PNAS
www.pnas.org

Supplementary Information for

Improvement in Municipal Wastewater Treatment Alters Lake Nitrogen to Phosphorus Ratios in Populated Regions

Yindong Tong[1], Mengzhu Wang[1], Josep Peñuelas[2,3], Xueyan Liu[4], Hans W. Paerl[5,6], James J. Elser[7], Jordi Sardans[2], Raoul-Marie Couture[8], Thorjørn Larssen[9], Hongying Hu[10], Xin Dong[10]*, Wei He[11], Wei Zhang[12], Xuejun Wang[13], Yang Zhang[14], Yi Liu[10], Siyu Zeng[10], Xiangzhen Kong[15], Annette B.G. Janssen[16] and Yan Lin[9]**

Affiliations

[1]School of Environmental Science and Engineering, Tianjin University, Tianjin 300072, China

[2]CSIC, Global Ecology Unit CREAF-CSIC-UAB, Bellaterra, Barcelona, Catalonia 08193, Spain

[3]CREAF, Cerdanyola del Valles, Barcelona, Catalonia 08193, Spain

[4]Institute of Surface-Earth System Science, Tianjin University, Tianjin 300072, China

[5]Institute of Marine Sciences, University of North Carolina-Chapel Hill, Morehead City 28557, USA

[6]College of Environment, Hohai University, Nanjing 210098, China.

[7]Flathead Lake Biological Station, University of Montana, Polson 59860, USA

[8]Department of Chemistry, Université Laval, Québe G1V0A6, Canada

[9]Norwegian Institute for Water Research, Oslo 0349, Norway

www.pnas.org/cgi/doi/10.1073/pnas.1920759117

[10]School of Environment, Tsinghua University, Beijing 100084, China
[11]School of Water Resources and Environment, China University of Geosciences (Beijing), Beijing 100083, China
[12]School of Environment and Natural Resources, Renmin University of China, Beijing 100872, China
[13]College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
[14]China-ASEAN Environmental Cooperation Center, Beijing 100035, China
[15]Department of Lake Research, Helmholtz Centre for Environmental Research-UFZ, Magdeburg 39114, Germany
[16]Water Systems and Global Change Group, Wageningen University & Research, Wageningen 6700, the Netherlands

Corresponding to Xin Dong and Yan Lin
Email: dongxin@tsinghua.edu.cn and Yan.Lin@niva.no

This PDF file includes:

- Supplementary text S1-S3
- Figures S1 to S8
- Tables S1 to S6
- Legends for Datasets S1 to S5
- SI References

Other supplementary materials for this manuscript include the following:

- Datasets S1 to S5

Supplementary Information Text S1

1. National Anthropogenic N and P discharges

We had analyzed temporal dynamics of N and P discharges from major anthropogenic sources during 2008–2017 in China, in order to identify driving forces for the alterations of TN, TP concentrations, and TN/TP mass ratios in lakes, by applying a hierarchical model of national nutrient cycling updated from Cui et al. (1), Liu et al. (2) and Tong et al (3). As the human activities had contributed to the bulk of N and P discharges into waterbodies in recent decades (1-3), in this study, five major anthropogenic sources were considered, including municipal and rural wastewater, livestock operation, crop farming and aquaculture. Since most of the statistical data is provided in the province-level in China (Data Set S3), all the calculated N and P discharges from five sources were presented in the province-level. The national-scale estimation of nutrient discharges was further aggregated from the province-level calculations. The nutrient discharges in the surrounding watersheds of 46 lakes with continuous nutrient monitoring data were estimated based on the province-level and the raster data comprised of population, livestock and land use distributions in China (see the Methods). A total of 31 provinces in China were included in this study. Taiwan, Hong Kong and Macao were not included due to the lack of data. Uncertainty analyses for the sectors were estimated based on the distributions of parameter values in calculations. Municipal wastewater discharge was obtained by a national-scale investigation during 20088–2017. Details in the estimating nutrient discharges in each sector were explained as follows:

