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Research article

Sources of nitrogen in reservoirs of the Haihe basin (China) 2012–2017

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ABSTRACT

Nitrogen (N) is essential for agricultural production. However, too much N can pollute waters. The Chinese government published several policies to reduce N losses from agricultural production to waters since 2015, which may influence river export of N to reservoirs and lakes and their pollution sources. This study aimed to quantify the trends of river export of N to five reservoirs in the Haihe basin and analyze the main sources of this N pollution from 2012 to 2017. This was done by upscaling the MARINA-Lakes (Model to Assess River Inputs of Nutrients to LAkes) model to the Haihe basin, including 22 sub-basins. From 2012 to 2017, river export of total dissolved nitrogen (TDN) to the Haihe reservoirs decreased by 11–51%, associated with a decreased contribution of point sources and an increased contribution of diffuse sources for the whole study area. Sub-basins draining into Reservoir Pan-Da contributed over one-third to the total TDN export by rivers in 2012 and 2017. The share of diffuse sources in river export of TDN to the Guanting reservoir reached 63% in 2017. Among the TDN diffuse sources, the contribution of animal manure (a diffuse source) to river export of diffuse TDN increased to 28%, 25%, and 23% for the sub-basins of Reservoir Miyun, Pan-da, and Guanting from 2012 to 2017, respectively. Among the TDN point sources, direct manure discharges were the main contributors to the river export of point TDN to the Haihe reservoirs in 2012. By 2017, direct discharges of untreated human waste became another important point source, especially for the Lake Baiyangdian and Reservoir Gang-Huang. This study concludes the need for specific agricultural N management options for different reservoirs of the Haihe basin.

1. Introduction

Nitrogen (N) plays various roles across social and natural systems. N is a necessary resource for society, sustaining our food demand through agricultural production (Erisman et al., 2008). However, in nature, excessive N can cause problems. For example, N that exceeds the environmental capacity can lead to eutrophication in water systems (Basu et al., 2022; Schulte-Uebbing et al., 2022). Several studies indicate that crop and animal production are the main sources of N pollution in water systems (Chen et al. 2020, 2022; Stokal et al., 2016b). This illustrates the potential conflicting aims between Global Sustainable Development Goals (SDGs) 2 “Zero Hunger” and SDG 6 “Clean Water and Sanitation” (UN, 2015). Several studies analyzed the relationships between different SDGs (Wu et al., 2022). For example, Wang et al. (2022) found strong trade-offs between food production (SDG 2.3) and water quality (SDG 6.3) in China. Therefore, it is important to study ways to maintain agricultural production while ensuring low water pollution.

Since 2015, the Chinese government published several policies and plans to guide sustainable development with less water pollution. Examples of these policies the Zero Fertilizer Growth (MOA, 2015), Increasing recycled manure (Zhang et al., 2022), Restricted Livestock production (MEP and MOA 2016), Improvement of sewage systems, and Prevention of Water Pollution (SCPRC, 2015). Particularly, agricultural production and water quality have gained considerable attention because of excessive fertilizer use and direct animal manure discharges to water systems (Stokal et al., 2016b). In addition, Agriculture Green Development (AGD, i.e., high productivity, high resource use efficiency, and low environmental impact) is recommended for implementation in China (Shen et al., 2020). Consequently, measures and technologies were applied across Chinese basins. However, it needs to be clarified how these top-down actions affect N losses from agricultural production to water systems in different basins of China. Moreover, there is also a need to understand the underlying factors which drive N losses to water systems. Analyses of driving factors may help to explore basin-specific N

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management options to further reduce N pollution in water systems.

Reservoirs and lakes provide drinking water and other ecosystem services such as flood protection, recreation, and biodiversity conservation (Lin and He, 2003). China has many reservoirs. Some in the Haihe basin suffer from water scarcity (Xia, 2002, 2004). The Haihe Basin is among China's largest river basins. It is an economically developed area that covers 320,000 km² and includes mountainous (60%) and plain areas (40%) (Sun et al., 2017) (Fig. 1). At the same time, the Haihe basin covers most of the North China Plain (Zhao et al., 2015), which is an essential agricultural production base. Accordingly, to meet water demands from the domestic, agricultural, and industrial sectors, more than 1000 reservoirs have been constructed since the 1950s in mountainous areas such as Reservoir Miyun and Guanting, which are sources of drinking water for Beijing (Ding et al., 2016; Jiang et al., 2011). Also, several natural lakes are located in the plain areas of the Haihe Basin (e.g., Lake Baiyangdian). On the other hand, water systems are largely polluted as described in Figure S3 in the Haihe basin. For example, the total N concentration in the rivers was quadruple the Chinese water quality standard V (i.e., 2 mg L⁻¹), and 85% of the rivers were mesotrophic and eutrophic in 2011 (Rong et al., 2016; Zhang et al., 2015). Polluted rivers transport excessive nutrients downstream, posing a threat to reservoirs and lakes (Tang et al., 2019). Considering the importance of agriculture production and the urgency of water pollution in the Haihe basin, we chose reservoirs and lakes in the Haihe basin as examples to show the changes in nutrient losses resulting from agriculture production and associated factors.

This study aims to quantify the trends of river export of N to five reservoirs (four artificial reservoirs and one natural lake) in the Haihe basin and analyze the main sources of this N pollution from 2012 to 2017. To this end, we upscaled the MARINA-Lakes (Model to Assess

River Inputs of Nutrients to Lakes) model to quantify N inputs to 22 draining sub-basins and river export of Total Dissolved Nitrogen (TDN, the sum of dissolved inorganic nitrogen (DIN) and dissolved organic nitrogen (DON)) to the five reservoirs in 2012 and 2017. Then we analyzed several social, agricultural, and hydrological factors affecting the river export of TDN. Finally, we discussed options for N management for the specific reservoir in the Haihe basin.

