sub-basins. Results from the PCLake model are used to identify to what extent river export of nutrients exceeds critical loads. We calculate that rivers exported 61 kton of TDN and 2 kton of TDP to Lake Taihu in 2012. More than half of these nutrients were from human activities (e.g., agriculture, urbanization) in Sub-basins I (north) and IV (south). Most of the nutrients were in dissolved inorganic forms. Diffuse sources contributed 90% to river export of TDN with a relatively large share of synthetic fertilizers. Point sources contributed 52% to river export of TDP with a relatively large share of sewage systems. The relative shares of diffuse and point sources varied greatly among nutrient forms and sub-basins. To meet critical loads, river export of TDN and TDP needs to be reduced by 46–92%, depending on the desired level of chlorophyll-a. There are different opportunities to meet the critical loads. Reducing N inputs from synthetic fertilizers and P from sewage systems may be sufficient to meet the least strict critical loads. A combination of reductions in diffuse and point sources is needed to meet the most strict critical loads. Combining improved nutrient use efficiencies and best available technologies in wastewater treatment may be an effective opportunity. Our study can support the formulation of effective solutions for lake restoration.

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1. Introduction

Lakes are important sources of freshwater for human needs in China. However, many Chinese lakes are eutrophied, as a result of over-enrichment of nutrients such as nitrogen (N) and phosphorus (P). Environmental problems caused by eutrophication are among the major concerns for future sustainable development in China (Le et al., 2010). A fast growing economy, rapid urbanization and increasing demand for meat production have resulted in increasing amounts of nutrients in Chinese rivers (Gu et al., 2015; Ma et al., 2012; Strokal et al., 2016b; Wang et al., 2018b). Rivers may export nutrients to lakes. Too much nutrients may deteriorate water quality and promote toxic algal blooms. Toxic algal blooms often lead to disrupted drinking water supply, health issues and odor nuisance for people living in the vicinity of the lake. As a result, the economic costs of algal blooms are estimated at millions of dollars each occurrence (Glibert et al., 2005; Le et al., 2010).

Lake Taihu (south-east China) is one of the Chinese lakes suffering from eutrophication (Qin et al., 2007). Lake Taihu is located in an area of strong socio-economic development (Le et al., 2010) (see also Section 2.1). Algal blooms in Lake Taihu have been reported since 1987 (Duan et al., 2009). Since then, the situation of eutrophication in Lake Taihu has worsened (Janssen et al., 2014). Wuxi's city is located in the Taihu basin. This city depends on Lake Taihu as the major drinking water source. In 2007, the algal blooms in Lake Taihu were so severe that Wuxi's city had to shut down its drinking water station. As a result, many people had to find other water resources for up to a month (Qin et al., 2010).

Many attempts have been made to reduce the eutrophication in Lake Taihu. Examples are the restoration of wetlands (Sun et al., 2015), mechanical removal of algae (Chen et al., 2012) and flushing with Yangtze water (Hu et al., 2008; Li et al., 2013). So far, these attempts have had limited effects. A possible reason for this is poor understanding of river export of N and P to the lake and their sources. Reducing external nutrient loads is a precondition for lowering Lake Taihu's algal growth sufficiently (Janssen et al., 2017; Xu et al., 2015). It was estimated that most of the nutrients in Lake Taihu were delivered by rivers draining into the lake, whereas only a minor part came from atmospheric deposition and sediment release (Xu et al., 2015). However, the relative shares of sources in these nutrient inputs are not known. A better understanding of the underlying causes of nutrient export by rivers to the lake is needed to formulate effective nutrient management options to reduce eutrophication in Lake Taihu.

A number of modelling studies exist on nutrient inputs to Lake Taihu from different sources (e.g., Huang et al., 2018, 2017; Lai et al., 2006; Liu et al., 2013). However, these studies often focus on particular areas (e.g., lowland polders), and on total N and P loads to the lake. Modelling studies that distinguish between nutrient forms (dissolved inorganic and organic) are lacking. However, distinguishing different nutrient forms is important for effective policy making because different nutrient forms may have different sources. For example, synthetic (mineral) fertilizers are often the main sources of dissolved inorganic N and P in rivers, whereas animal manure and sewage are often the main sources of dissolved organic N and P (Strokal et al., 2016a). Effective management of these different causes of eutrophication requires ability to differentiate between nutrient forms. For each nutrient form the loads can be compared with critical nutrient loads, which are the levels above which we may expect eutrophication problems (Janse et al., 2008). The difference between critical nutrient loads and the actual river export indicates the gap that needs to be closed to ensure a good ecological status of the lake. Moreover, combining this with the information on the sources and forms of N and P in the lake will largely contribute to the formulation of effective environmental policies. Such analyses are lacking for Lake Taihu.

The main objective of our study is, thus, to better understand the sources of river export of different nutrients forms (external nutrient loads) to Lake Taihu in relation to the critical nutrient loads. To this end, we implement the MARINA-Lake model (Model to Assess River Inputs of Nutrients to seAs) to quantify river export of N and P in different forms (dissolved inorganic and dissolved organic) from sub-basins draining into Lake Taihu. We focus on dissolved N and P, since the bioavailability for harmful algae is considered higher for dissolved than for particulate forms (Garnier et al., 2010). We link the results of the MARINA-Lake model to the PCLake model to analyze to what extent the river export of N and P exceeds the critical loads. Finally, we discuss the opportunities for nutrient reduction at the sub-basin scale, based on the modelled sources of nutrients in the rivers.

2. Methods

2.1. Lake Taihu

Lake Taihu is a large shallow sub-tropical lake located in Jiangsu province (Fig. 1). The lake extends 2350 km^2 and has an average depth of 2 m. The water retention time of the lake is about half a year. In response to regular flooding, the lake has been dammed by a dike surrounding the lake. About 117 rivers and tributaries discharge into the lake (Xu et al., 2015).