1.1 Municipal Wastewater Discharge

The estimation of nutrient discharge form municipal wastewater was based on a national-level investigation about nutrient concentrations in influent and effluent of WWTPs, and volumes of treated and untreated wastewater during 2008–2017. During the decadal investigation, a total of 4,960 WWTPs were included. The N and P discharges from municipal wastewater were divided into two parts: direct nutrient discharge (through the untreated wastewater) and effluent nutrient discharge (through the effluent from treatment facilities). The treated nutrient discharge was calculated as follows:

$$M_{D1} = \sum \sum_{i=1}^{12} C_E \times V_E \times 10^{-2} \qquad \text{Equation 1}$$

M_{D1} represents the nutrient discharge after wastewater treatment in one year (Mg); C_E represents the average TN or TP concentrations in the effluent of WWTPs (mg/L); V_E represents the volume of treated wastewater in the month (10^4 m^3); 10^{-2} is the unit conversion coefficient.

The untreated discharge was calculated as follows:

$$M_{D2} = C_I \times V_I \times 10^{-2} \qquad \text{Equation 2}$$

M_{D2} represents the nutrient discharge by untreated municipal wastewater in one year (Mg); C_I represents the TN or TP concentration in the influent of WWTPs (mg/L); V_I represents the volumes of untreated municipal wastewater (10^4 m^3); 10^{-2} is the unit conversion coefficient. Details about the investigation results in each province were provided in the Supplementary Data Set S2. In this dataset, temporal changes of the number of WWTPs, volumes of treated and untreated municipal wastewater, TN and TP concentrations in the influent and effluent of WWTPs, nutrient discharges through untreated wastewater and effluent were provided.

1.2 Rural Wastewater Discharge

The nutrient discharges from rural wastewater were estimated based on the rural population and region-specific per-capita export coefficients of TN and TP (4-5). Sanitation facility in the rural areas is an important factor in determining nutrient discharges through domestic wastewater (6). Thus, the nutrient discharges from rural wastewater were generally divided into two categories in this study: discharge from rural people with access to compositing toilets, and discharge from rural people without access to compositing toilets. Based on the results of first Chinese pollution source census (7), different export coefficients were applied for these two categories (see details in Supplementary Data Set S3). Nutrient discharge by the rural residents with assesses to the compositing toilets was estimated as follows:

$$\mathrm{U}_{D1} = R_P \times P_C \times E_I \times 365 \times 10^{-2} \qquad \text{Equation 3}$$

U_{D1} represents the nutrient discharges by rural residents with access to compositing toilets in each province in one year (Mg). R_P represents number of rural residents in each province during 2008–2017 (10^4 people, see Supplementary Data Set S3); P_C represents the percentage of rural residents with the accesses to compositing toilets in each province (%, see Supplementary Data Set S3); E_I represents the daily N and P discharges for each people with accesses to the compositing toilets (g per day per person). Export coefficients are based on results of the first Chinese pollution source census, and varied significantly among regions (see Supplementary Data Set S3). The nutrient discharge by rural residents without assesses to compositing toilet was estimated as follows:

$$\mathrm{U}_{D2} = R_P \times (1 - P_C) \times E_L \times 365 \times 10^{-2} \qquad \text{Equation 4}$$

U_{D1} represents the nutrient discharges from rural residents with accesses to the compositing toilets in each province (Mg); E_L represents the daily N and P discharges for rural residents without access to the compositing toilet (g per day per person). The export coefficients are based on the results of first Chinese pollution source census, and varied among different provinces (see details in Supplementary Data Set S3).