2. Methodology

2.1. Study area

We chose five reservoirs, including Lake Baiyangdian, and four reservoirs: Gang-Huang, Guanting, Miyun, and Pan-da. These four reservoirs are in the mountainous areas, and Lake Baiyangdian is in the plain. Twenty-two sub-basins drain into the five reservoirs (Fig. 1a), delineated using the information from an online database and our earlier study (Lehner and Grill, 2013; Yang et al., 2019). According to the Chinese surface water standard, the required water quality varies among the study areas (Fig. 1b). Grade II is required for Reservoir Gang-Huang, Miyun, and Pan-da because they provide drinking water for several megacities in the Haihe basin. Grade III is required for Lake Baiyangdian and Reservoir Guanting. From 2012 to 2017, the urbanization ratio increased in all study areas (Fig. 1b). Reservoir Miyun and Lake Baiyangdian experienced decreases in the crop-sown areas and animal-raising numbers from 2012 to 2017. Reservoir Pan-Da held a steady crop area but a noticeable increase in livestock numbers (51%). The agricultural activities turned more intensive in Reservoir Gang-Huang between 2012 and 2017 (20% and 34%). The changes for decreasing crop areas and increasing animal numbers were below 10%

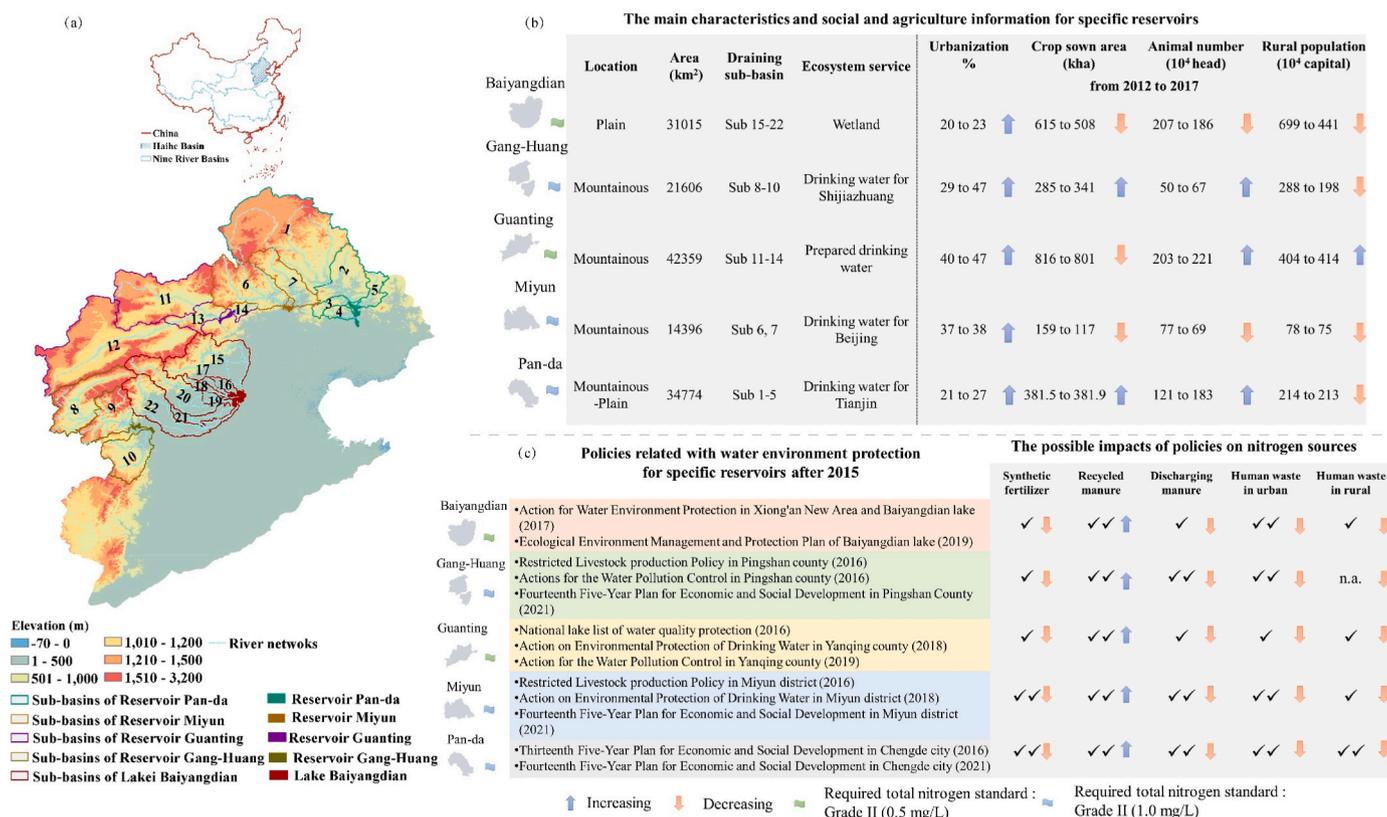


Fig. 1. (a) The locations of the five reservoirs in the Haihe basin. (b) The main characteristics of the five reservoirs, including social and agricultural information for 2012 and 2017. (c) The policies related to water protection for five reservoirs. The number of marks refers to the number of related policies. The blue and orange arrows refer to the policies that could increase or decrease the amounts of nitrogen sources, respectively. Source (BJMPG, 2018; CDMPG, 2016, 2022; HBMPG, 2018, 2019; Lehner and Grill, 2013; MEP, 2002; MYMPG, 2016, 2021; PSMGP, 2016b, 2021; Yang et al., 2019; YQMPG, 2019, 2021; ZJKMEPB, 2017). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

in Reservoir Guanting (Fig. 1b). After 2015, policies related to water quality protection were published for the specific reservoir (Fig. 1c). Most of these policies are comprehensive, increasing or decreasing N losses from different N sources to five reservoirs (Fig. 1c, Table S1). Reducing synthetic fertilizer application and improving sewage facilities in urban were more emphasized in the policy document compared to the past years. The management of animal excretion gradually attracted attention. However, rural sewage systems were less concerned than other sources from 2012 to 2017 (Fig. 1c, Table S1).