In this study, we defined a drainage area of Taihu basin covering 15,723 km^2. We divide the drainage area into five sub-basins (I—V) according to the flow direction of Wu et al. (2011) at 5′ resolution (Fig. 1). Sub-basins I and III are upstream sub-basins draining into downstream sub-basins. Sub-basins II, IV and V are downstream sub-basins draining into Lake Taihu. The upstream and downstream sub-basins cover 49% and 51% of the total basin area, respectively.

Taihu basin is located in an area with rapid urbanization and fast socio-economic development with a population of 10.5 million people in 2012 (Fig. S1 in Supplementary Materials). Around 70% of the population live in urban areas. And more than 85% of the people live in Sub-basins I, II and IV. In urban areas, the people are relatively well connected (more than 70%) to sewage systems, whereas the rural population is less well connected (less than 2%). Increasing food demand has driven intensive agricultural production in the Taihu basin. As a result, agriculture has become an important contributor to N and P inputs to the basin (Figs. S2, S3 in Supplementary Materials).

2.2. Model implementation

Our study was performed in three steps. First, we quantified river export of nutrients to Lake Taihu using the MARINA-Lake model. MARINA-Lake is a version of the MARINA 1.0 model specifically focusing on river export of nutrients to lakes. Second, we used results from the PCLake model to identify critical nutrient loads for Lake Taihu. In the last step, we identified to what extent the river export of nutrients exceeds the critical load (nutrient reduction gap) by comparing the output from the MARINA-Lake model with the critical nutrient loads from the PCLake model, and discussed possibilities to reduce the nutrient exceedance gap.

2.2.1. The MARINA-Lake model for river export of nutrients

We applied the MARINA-Lake (Model to Assess River Inputs of Nutrients to seAs) model to quantify river export of dissolved inorganic (DIN, DIP) and organic (DON, DOP) N and P to Lake Taihu for 2012. The original MARINA 1.0 model was developed for Chinese rivers at the sub-basin scale (Strokal et al., 2016a). This model quantifies annual river export of N and P in different forms: dissolved inorganic and dissolved organic. River export of dissolved inorganic and organic N and P is quantified from diffuse (e.g., use of synthetic fertilizers, manure on land) and point (e.g., direct discharges of animal manure, sewage systems) sources. This model has been recently used to model several lakes in China, resulting in the MARINA-Lake model. The MARINA-Lake model has been used so far to analyze the sources of dissolved inorganic and organic N and P pollution for the past and in the future (up

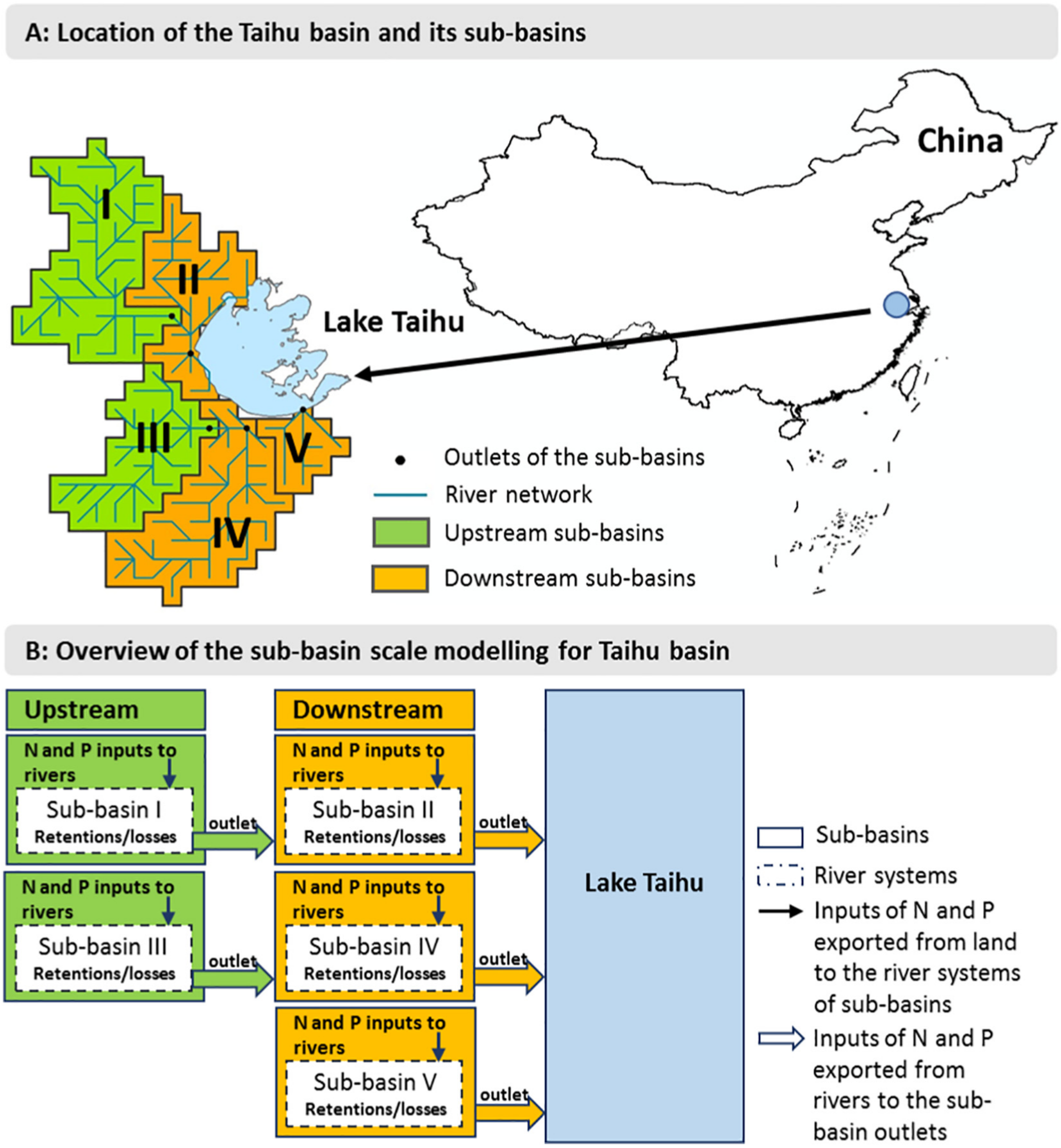

Fig. 1. (A) Sub-basins draining into Lake Taihu, located in China; (B) The overview of the sub-basin scale modelling for Taihu basin. The sub-basins are delineated based on the 5′ resolution river network of Wu et al. (2011). The borders for China is from RESDC (2013).

to 2050) in a spatially explicit way for three lakes: Dianchi (Li et al., 2019), Bayandian and Guanting (Yang et al., 2019).