1.3 Crop Farming Discharge

Crop farming refers to agricultural activities in the arable lands where nutrient discharges from the non-point sources were generated by the applications of N and P containing fertilizers. The hydrological characteristics varied distinctly among different types of arable lands, particularly between the paddy fields and dry lands (2-3). Therefore, in this study, we divided nutrient discharges from the crop farming into two categories: discharge from the paddy fields and discharge from the dry lands. Nutrient discharge from the paddy fields was estimated as follows:

$$C_{D1} = A_P \times E_P \times 10^{-3} \qquad \text{Equation 5}$$

C_{D1} represents the nutrient discharges from the paddy fields in each province in one year (Mg). A_P represents the areas of paddy fields in each province during 2008–2017 (ha, see the Supplementary Data Set 3). E_P represents the nutrient loss coefficient through the runoff in paddy fields (kg per ha). This coefficient was obtained from the first Chinese pollution source census (7), and varied significantly among different regions (see the details in Supplementary Data Set S3). Nutrient discharge from the dry lands was estimated as follows:

$$C_{D2} = A_D \times E_D \times 10^{-3} + A_D \times E_R \times 10^{-3} \qquad \text{Equation 6}$$

In the dry lands, two processes which can generate nutrient discharges were considered, including runoff and leaching. C_{D2} represents nutrient discharges from the dry lands in each province (Mg). A_D represents the area of dry lands in each province (ha, Supplementary Data Set S3). It was calculated by subtracting area of paddy fields from the total crop land area in each province. E_D represents the nutrient loss coefficient through runoff in the dry lands (kg per ha); E_R represents the nutrient loss coefficient through leaching in the dry lands (kg per ha). These export coefficients are based on the results of the first Chinese pollution source census, and vary largely among different provinces (see the details in Supplementary Data Set S3).

1.4 Livestock Operation Discharge

In this sector, the most farmed animal types in China were considered, including pig, cattle, sheep and poultry (2). In each province, livestock practices were generally divided into large-scale intensive farming and household low-intensive farming (3). The number of farmed animals and their generation coefficients of feces and urine were used to estimate nutrient discharges in each province. Nutrient discharges from the intensive and low-intensive farming were calculated as follows:

$$C_{D1} = (0.542N_A + N_G) \times P_I \times E_A \times P_R \times 365 \times 10^{-2} \quad \text{Equation 7}$$

$$C_{D2} = (0.542N_A + N_G) \times (1 - P_I) \times E_A \times P_L \times 365 \times 10^{-2} \quad \text{Equation 8}$$

C_{D1} and C_{D2} represents the nutrient discharges from the intensive and low-intensive farming for each type of animal in each province (Mg); N_A represents the slaughter amounts of pig, cattle, sheep and poultry in each province during 2008–2017 (10^4 head, Supplementary Data Set S3). To correct for the fact that the slain animals would no longer produce feces or urine, we corrected the annual number of slain animals in each province with a factor of 0.542 (8). N_G represents the number of animals on hands (10^4 head, see Supplementary Data Set S3). P_I represents the ratio of animals in the intensive farming out of all the domestic animals in each province (unitless). These values varied largely among different provinces and among different years (see Supplementary Data Set S3). E_A represents the daily nutrient discharge by the farmed animals in large-scale farming (gram per day per head). This value is based on the results of the first Chinese pollution source census (Supplementary Data Set S3). P_R represents the percent of nutrient loadings discharged into the waters in the large-scale farming (%). A value of 2–8% was adopted for P_R based on a previous study (9). P_R represents the percent of nutrient loadings discharged into the waters in the low-intensive farming (%). Based on the previous studies, a percentage of 40–70% was used for the provinces in South China, and a percentage of 20–40% was used for the provinces in North China (9-10).

1.5 Aquaculture Discharge

In cultivation of aquatic products, feed additives were used to promote the growth of aquatic animals (2). However, most of added feed additives cannot be effectively eaten by aquatic animals. In this study, aquaculture discharge was divided into three typical categories based on the types of the farmed aquatic animals, including fish, crustacean and shell. It should be noted that, based on the results from the first Chinese pollution source census, the nutrient export coefficients varied significantly among different specific types even within the same category. For the shellfish, most shellfish types, except river mussel, were fed on the uneaten food and excretions from other aquatic animals. Therefore, in each category of aquatic animals, 3–6 specific animal types were further divided (see details in Supplementary Data Set S3). Nutrient discharge from aquaculture was calculated as follows:

$$\mathrm{A}_D = P_F \times E_F + P_R \times E_C + P_S \times E_S \quad \text{Equation 9}$$

A_D represents the nutrient discharge from aquaculture in each province (Mg). P_F, P_R and P_S represent the annual productions of fish, crustacean and shell in each province during 2008–2017 (Mg). Detailed types of the aquatic animals and their annual productions in each province were provided in Supplementary Data Set S3. E_F, E_C and E_S represent the nutrient export coefficients for each type of farmed aquatic animals (g per kg). Values of export coefficients for each type of aquatic animal were provided in Supplementary Data Set S3.