2.2. The MARINA-lakes model

MARINA-Lakes model was used to estimate N inputs and river export of TDN to five reservoirs in the Haihe basin for 2012 and 2017. This model has been applied to several lakes, including Lake Taihu, Lake Dianchi, Reservoir Guanting, Lake Baiyangdian in China, and Tana Lake in Ethiopia (Goshu et al., 2020; Li et al., 2019; Wang et al., 2019; Yang et al. 2019, 2021).

MARINA-Lakes model accounts for anthropogenic activities (e.g., agricultural production and human waste), natural processes (e.g., weathering) and hydrological factors (e.g., runoff and river discharges) (Yang et al., 2021). The details of the model structure and equations are described by Yang et al. (2019). This model does not account for the relation between phosphorus concentrations in soil and biological N₂ fixation. The calculation flow is shown in Section S2 in the supplementary materials. All input data sources for this study are derived from statistical books and previous studies (Wang et al., 2018; Yang et al., 2019), see details in Tables S2-S4 of the supplementary materials. To build trust in our model results, we did the model validation (Section S3 in the supplementary materials) and discussed model uncertainty (Section 4.3).

2.3. Definition of nitrogen hotspot and non-hotspot areas

We define “N hotspot” based on the N inputs to sub-basins and river export of TDN to five reservoirs, and by using the approach of Wang et al. (2018) and Li et al. (2021). The year 2012 was chosen as a base year. Firstly, we calculate the N input (for sub-basins, land, and rivers, per km² of sub-basin area) and the river export of TDN (for total river export of TDN, river export of diffuse TDN, river export of point TDN, per km² of sub-basin area) in 2012. Secondly, we rank these values from the highest to the lowest: from Rank IV (highest) to Rank I (lowest). Thirdly, sub-basins with the top 25% of the highest N inputs and river exports of TDN in 2012 were defined as hotspot sub-basins (Rank IV). The non-hotspot sub-basins (Rank I-III) have lower values. All ranges are described in Table S5, Figs. 2 and 3.

2.4. Driving factors affecting river export of N

Several agricultural, socio-economic, and hydrological factors were compared between hotspots (rank IV in Fig. 3) and non-hotspot sub-basins (ranks I, II, III in Fig. 3) in 2012 and 2017. The agricultural factors are (i) the use of N synthetic fertilizers for crops, (ii) animal numbers, (iii) N recycled animal manure to crops, (iv) N from discharging animal manure to rivers, and (v) N recycled ratio (calculated by N recycled animal manure to crops and total animal manure excretion excluded gas N). The socioeconomic factors are (i) urbanization ratio (the proportion of the urban population to the total population), (ii) rural population, and (iii) Gross Domestic Production (GDP). The hydrological factors are (i) nature water discharges and (ii) river discharges in the sub-basins. The results of the comparisons are shown in Fig. 4 and Table S7.

3. Results

3.1. Spatial and temporal variation of N inputs to sub-basins

Inputs of N to the sub-basins include N inputs to land from diffuse sources and N inputs to rivers from point sources (see detailed description in section 2.2). N inputs to 22 sub-basins decreased from 1237 Gg year⁻¹ in 2012 to 1114 Gg year⁻¹ in 2017 (Fig. 2a). N inputs to land contributed 77–86% of all N inputs to the sub-basins in 2012, and these percentages increased to over 88% in 2017 (Fig. 2a). Sub-basins of Lake Baiyangdian were responsible for over 50% of N inputs to 22 studied sub-basins in 2012 and 2017. The sub-basins of Reservoir Miyun received the lowest N inputs compared to the others in both years. However, N inputs to the sub-basins of Reservoir Pan-da increased from 2012 to 2017 (Fig. 2a). From 2012 to 2017, the hotspot area for N inputs to all studied sub-basins remained relatively the same, but the hotspot area for N inputs to rivers decreased from 2012 to 2017. Moreover, most hotspots were mainly concentrated in the sub-basins of Lake Baiyangdian in both years (Fig. 2b, Table S7).

The sources of N inputs to land and rivers differ over the years and sub-basins. In 2012, more than 49% of the N inputs to land were from synthetic fertilizers, and less than 20% were from animal manure applied to crops (Fig. 2c). By 2017, synthetic fertilizers' contribution decreased, but animal manure's contribution increased. The most obvious changes happened in the sub-basins of Reservoir Miyun. The shares of synthetic N fertilizers changed from 50% to 35%, and the shares of animal N manure were from 20% to 35% from 2012 to 2017, respectively (Fig. 2c). However, the reductions of synthetic N fertilizers were not apparent in the sub-basins of Lake Baiyangdian and Reservoir Gang-Huang, although animal manure N applied to crops increased (Fig. 2c). For N inputs to rivers, direct discharges of manure to rivers dominated across all basins in 2012 (60–84%). The situation changed in 2017 for several reservoirs. For example, treated and untreated human waste contributed over 60% to N inputs from point sources to rivers in the Reservoir Gang-Huang and Guanting sub-basins in 2017 (Fig. 2c).