The MARINA-Lake model quantifies annual river export of N and P by sub-basins and by source to Lake Taihu based on the overall equation (Eq. (1)) as follows:

$$M_{F.y.j} = \left(RSdif_{F.y.j} + RSpnt_{F.y.j}\right) \cdot FE_{riv.F.outlet.j} \cdot FE_{riv.F.mouth.j} \tag{1}$$

where

$M_{F.y.j}$ is river export of forms F (DIN, DON, DIP, DOP) to the river mouth by source y from sub-basin j (kg/year). The model distinguishes N and P inputs to rivers between diffuse sources and point sources.

$RSdif_{F.y.j}$ refers to inputs of nutrient form F to river systems (surface waters) from diffuse source y in sub-basin j. Diffuse sources included in this model are synthetic fertilizers, animal manure, human waste, atmospheric N deposition (for DIN) and biological N_2 fixation (for DIN) over agricultural land, and atmospheric N deposition (for DIN) and biological N_2 fixation (for DIN) over natural land.

$RSpnt_{F.y.j}$ refers to inputs of nutrient form F to river systems (surface waters) from point source y in sub-basin j. Point sources include human waste from the sewage systems, human waste from the urban and rural population that is not connected to sewage systems and direct discharge of animal manure to rivers.

$FE_{riv.F.outlet.j}$ is the fraction of ($RSdif_{F.y.j}$ + $RSpnt_{F.y.j}$) exported to the outlet of sub-basin j accounting for the retention of nutrient form F within the sub-basins.

$FE_{riv.F.mouth.j}$ is the fraction of ($RSdif_{F.y.j}$ + $RSpnt_{F.y.j}$) · $FE_{riv.F.outlet.j}$ exported to the river mouth accounting for the retention of nutrient form F between sub-basin outlets and the river mouth. Both the $FE_{riv.F.outlet.j}$ and $FE_{riv.F.mouth.j}$ are calculated based nutrient retention by denitrification, water consumption, and dams and lakes in the river systems. The detailed equations on how $RSdif_{F.y.j}$, $RSpnt_{F.y.j}$, $FE_{riv.F.outlet.j}$, and $FE_{riv.F.mouth.j}$ are quantified in the MARINA-Lake model is available in the Box S1 in the Supplementary Materials.

We took the MARINA-Lake model (Yang et al., 2019) and updated the model inputs for Lake Taihu (see Fig. S5 in Supplementary

Materials). The model inputs for diffuse sources of N and P inputs on land (e.g., synthetic fertilizers, animal manure, biological N_2 fixation) were derived from the NUFER model (Nutrient flows in Food Chains, Environment and Resources use) (Wang et al., 2018b). The model inputs for point sources of N and P to river systems (e.g., N and P in human excretion, population with sewage connection, N and P removal by sewage treatment) were updated with gross domestic product (GDP) and population densities from Chinese statistic yearbook (RESDC, 2013), information on sewage systems from the dataset for sewage treatment plants from Ministry of Ecology and Environment of the People's Republic of China (MEP, 2013) and the study on Lake Taihu by Liu et al. (2013). The model inputs for hydrology (e.g., river discharge, water consumption in the river systems) were derived from the Community WATer Model (CWATM) (Burek et al., 2017). CWATM is a distributed rainfall-runoff-routing model which includes all necessary hydrological processes for addressing water supply and demand modelling.

2.2.2. The PCLake model for critical nutrient loads

We compared the results from MARINA-Lake for nutrient export by rivers (sub-basins) to Lake Taihu with the critical nutrient loads as calculated by Janssen et al. (2017) (Fig. 3). These critical nutrient loads were calculated with the water quality model PCLake and based on four water quality thresholds for phytoplankton chlorophyll-a starting at strict (<20 μg/L) to more tolerant (<50 μg/L) levels (Table S5). The level of the thresholds depends on the purpose of water use: drinking water has higher water quality requirements than, for example, water for irrigation (Edition, 2011; Fewtrell and Bartram, 2001). Water quality will start to deteriorate when nutrient levels exceed the lower critical nutrient loads. Most of the in-lake chlorophyll-a will be higher than the threshold when nutrient levels exceed the upper critical nutrient load. River export of nutrients as calculated by MARINA-Lake is compared to the dissolved fraction of the critical nutrient loads: 50% of total P, and 90% of total N is in dissolved form. Combining the calculated nutrient losses by source with critical nutrient loads will reveal possible options for nutrient management.

3. Results

We first evaluate the model performance in Section 3.1. Second, we present the results for river export of DIN, DON, DIP and DOP and their sources for the year 2012 in Section 3.2. Next, we compare how our modelled river export of TDN and TDP exceeds the critical nutrient loads and show the opportunities to meet the critical loads in Section 3.3.

3.1. Model performance

We evaluated the results from the MARINA-Lake model for Lake Taihu in three ways.