Supplementary Information Text S2

In order to quantify the impacts of WWTPs on temporal changes in lake nutrient concentrations and their ratios during 2008–2017, we used the null hypothesis test to compare two datasets, one being the results of nutrient concentrations (and their ratios) assuming no new WWTPs were built after 2008, and wastewater treatment efficiency also remained unchanged during 2008–2017 (null scenario: NUS); one being the result of nutrient concentrations (and their ratios) where new WWTPs were built and efficiency was improved following the observed trajectory (real situation scenario: REA). Improvement in wastewater treatment was provided in Supplementary Data Set S2, where annual TN and TP removal efficiency were provided from 2008 to 2017 for all 31 provinces in China. Take the city of Beijing as an example, with a growing population and urbanization, the volume of municipal wastewater increased from 980 × 10^6 m^3 in 2008 to 1,504 × 10^6 m^3 (~50% increase) in 2018. However, the nutrient discharges were 37,993 Mg (for TN) and 2,868 Mg (for TP) in 2008 compared with 17,488 Mg (for TN) and 471 tons (TP) in 2017, representing a 50% reduction for TN and 84% reduction for TP. We calculated the nutrient discharges for both NUS and REA for all the target watersheds. Results showed that the null hypothesis are rejected for TN, TP concentrations and TN/TP mass ratio (P value < 0.05 and $F > F$ critical) (Table S5 and S6). Figure S7 provides the annual average nutrient concentration and their ratios for the REA and NUS scenarios. The results indicate that for the NUS scenario, if no new WWTPs were built, lake TN and TP concentrations would be significantly higher values than REA scenario by 2017 (~160% increase in TN concentration and ~50% increase in TP concentration). However, with the increase in the percentage of wastewater being treated, TN/TP mass ratios in lakes increased to 52 under REA in 2017, while if without newly built WWTPs, the corresponding TN/TP mass ratios in lakes would be 40 (NUS) in 2017.

Supplementary Information Text S3

To examine impacts of improvement in TN removal efficiency on future changes of TN/TP mass ratios in the lakes, we simply simulated three scenarios with different levels of improvement (1%, 2% and 3% per year) on TN removal efficiencies by WWTPs. Since China is still under the rapid urbanization in the next 10 years, future TN discharge is predicted by assuming that China's urbanization rate will increase by 1% per year, and under this assumption, TN discharge from the urban residents also increase by 1% per year. TN discharge and TN removal capacity in all the 31 provinces of China were provided in Supplementary Data Set S2. The following equations were used to estimate N discharges from municipal wastewater:

$$\mathrm{TN}_{rem\_i+1} = \mathrm{TN}_{rem\_i} \times (1 + e_{imp}) \quad \text{Equation 10}$$

$$\mathrm{TN}_{tot\_i+1} = \mathrm{TN}_{tot\_i} \times (1 + U_{rb}) \quad \text{Equation 11}$$

$$\mathrm{TN}_{dis\_i+1} = \mathrm{TN}_{tot\_i+1} - \mathrm{TN}_{rem\_i+1} \quad \text{Equation 12}$$