3.2. Spatial and temporal variation of TDN exports by rivers to reservoirs

Our analyses show that the decrease in river export of TDN to reservoirs is associated with the decreased share of point sources (i.e., direct discharges of animal manure) and increased share of diffuse sources (i.e., animal manure used for crops) at the sub-basin scale (Fig. 3a, b and 3c). From 2012 to 2017, river export of TDN is estimated to decrease by 11–51% (23.1–14.4 Gg year⁻¹, Fig. 3a) for five reservoirs. Point sources contributed by 75–95% to the river export of TDN to five reservoirs in 2012. Diffuse sources became the main contributor of TDN, reaching 63% for the Guanting reservoir in 2017 (Fig. 3a). Reservoir Guanting received the lowest river export of TDN in both 2012–2017 years compared to the other reservoirs (Fig. 3a). About 37% of the river export of TDN was exported to the Reservoir Pan-da in 2012 and 2017. Lake Baiyangdian, Reservoir Gang-Huang and Miyun received considerable amounts of TDN in the years 2012 (4.2 Gg year⁻¹, 5.4 Gg year⁻¹ and 3.6 Gg year⁻¹, respectively) and 2017 (2.4 Gg year⁻¹, 3.5 Gg year⁻¹ and 2.0 Gg year⁻¹, respectively). A 92% reduction in hotspot areas for river export of TDN and point TDN could be found from 2012 to 2017, mainly in the Baiyangdian and Pan-da basins (Fig. 3b, Table S7). However, the hotspot areas for river export of diffuse TDN did not show such apparent decreasing trends (Fig. 3b, Table S7). For example, the amounts of diffuse TDN export per km² in the hotspot sub-basins of Lake Baiyangdian (Rank IV) were even higher in 2017 compared with 2012 (Fig. 3b). One sub-basin of Reservoir Guanting changed to the hotspot (Rank IV) in 2017 but was in the Rank III level in 2012 (Fig. 3b).

The share of river export of diffuse and point TDN to five reservoirs differs over the years and sub-basins. Our results indicate that the contributions of animal manure to river export of diffuse TDN increased between 2012 and 2017 but the share of synthetic fertilizers in the river

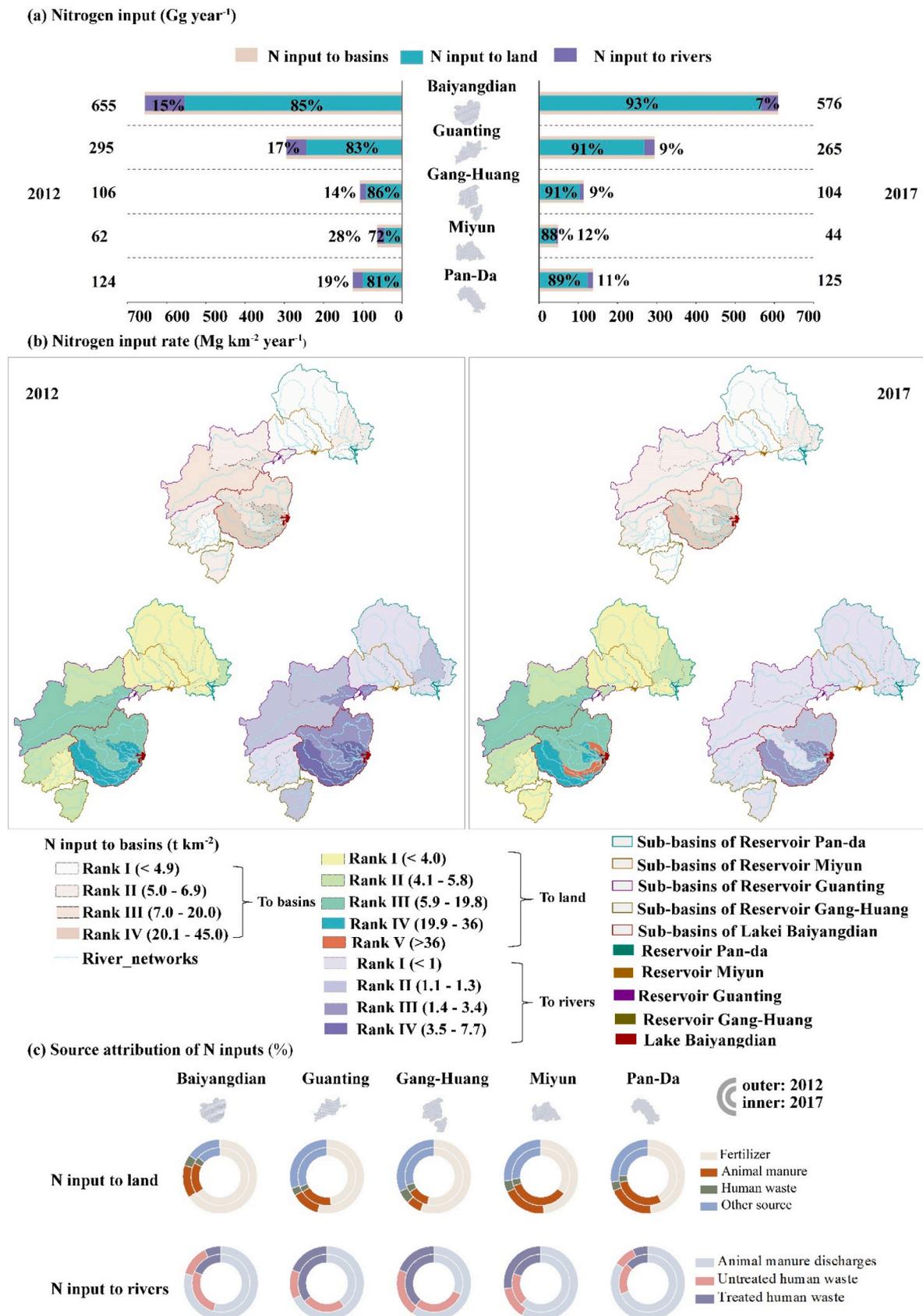
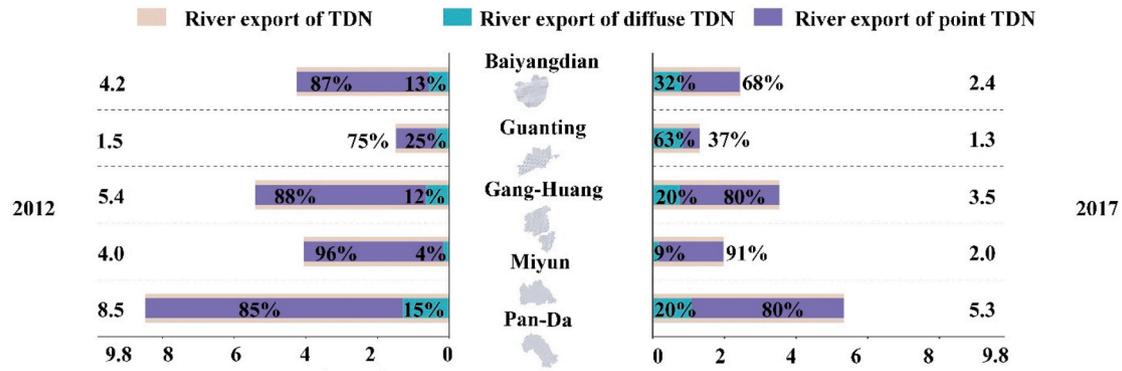
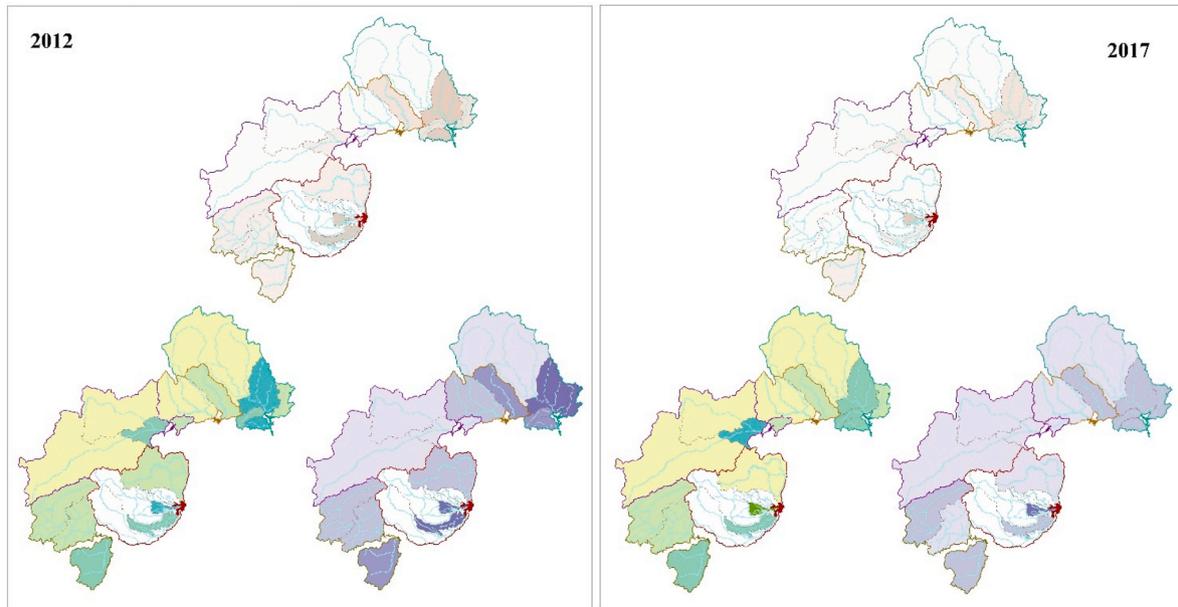