First, we compared our modelled fluxes of N and P with the measured fluxes of N and P in the lake (Tables 1 and S4). Measured fluxes of N and P to Lake Taihu were available from Xu et al. (2015) and Li et al. (2011a). The comparison shows we model somewhat higher N inputs into the lake compared to the measurements. There are a number of explanations for this. One is the difference in the studied period and location: our model is for 2012 and the measurements are for the period 2000 to 2012. We model the river export of nutrients (external loads into the lake) whereas the measurements might be from water samples at different locations in the lake. Differences in nutrient forms are another reason: we model river export of dissolved inorganic and organic nutrients whereas the measurements are for total N and P thus including particulate forms. Furthermore, our model quantifies annual river export of N and P. However, the measurements are often derived from water samples collected at a specific time during certain periods (e.g., days, seasons) and then averaged to annual values.

Second, we compared our modelled river export of N and P with some other modelling studies (Table 1 and S4). Only few studies subdivide the draining area of Taihu basin into sub-basins (e.g., Yu et al. (2007)) and they delineated sub-basins using a different approach than the one we took. Therefore, we compared our results with other studies at the basin scale rather than at the sub-basin scale. Several studies (e.g., Liu et al., 2013) quantified nutrient loads into Lake Taihu using, for example, the SWAT model (e.g., Lai et al., 2006). The comparison shows that our modelled results are within the range of the results from other studies (Table 1). For example, we model 61 kton of N and 2.0 kton of P exported by rivers in 2012. The other studies estimate 33–66 kton of N and 1.6–5.2 kton of P for different years (1995–2012, see Table 1).

Third, we compared the source attribution from the MARINA-Lake model with some existing studies (Table S4). Results of this comparison are promising and show that our results are generally in line with other studies. For example, the other studies emphasize the importance of agriculture in nutrient pollution of the lake. The results of the MARINA-Lake model are in line with this conclusion. More information on comparing our results with other studies and on model uncertainties can be found in Section 4.1.

Based on this model evaluation, we consider the performance of the MARINA-Lake model good enough for analyses of river export of nutrients, and their sources, to Lake Taihu. As mentioned above, the MARINA-Lake model has been successfully implemented to a few other lakes in China: Dianchi (Li et al., 2019), Bayandian and Guanting (Yang et al., 2019). This gives trust in implementing the MARINA-Lake model for Lake Taihu.

3.2. River export of nutrients and their sources

River export of nutrients differed among sub-basins in 2012. Rivers exported 61 kton of TDN and 2 kton of TDP to Lake Taihu in 2012 (Fig. 2). Most of these nutrients were in dissolved inorganic forms.

Table 1
Comparison of our modelled annual river export of total N and P to Lake Taihu with other studies. Our study is in the grey shaded row.

Studies	TN		TP		Temporal resolution	Type of study
	g/m^2/year	kton/year	g/m^2/year	kton/year		
Xu et al. (2015)	20 (17–23)	46 (41–54)	0.9 (0.8–1.1)	2.1 (1.8–2.6)	2007–2012	Measurements
Li et al. (2011a)	9.3–12*	21–27	0.4–0.5	0.8–1.2	2000–2005	Measurements
Liu et al. (2013)	14.7*	33	2.3	5.23	2008	Modelled results
Huang et al. (2018)	–	–	0.8	1.9	2014–2016	Modelled results, polders
Reidsma et al. (2012)	29.3*	66	0.7	1.6	2008	Modelled results
Lai et al. (2006)	17.8* (16.4–18.7)*	40 (37–42)	0.9 (0.8–0.9)	2.0 (1.8–2.0)	1995, 1998, 2002	Modelled results
This study**	27	61	0.9	2.0	2012	Modelled results

*Estimated from kton/year using the surface area of the lake. **In this study we model total dissolved N and P, which is the sum of dissolved inorganic and organic N and P.

More than half of the nutrients resulted from activities in Sub-basins I and IV that cover around 60% of the Taihu basin drainage area. Sub-basin II contributed by 15–22% to the nutrients transported to Lake Taihu depending on the nutrient form. The share of Sub-basin III in the total nutrient loads to the lake was estimated at 8–16% for different nutrient forms. Sub-basin V contributed <10% to the nutrients in 2012 (Fig. 2). Rivers of Sub-basin III exported the smallest amount of these nutrients per km^2 whereas rivers of Sub-basin IV exported the largest amounts of the nutrients per km^2 in 2012 (Fig. S6 in Supplementary Materials).

Diffuse sources were responsible for about 90% of the total TDN river export to Lake Taihu in 2012 (Figs. 2 and 3). Around 40% of this TDN river export from diffuse sources was from synthetic fertilizers. However, the shares of diffuse sources varied greatly among sub-basins and N forms (Fig. 2). For example, the share of diffuse sources ranged from 83 to 98% for DIN and from 43 to 89% for DON among the sub-basins (Fig. 2). Use of synthetic fertilizers on land was an important diffuse source of DIN in rivers of all sub-basins. Atmospheric N deposition also contributed to DIN in rivers of the sub-basins (16–30% in the total DIN export). This was different for DON. Leaching of organic matter was an important source of DON in rivers (with a share of 36–74% for different sub-basins and N forms). In addition, sewage systems (point source) were responsible for half of DON in rivers of Sub-basin II and for 7–38% of DON in rivers of the other sub-basins.