Fig. 2. (a) Nitrogen (N) inputs to Haihe sub-basins in 2012 and 2017 (Gg year⁻¹). (b) Hotspots for N inputs to sub-basins, land, and rivers in 2012 and 2017 (Mg km⁻² year⁻¹). (c) Source attributions of N inputs to land and rivers in 2012 and 2017 (%). N inputs to sub-basins include N inputs to land and N inputs to rivers. N inputs to land include N from synthetic fertilizers, animal manure, human waste, other source (i.e., atmospheric N deposition and biological N₂ fixation). N inputs to rivers include the point sources of N in rivers from direct discharges of animal manure, untreated human waste, and treated human waste from sewage systems. Source: MARINA-Lakes model (see sections 2.2 and 2.3).

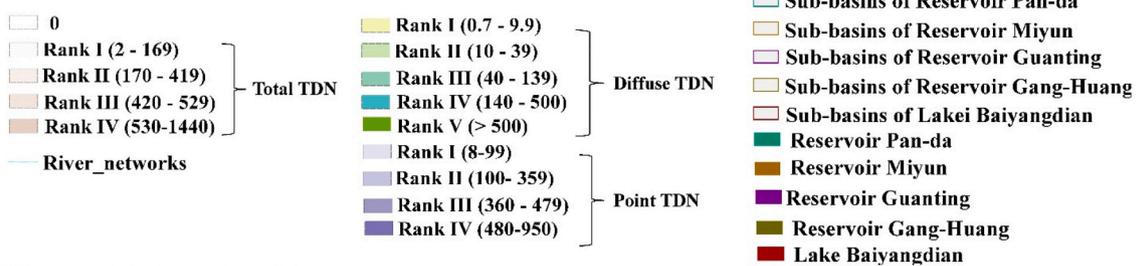
(a) River export of Total Dissolved Nitrogen (TDN, Gg year⁻¹)



(b) River export of TDN rate (kg km⁻² year⁻¹)



River exports of TDN to reservoirs (kg km⁻² year⁻¹)



(c) Source attribution of TDN (%)

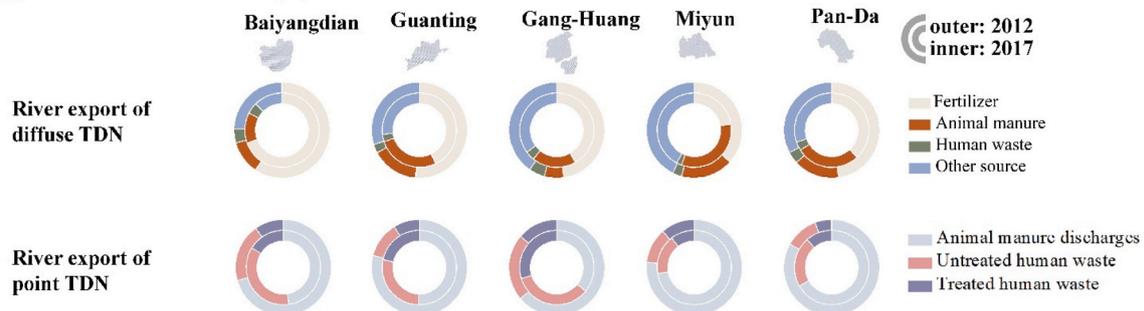


Fig. 3. (a) River export of Total Dissolved Nitrogen (TDN) to Haihe reservoirs in 2012 and 2017 (Gg year⁻¹). (b) Hotspots for river export of TDN, river export of diffuse TDN and river export of point TDN in 2012 and 2017 (Kg km⁻² year⁻¹). (c) Source attributions for river export of diffuse TDN and point TDN in 2012 and 2017 (%). Source: MARINA-Lakes model (see sections 2.2 and 2.3).

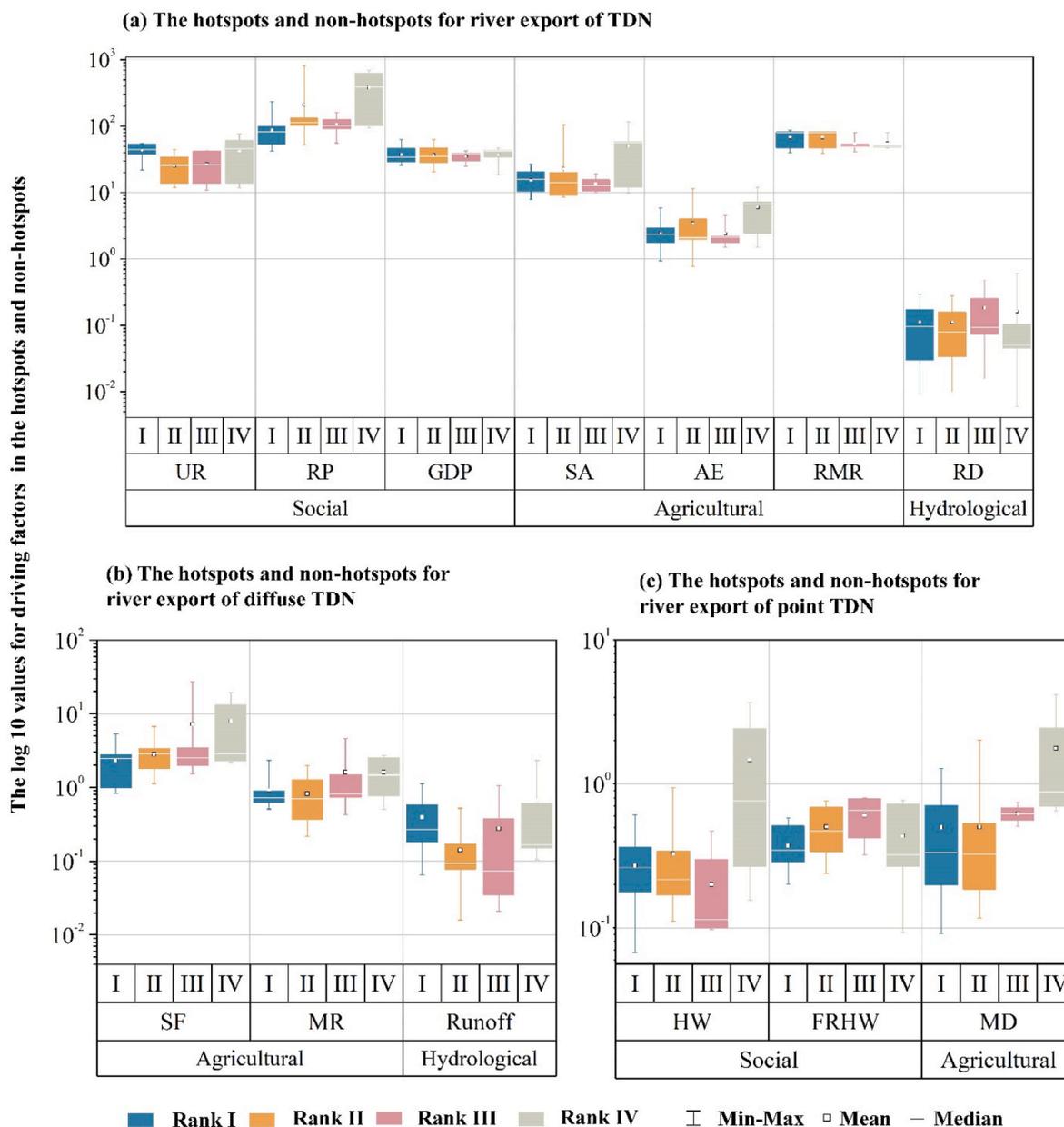


Fig. 4. The social, agricultural and hydrological factors in the hotspot and non-hotspots for river export of Total Dissolved Nitrogen (TDN) to Haihe reservoirs. The values of these factors were transformed to log10 for the same unit scale. (a) For the hotspot and non-hotspots of river export of TDN: social factors include urbanization rate (UR, %), rural population (RP, capital km⁻² year⁻¹) and Gross Domestic Product (GDP, 1000 RMB capital⁻¹). Agricultural factors include sown area (SA, ha km⁻² year⁻¹), animal excretion N (AE, ton km⁻² year⁻¹) and recycled N manure ratio (RMR, %). Hydrological factors include river discharges in the sub-basins (RD, km³ year⁻¹). (b) For the hotspot and non-hotspots of river export of diffuse TDN: agricultural factors include synthetic fertilizers N (SF, ton km⁻² year⁻¹) and recycled manure N (MR, ton km⁻² year⁻¹). Hydrological factors include runoff in the sub-basins (km³ year⁻¹). (c) For the hotspot and non-hotspots of river export of point TDN: social factors include human waste N (HW, ton km⁻² year⁻¹) and fractions of rural human waste N in total human waste (FRHW, %). Agricultural factors include discharging manure N (MD, ton km⁻² year⁻¹). Source: results in section 3.2 and Table S7 in the supplementary materials.