Point sources were responsible for 48% of the total TDP river export to Lake Taihu in 2012 (Figs. 2 and 3). Most of this TDP from point sources were from sewage systems. This varied among sub-basins and P forms (Fig. 2). The share of sewage systems in DIP river export ranged from

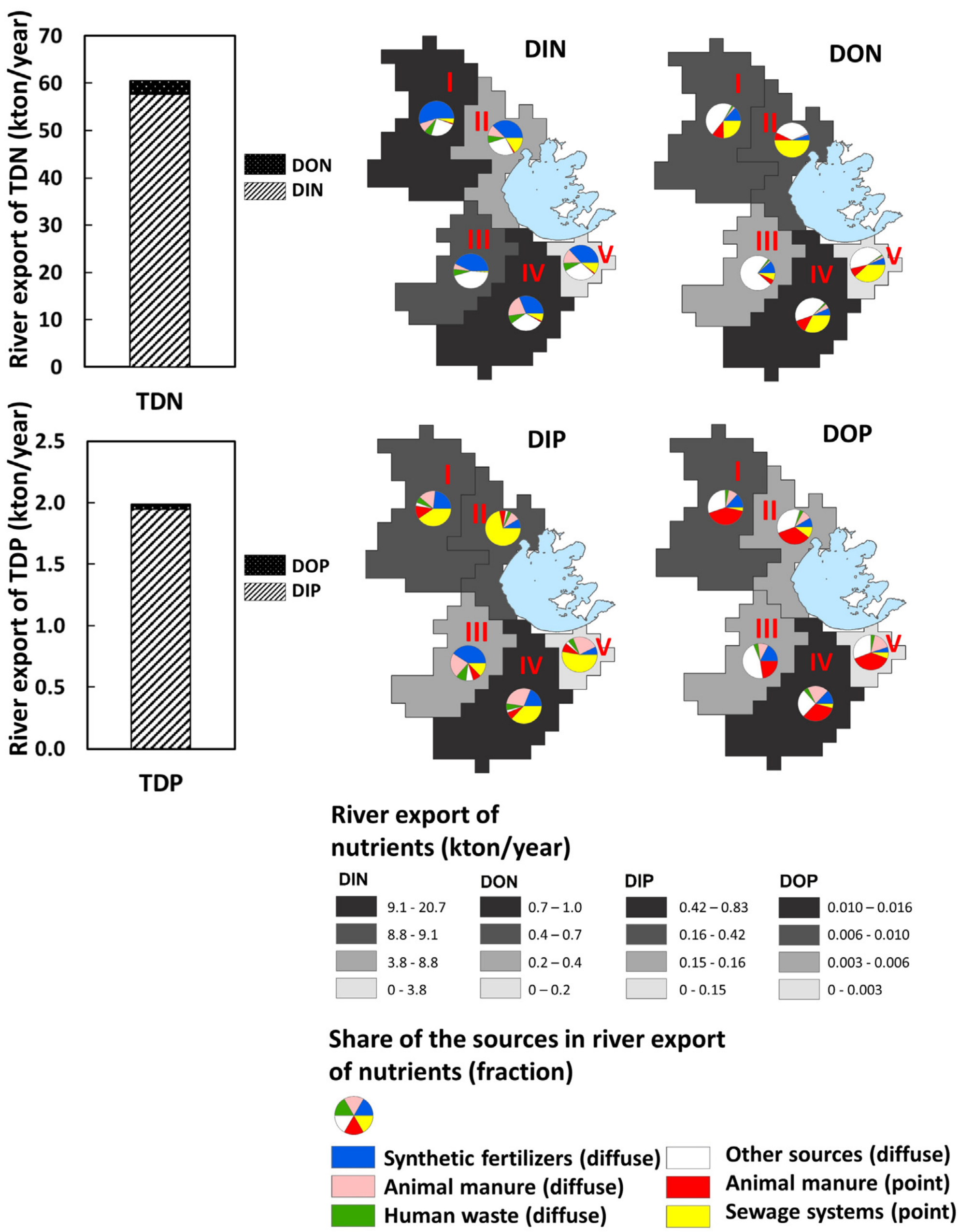

Fig. 2. River export of nitrogen and phosphorus by source from sub-basins to Lake Taihu in 2012. TDN and TDP are total dissolved nitrogen and phosphorus, respectively. DIN and DIP are dissolved inorganic nitrogen and phosphorus, respectively. DON and DOP are dissolved organic nitrogen and phosphorus, respectively. "Others" include atmospheric N deposition over agricultural and non-agricultural areas (for DIN), biological N_2 fixation by natural vegetation and crops (e.g., legumes) (for DIN), leaching of organic matter (for DON and DOP) and weathering of P-contained minerals (for DIP) from agricultural and non-agricultural areas. The results are from the MARINA-Lake model (see Section 2.2).

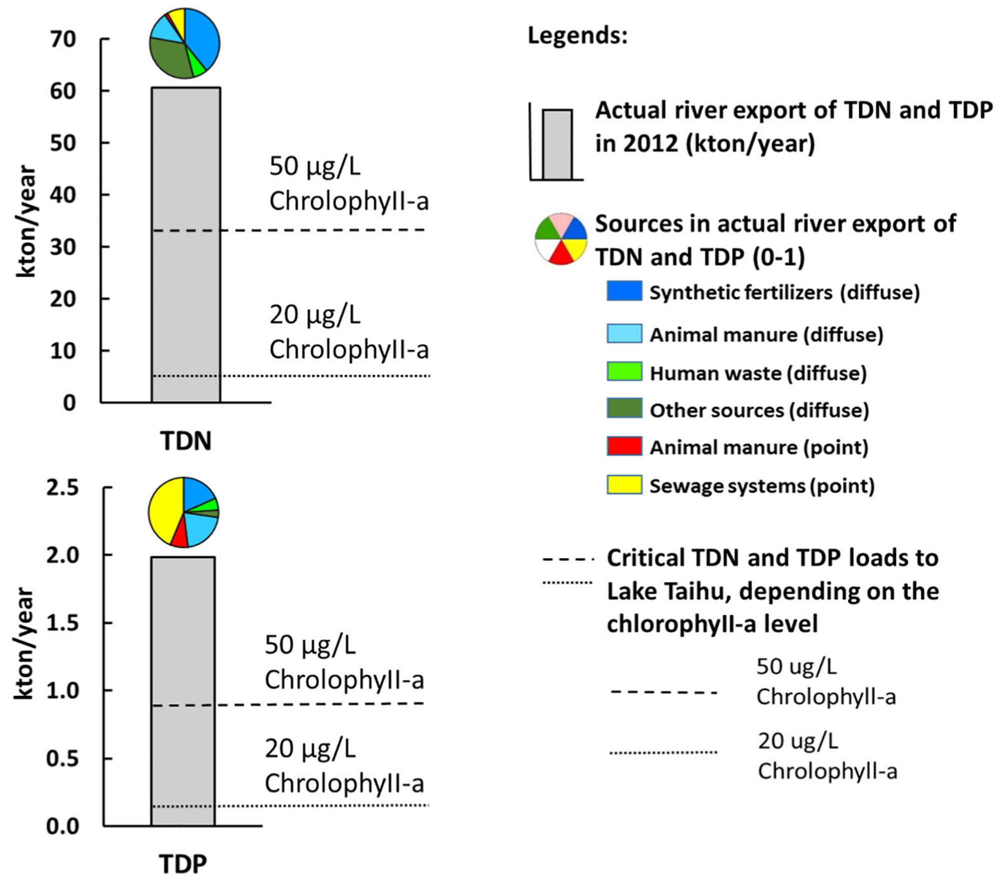

Fig. 3. Actual river export of total dissolved nitrogen (TDN) and phosphorus (TDP) in 2012 (kton/year, shown by the bars), the share of the sources (0–1, shown by the pies) and the critical TDN and TDP loads (kton/year, shown by the dash lines) according to the two levels of Chlorophyll-a (20 and 50 µg/L). Actual river export of TDN and TDP is from the MARINA-Lake model (see Sections 2.2, 3.2). TDN and TDP are the sum of dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen and phosphorus. "Others" include atmospheric N deposition over agricultural and non-agricultural areas (for DIN), biological N_2 fixation by natural vegetation and crops (e.g., legumes) (for DIN), leaching of organic matter (for DON and DOP) and weathering of P-contained minerals (for DIP) from agricultural and non-agricultural areas. The critical TDN and TDP loads according to the levels of Chlorophyll-a are from the PCLake model (see Section 2.2.2) (Janssen et al., 2017). The levels of chlorophyll-a of 20 and 50 µg/L were used to determine the levels of the critical loads of TDN and TDP in Lake Taihu (see Table S5 for details).

11 to 60% among sub-basins. In rivers of Sub-basins II and V, more than 40% of DIP resulted from sewage systems, indicating rapid urbanization in these sub-basins. In addition, diffuse sources namely use of synthetic fertilizers (for Sub-basins I, III and IV) and animal manure on land (for Sub-basins I, III, IV and V) were important sources of DIP in rivers. For river export of DOP, direct manure discharges were more important point sources of DOP in rivers than sewage systems. The share of manure discharges in DOP river export ranged from 22% (Sub-basin III) to 42% (Sub-basin V) among sub-basins (Fig. 2). Leaching of organic matter, use of animal manure and synthetic fertilizers also contributed to DOP in rivers.

3.3. Opportunities to meet critical nutrient loads

In this section, we identify opportunities to meet critical nutrient loads for Lake Taihu. To this end, we compare the actual river export of nutrients with the critical loads to Lake Taihu to identify required reductions.

Critical nutrient loads to Lake Taihu depend on the desired level of Chlorophyll-a in the lake (Section 2.2.2 and Table S5). We present results for the two extreme levels of Chlorophyll-a: 20 µg/L (most strict) and 50 µg/L (least strict). For 20 µg/L of Chlorophyll-a, the most strict critical load is 5 kton/year for N and 0.15 kton/year for P. For 50 µg/L of Chlorophyll-a, the least strict critical load is 33 kton/year for N and 0.9 kton/year for P (Table S5). In our analysis, we compare the modelled actual river export of N and P in kton/year with these critical loads (Fig. 3). Water quality that meets the critical nutrient loads of 20 µg Chl-a/L can be considered as very good and appropriate for, for example, drinking purposes (Janssen et al., 2017). Water quality that meets the critical nutrient loads of 50 µg Chl-a/L can be considered as just acceptable, and is more appropriate for purposes such as irrigation (Edition, 2011; Fewtrell and Bartram, 2001).

Results show that the actual river export of nutrients exceeded largely the critical loads in 2012 (Fig. 3). To meet critical loads of TDN, river export needs to be reduced by 46–92%, depending on the level of Chlorophyll-a. This implies that river export of TDN needs to be reduced by around 28 kton to meet the least strict critical N load (50 µg Chl-a/L). To meet the most strict critical N load (aiming for 20 µg Chl-a/L), the required reduction is around 56 kton for TDN. Most TDN in river export was DIN (see Fig. 2 and Section 3.2). Thus, the required reductions apply to DIN. Most DIN in rivers draining in Taihu were from diffuse sources with the large share of synthetic fertilizers (see Figs. 2 and 3, Section 3.2). Thus, improving N use efficiencies in agriculture (e.g., use of fertilizers according to crop needs) may effectively reduce DIN in rivers to meet the least strict critical N load. However, to meet the most strict critical N load (aiming for 20 µg Chl-a/L), additional reductions in DIN in rivers from other sources are required.

To meet critical loads of TDP, river export needs to be reduced by 55–92%, depending on the level of Chlorophyll-a (Fig. 3). We estimate a required reduction of 1.1 kton for TDP river export from all sub-basins to meet the least strict critical P load (aiming for 50 µg Chl-a/L). A reduction of 1.8 kton in TDP is needed to meet the most strict critical

P load (20 μg Chl-a/L). Most TDP river export is DIP (see Section 3.2, Fig. 2). Thus, reducing DIP in rivers can largely contribute to meet the critical P loads in Lake Taihu. Our result shows that 11–60% of the DIP in river export from sub-basins was from point sources (sewage systems). Thus improving removal efficiencies for nutrients during wastewater treatment in sewage systems is an effective way to meet the least strict critical P load. However, meeting the most strict critical P load will be difficult without considering other sources (e.g., use of synthetic fertilizers and animal manure on land, Figs. 2 and 3). Combining better nutrient management in agriculture with improved sewage treatments is needed to meet the critical loads.