export of diffuse TDN decreased for most sub-basins (46%–39%). However, synthetic fertilizers were still essential contributors to the river export of TDN from diffuse sources to five reservoirs across all sub-basins in 2012 and 2017 (Fig. 3c). There is one exception. Synthetic fertilizers in the sub-basins of Lake Baiyangdian contributed 59% and 71% of river export of diffuse TDN in 2012 and 2017, respectively. The contribution of animal manure to the river export of diffuse TDN reached 28%, 25% and 23% in the sub-basins of Reservoir Miyun, Panda and Guanting in 2017, respectively. Other sources such as atmospheric N deposition and biological N₂ fixation, accounted for for around 40% of river export of diffuse TDN to reservoirs in the sub-basins of Guang-Huang and Miyun (Fig. 3c). Results indicate that the main

contributor to river export of point TDN changed from discharging animal manure to untreated human waste from 2012 to 2017 gradually. In 2012, the dominant point source was the direct discharges of manure, accounting for 64–88% of river export of point TDN to reservoirs (Fig. 3c) across sub-basins and years. In 2017, river export of point TDN was largely from untreated human waste that contributed by 11–36%. Lake Baiyangdian and Reservoir Gang-Huang were representative sub-basins where the dominant point source varied from direct manure discharges to untreated human waste between 2012 and 2017 (Fig. 3c).

3.3. Driving factors affecting N exports by rivers to reservoirs

For the river export of TDN to five reservoirs, the mean amount of N excreted by animals in hotspots (Rank IV) was two times higher than the N excretion in non-hotspot sub-basins (Rank I-III). But, the mean recycled manure ratios in the hotspots were lower than in non-hotspots (Fig. 4a, Table S7). Meanwhile, social indicators differ between hotspots and non-hotspot sub-basins. Hotspots were more urbanized but still had the highest rural population. On average, the number of rural people in hotspots was three times that in non-hotspot sub-basins (Fig. 4a, Table S7). No significant differences were observed in the mean Gross Domestic Product (GDP) between the hotspots and the non-hotspots (Fig. 4a, Table S7). The hydrology does not explain the variations in river export of TDN to five reservoirs because the river discharges were almost the same between the hotspot and the non-hotspot sub-basins (Fig. 4a

Table S7).

For the river export of diffuse TDN, the hotspot sub-basins held higher synthetic fertilizers N and recycled manure N compared with non-hotspots (Fig. 4b). The mean synthetic fertilizers N in the hotspots were two times that in non-hotspots (Table S7). Moreover, hotspot sub-basins held the highest mean runoff (Fig. 4b, Table S7). However, higher runoff in non-hotspot sub-basins could be found occasionally (Fig. 4b). It indicated that the runoff is an essential factor but may not be the determining one which can affect the river export of diffuse TDN to reservoirs.

For the river export of point TDN, the mean amount discharging animal manure N in hotspots was three times that in non-hotspot sub-basins (Fig. 4c). In addition, human waste N differed between hotspots and non-hotspots with an increasing trend from Rank I to Rank IV. Moreover, the fractions of rural human waste N increased from Rank I to Rank III. It indicated that human waste from rural areas accounted for much of the river export of point TDN to five reservoirs. But these fractions were a bit lower in the hotspots (Rank IV, 43%) than in non-hotspots (Rank I-III, 49%). It indicates that human waste in rural areas was not the only factor resulting in the highest river export of point TDN to five reservoirs (Fig. 4c, Table S7).

4. Discussion

4.1. Lessons from this study

Results in this study show that existing policies could have supported the mitigation of N pollution. For example, N input to basins and river export of N to five reservoirs decreased from 2012 to 2017 (Figs. 2a and 3a). Likewise, hotspot areas for river export of N to reservoirs shortened from four sub-basins to only one sub-basin between 2012 and 2017 (Fig. 3b, Table S7). The reasons explained here were the decrease in animal manure excretion and increased recycled manure ratios between 2012 and 2017 (Fig. 4a and c). That greatly decreases the percentage of discharging animal manure N to the reservoirs (Fig. 3b). In other words, policies about recycling more animal manure to crops associated with reducing manure discharges could have influenced inputs of N to rivers from land after 2015 (Fig. 1, Table S1). Therefore, manure management policies could influence the share of water pollution sources. In our study, we calculate a decrease in the share of point sources (i.e., less discharges of animal manure) and an increase in the share of diffuse sources (i.e., more animal manure used for crops) in the river export of TDN. This is because of the recycling of animal manure to crops (Fig. 1, Table S1). However, river export of diffuse N to the reservoir kept the number of sub-basins with hotspots in the period 2012–2017 years (Fig. 3b, Table S7). The possible reasons were the increased use of animal manure in crops but not an obvious decrease in the use of synthetic fertilizers. Synthetic fertilizers were still the main source affecting the river export of diffuse TDN to reservoirs (Fig. 4b). That means that the implementation of reduced synthetic fertilizers-

related policies (Fig. 1, Table S1) may still need further efforts and/or extra policies to reduce the river export of N from diffuse sources. In addition, untreated human waste gradually became the main contributor to river export of point TDN from 2012 to 2017. The first reason could be associated with the decreased share of discharging animal manure N to the river export of TDN (Fig. 3c). This could be related to animal manure management policies (Fig. 1, Table S1). The second reason could be the increased share of untreated human waste N in the river export of TDN (Fig. 3c) resulting from the higher rural population and poor wastewater treatment in villages (Fig. 4a and c). This implies that policies that are related to rural developments may need more attention to reduce N losses to reservoirs (Fig. 1, Table S1).