4. Discussion

4.1. Sources of nutrient export to Lake Taihu

We linked the MARINA-Lake model with the PCLake model for Lake Taihu to better understand the sources of river export of nutrient to Lake Taihu in respect to the critical nutrient loads of the lake. In general, our model results compare well with the existing studies (see also Section 3.1). This also holds for the source attribution. For example, we estimate that around 15% of N and 49% of P river export from all sub-basins resulted from households (sewage systems and human waste as well as fertilizers, see Table S4). This is in the range of other studies indicating that the share of households is 17–43% for N and 28–50% for P (Lai et al., 2006; Wang et al., 2006). We estimate that 1–8% of N and P in Lake Taihu is from direct discharge of animal manure to rivers. Exiting studies estimate 5–20% (Liu et al., 2013; Wang et al., 2006). Our estimates for the share of livestock activities (use of manure on land and direct discharges) are in line with the estimates of Liu et al. (2013) (see Table S4).

There are also some differences between our study and other studies (Table S4). For example, our study estimates a larger share of diffuse sources for N than existing studies (e.g., Cao et al. (2013); Lai et al. (2006); Wang et al. (2006)). The difference can be explained by the fact that we account for more sources of N in rivers (e.g., biological N_2 fixation) from agricultural and non-agricultural areas. Another reason is that there is a difference in the study areas and the level of temporal detail between our study and existing studies (Table 1). This may explain the differences between our estimates and other studies for sources of nutrients in rivers of Taihu basin. Furthermore, we calculated slightly higher contributions from sub-basins III, IV and V in the south than from sub-basins I and II in the north, which was also found by Yu et al. (2007). Our result shows sub-basins III, IV and V contributed in total 56–61% to the nutrients transported to Lake Taihu depending on the nutrient form. This spatial pattern can be conceived counterintuitive as most cyanobacterial growth was observed in the north (Duan et al., 2009; Li et al., 2018). However, due to hydrological patterns that limit the exchange of water between the bays and the rest of the lake, the nutrient load in the northern bays of the lake is spread over a relatively small area. The nutrients from the northern sub-basins are mixed over a large part of Lake Taihu (Li et al., 2011b). When nutrient loads are spread over a relatively small area, the result is locally higher nutrient concentrations that boost algal growth. Besides the reason of the nutrient loads, the effect of wind is another important reason to explain higher algal biomass in the north. The general wind direction at Lake Taihu is towards the northwest. In the center of Lake Taihu, the wind is generally too strong to sustain high algal growth. Besides, algae that grow in the center of the lake are blown to the northern parts of the lakes.

With respect to the uncertainties in critical nutrient loads, Janssen et al. (2017) made a comparison with Xu et al. (2015). Xu et al. (2015) estimated a required reduction of 20–71% for nutrients in the lake to meet the critical nutrient threshold (condition of $<20\ \mu g\ L^{-1}$ chlorophyll *a*). This is lower than our study (46–92%, see Fig. 3). The reason for this can be that Xu et al. (2015) calculated the reduction for TN and TP, while in this study we focus on TDN and TDP. In addition, Xu et al. (2015) used experiments to determine the critical nutrient thresholds whereas the critical nutrient loads in this study are obtained using the model PCLake (Section 2).

4.2. Combing MARINA-Lake and PCLake: strengths and limitations

We combined the sub-basin scale MARINA model and the lake ecosystem model PCLake. This approach allows us to identify opportunities to reduce the gap between the actual and critical nutrient loads. To our knowledge, such a comparison was not done before for Lake Taihu. This comparison is very useful for Lake Taihu since many existing studies have shown that eutrophication problems are increasing in this lake in the last decades (Duan et al., 2009; Janssen et al., 2014; Qin et al., 2010).

There are also limitations in combing these two models. For example, to compare the critical nutrient loads with the results from MARINA, we converted critical loads for total N and P to dissolved N and P (Section 2). This means that the gap between the actual loading and the critical nutrient loads in our study needs to be interpreted in the light of dissolved forms. Since dissolved forms of N and P generally are considered to have a higher bioavailability for harmful algae than their particulate forms (Garnier et al., 2010), we consider this as an acceptable approach to combine the results of two models.

Our model has uncertainties associated with model inputs, parameters and nutrient sources. For example, we derived many model inputs for agricultural activities (e.g., use of synthetic fertilizers, manure excretion) from the NUFER model (Wang et al., 2018b). This model provides inputs for over three thousand counties in China. We aggregated county-scale inputs to sub-basin-scale inputs using an area-weighted method in ArcGIS (see Table S3 for more explanation). Model inputs for water discharge at the sub-basin outlets were derived from the CWATM model at 5 min resolution. This model was not calibrated for rivers draining to Lake Taihu. We realize that an un-calibrated hydrological model may introduce uncertainties in our estimates. For example, a higher discharge will lead to more nutrients exported to Lake Taihu which increases algal blooming and requires stronger nutrient reductions (Paerl and Huisman, 2009). However, to calibrate the model an extensive number of measurements at the outlets of the Taihu sub-basins is needed which are not at our disposal. Some of the MARINA-Lake model parameters were based on literature and in this study do not vary among sub-basins (e.g., the fraction of manure discharges to rivers (Section 2)). Regarding the nutrient sources, the MARINA-Lake model ignores sources such as P deposition from atmosphere and aquaculture. To our knowledge, these sources do not contribute largely to the total nutrient pollution in Lake Taihu (e.g., Cai et al. (2013); Liu et al. (2013); Wang et al. (2006)) compared to agricultural activities and sewage systems.

The PCLake model is well known in the context of critical nutrient loads (Janse et al., 2008; Janssen et al., 2014; Kong et al., 2017; Li et al., 2019). PCLake quantifies critical nutrient loads based on the average temporal and spatial conditions in the lake. In reality, conditions of lakes fluctuate among years, such as wetter or dryer years and warmer or colder years. These fluctuations cause uncertainties in the critical nutrient loads when applied for specific years. Moreover, Janssen et al. (2017) showed an application of the PCLake model in which spatial differences in critical nutrient loads exists: some parts of the lake have a lower critical nutrient load and are thus more sensitive to a shift than other parts of the lake. Our results should be seen, therefore, as an indication of the temporally and spatially averaged critical nutrient load of Lake Taihu.

We believe that the model uncertainties do not change the main messages of our study. We validated the MARINA-Lake model with measurements for water quality that are available to us. Validation results are promising (Section 3.1). The critical nutrient loads found with the PCLake model has been validated by Janssen et al. (2017). In addition, we compared our results with results of other existing studies for Lake Taihu (see below and Tables 1 and S4). This gives trust in using the MARINA-Lake model to analyze the sources of N and P in rivers to Lake Taihu. Furthermore, the MARINA-Lake model has been successfully applied to other lakes in China: Lake Dianchi (Li et al., 2019) and Guanting reservoir and Lake Baiyangdian (Yang et al., 2019). In the case of Lake Dianchi the combination of MARINA-Lake model with the PCLake model was also applied.

4.3. Implications for environmental policies

Our study can help to search for effective environmental policies for lake restoration. We show the gap between the actual river export of N and P and the critical nutrient loads (Section 3.3). Reducing this gap will reduce eutrophication problems and facilitate lake restoration. This gap depends on the requirements of water quality for different purposes of water use (e.g. drinking water or irrigation). In our study, we use the two extreme critical nutrient loads depending on the desired level of Chlorophyll-a: least strict (e.g., water for irrigation) and most strict (e.g., water for drinking) critical nutrient loads.

The need for effective environmental policies to restore Lake Taihu is recognized in existing studies (e.g., Liu et al. (2013); Ma et al. (2014); Reidsma et al. (2012)). For example, policies for sustainable agricultural developments are needed to avoid nutrient pollution of Lake Taihu (e.g., Ma et al. (2014); Reidsma et al. (2012)). Our study provides useful information on from which human activities (sources) and areas (sub-basins) nutrient export by rivers should be reduced to meet the critical nutrient loads in Lake Taihu. We argue that reducing N inputs in rivers from synthetic fertilizers (diffuse source) and P inputs in rivers by improving treatment of wastewater (point source) are essential to meet the critical nutrient load under which water can be used for irrigation purposes. However, more efforts might be needed to meet the critical nutrient loads under which water can be used for drinking. This can be done by 1) fertilizing the crops according to their needs for nutrients, 2) recycling animal manure on land to avoid manure discharges to water, and 3) improving wastewater treatment in sewage systems of the Taihu basin (Strokal et al., 2017; Wang et al., 2018a; Zhang et al., 2012). Some of the current policies already aim to reduce nutrient loads in Lake Taihu. For example, implementing the 'Zero Fertilizer Policy' that aims for zero growth in the use of synthetic fertilizers after 2020 will reduce the fertilizer use in the Taihu basin (MOA, 2015). The national government has also introduced management strategies and technologies for improving sewage treatment of nutrients in rural area (Wang et al., 2010). These measures will help to reduce nutrient inputs to the lake from rural sewage. Changes in climate are also important for future water quality in the lake because climate-related factors (e.g. temperature, wind speed) also have effects on changes in critical nutrient loads in Lake Taihu, and in nutrient retention in the Taihu basin (Zhang et al., 2018). Thus future policies should combine management options to reduce river export of N and P from both diffuse (e.g., use of synthetic fertilizers, manure) and point (e.g., sewage systems) sources, with consideration of future changes in climate. In any case, it is important to realize that a full restoration of Lake Taihu will take time, even with strong nutrient reductions, as it will take years before the large pool of nutrients stored in the sediments will be released (Wu et al., 2019).

5. Conclusions

Our results show that the sources of nutrient inputs to Lake Taihu differed largely among nutrient forms and sub-basins in 2012. Rivers exported 61 kton of TDN and 2 kton of TDP to Lake Taihu, mostly in dissolved inorganic forms. More than half of the nutrients in Lake Taihu were from Sub-basins II (north) and IV (south). Diffuse sources contributed 90% to the TDN in rivers. More than 40% of this TDN from diffuse sources was from synthetic fertilizers used on land. The share of diffuse sources in river export of N ranged from 43 to 98% among the sub-basins and N forms. Atmospheric N deposition was another important source of DIN and sewage systems of DON in rivers. Point sources contributed 52% to TDP in the rivers. Important point sources were sewage systems. The share of sewage systems in river export of DIP ranged from 11 to 60% among sub-basins. Direct manure discharges were important sources for river export of DOP.

To meet the critical nutrient loads, river export of TDN and TDP needs to be reduced by 46–92%. This implies a reduction of 28–56 kton TDN and 1.1–1.8 kton TDP, depending on the level of Chlorophyll-a. In our analysis, we focus on the critical nutrient loads based on two extreme levels of Chlorophyll-a in the lake: 20 μg/L (most strict, e.g., for drinking purposes) and 50 μg/L (least strict, e.g., for irrigation purposes). The least strict nutrient loads may be reached by reducing river export of DIN from diffuse sources (synthetic fertilizers) and DIP from point sources (sewage systems). To meet the most strict nutrient loads, it requires combinations of reduction options for both diffuse and point sources. Examples are improved nutrient use efficiency in agriculture by better meeting the needs of nutrients by crops, combined with best available technologies to remove nutrients in sewage systems.

Our study shows the main sources of nutrients in rivers draining into Lake Taihu. This can help to identify opportunities to meet critical nutrient loads. A good understanding of nutrient sources is essential for formulating effective management options to reduce nutrient pollution. Our study may thus contribute to restoring the ecosystems in the lake, and serve as an example for other lakes.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.02.051.

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