This study can help to identify hotspots of N inputs to sub-basins and river export of N to reservoirs of the Haihe basin. Lake Baiyangdian and Reservoir Pan-da were the most polluted and thus need more attention in pollution reduction strategies. Among five reservoirs, the N inputs to sub-basins of Lake Baiyangdian were the highest, and Pan-da received the highest river export of TDN (Figs. 2a and 3a). Likewise, hotspots for N inputs to sub-basins were mainly located in Lake Baiyangdian (Fig. 2b). Sub-basins in Lake Baiyangdian and Reservoir Pan-da dominated the hotspots of river export of TDN to reservoirs (Fig. 3b). That was because the sub-basins of Lake Baiyangdian had a higher rural population (Fig. 1b) and a large amount of synthetic fertilizers (Fig. 3c), both of which were factors affecting river export of TDN from point and diffuse sources (Fig. 4b and c). An increase in animal numbers could be found for the sub-basins of Reservoir Pan-da from 2012 to 2017 (Fig. 1b). That could have led to increasing animal manure discharges which was a necessary factor affecting the river export of TDN (Fig. 4c).

This study can help to prioritize sources of river export of N to five reservoirs of the Haihe basin. In the sub-basins of Reservoir Panda, Miyun and Gang-Huang, priority needs to be given to control point source pollution, among which effective management of livestock manure discharge and improved sewage management and construction in rural areas were critical (Fig. 3c). Controlling diffuse source pollution should be the priority for the Reservoir Guanting, mainly focusing on fertilizer management (Fig. 3c). In Lake Baiyangdian, both diffuse and point sources must be controlled. The N involved in agricultural production (e.g., synthetic fertilizers and animal manure) and sewage discharge in rural areas must be managed simultaneously (Fig. 3c).

4.2. Implications for environmental policies

Based on the lessons described above, we suggest specific options to realize AGD and meet local water quality requirements for five reservoirs in the Haihe basin. **Reservoirs Panda, Miyun, and Gang-Huang** face more strict water quality limits. N losses need to be reduced (Fig. 1). Moreover, sub-basins of these reservoirs are located in mountain areas (Fig. 1) where arable land may not be sufficient to bear large amounts of animal manure. Thus, addressing point source pollution (i.e., animal manure discharge) must be prioritized as the requirements in existing rules demand (Fig. 1, Table S1). For **Reservoir Guanting**, the most effective strategy is to control diffuse source pollution (i.e., N synthetic fertilizers). Farmers should be encouraged to optimize N fertilizer inputs (Zhang et al., 2022) by adopting such technologies as integrated soil-crop system management (Cui et al., 2018). For sub-basins of **Lake Baiyangdian**, the manure application needs to be enhanced further. This could be achieved by recycling more manure to crops and reducing the direct manure discharges to water and using synthetic fertilizers on land (MOST, 2015).

4.3. Uncertainty

Although our modeled results compare well to the measurements, we still realize that our model has uncertainties. Uncertainties are related to the model structure, model inputs and parameters. For the model

structure, the MARINA-Lakes model is based on the steady-state approach, which is limited in accounting for N dynamics in the different environment compartments (e.g., soils and water). For example, this model does not explicitly consider the link between BNF and P concentrations in soil solution. Previous studies do not agree on the effect of P content on terrestrial BNF across different biomes, compartments, and N fixation types (Burns and Hardy, 1975; Zheng et al., 2019). Moreover, our model system combined mechanistic and process-based approaches for model parameters and inputs. This implies that processes associated with retentions of N in soils and rivers are lumped into model parameters. The MARINA-Lakes model has been validated for several lakes, including Baiyangdian, Guanting in the Haihe basin and Taihu, Dianchi in the south of China (Li et al., 2019; Wang et al., 2019; Yang et al., 2019). The MARINA-Lakes model is developed based on the MARINA 1.0 model, which has been applied in large rivers in China (Strokal et al., 2016a). All the above studies indicate an acceptable performance for quantifying N river export to seas or lakes. Most model inputs were derived from existing studies, local investigations and statistical books (Tables S2, S3). For example, N inputs for agricultural productions were from the NUFER model, which was applied in provincial, city and county scales for China (Ma et al., 2013; Wang et al., 2018). Hydrological data used to calculate soil and river retentions was derived from measurements by the hydrological department. All these inputs are the most reliable datasets to our knowledge.

5. Conclusion

From 2012 to 2017, the river export of TDN to reservoirs decreased by 11–51%. In both years, the highest and lowest river export of TDN to reservoirs were found in Reservoir Pan-da and Guanting. A 92% reduction in hotspot areas for river export of TDN and point TDN was calculated from 2012 to 2017. However, the hotspot areas of river export of diffuse TDN did not show such apparent decreasing trends. These hotspots were mainly in the sub-basins of Lake Baiyangdian and Reservoir Pan-da.

Diffuse sources gradually became the main contributor to river export of TDN to five reservoirs in 2017, reaching 63% for the Guanting reservoir. As the main contributor to diffuse sources, the decreasing trend for synthetic fertilizers (46%–39%) and the increasing trend for animal manure were calculated from 2012 to 2017. The dominant point source for the river export of point TDN to the Haihe reservoirs was the direct manure discharges in 2012. However, in the sub-basins of Lake Baiyangdian and Reservoir Gang-Huang, the main contributor of point sources gradually changed from discharging animal manure to untreated human waste from 2012 to 2017.

Sub-basins of Lake Baiyangdian and Reservoir Pan-da were the most polluted compared to the others and thus require more attention in pollution control. The specific reservoir options for agricultural N management are needed to realize AGD and keep good water quality for five reservoirs in the Haihe basin.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.jenvman.2023.118667>.

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