Where TN_{rem_i} represents the N removal capacity in the year of i (starting from 2017 and ending at 2026, Mt per year); TN_{remo_i+1} represents the N removal capacity in the year of i+1 (Mt per year); e_{imp} represents improvement in the percentage of TN removal efficiency by WWTPs. Improvements by 1%, 2% and 3% per year are applied respectively for the estimations under three scenarios; U_{rb} represents the increase of urbanization rate (with an annual increase of 1%); TN_{total_i} represents the TN discharge generated by the municipal waste water before treatment in the year of i (starting from 2017 and ending at 2026, Mt per year); TN_{total_i+1} represents the TN generated by municipal wastewater before treatment in the year of i+1 (Mt per year); TN_{dis_i+1} represents TN discharge from WWTPs after treatment in the year of i+1 (Mt per year). The calculation starts from year of 2018 predicted based on data of 2017, and ends at the year of 2027. The estimated future discharges would be used in the Equation 1 and 3 in Table S4 to predict the response in TN/TP ratios in lakes. Results showed that with an annual improvement of TN removal efficiency by 1%, TN/TP mass ratio in lakes could remain at the current level, while TN/TP ratios will start to decrease with an annual 2% improvement of TN removal efficiency (Figure S8). A bold assumption of annual improvement of TN removal efficiency by 3% can bring TN/TP mass ratios in lakes back to the level of 2008 over 10 years (Figure S8).

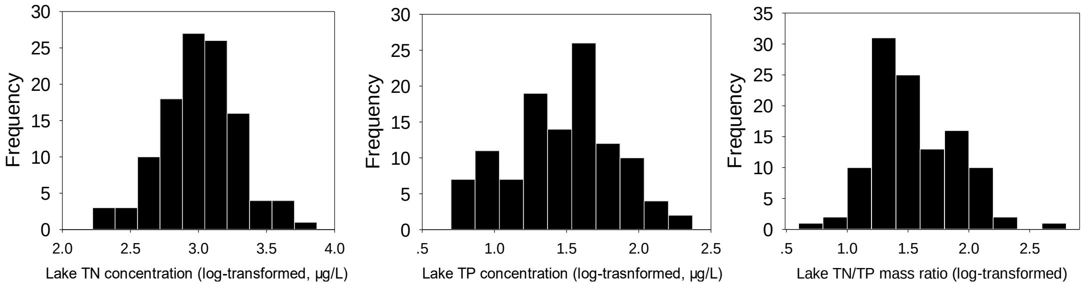


Figure S1. Statistical distributions of TN, TP concentrations and TN/TP mass ratios in 111 surveyed lakes in 2017. The details for each lake were provided in Supplementary Data Set S1.

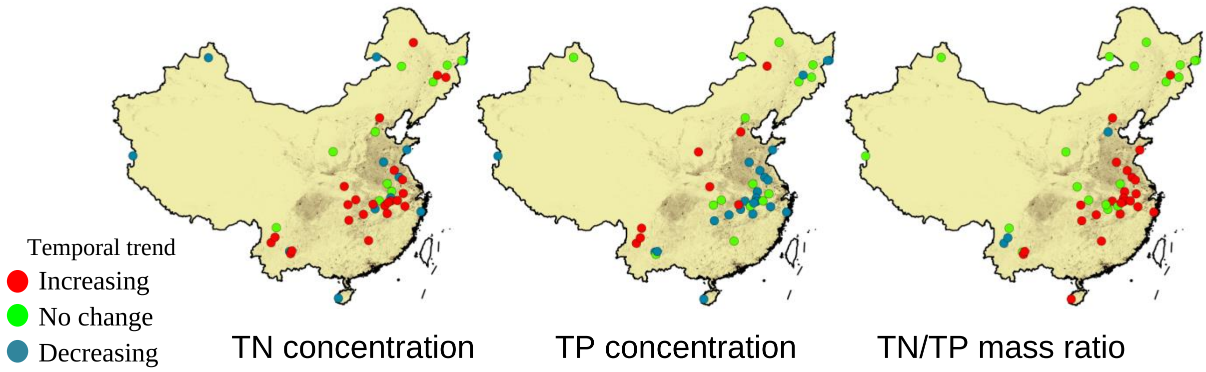


Figure S2. Temporal trends of TN, TP concentrations and TN/TP mass ratios in 46 lakes during 2008–2017. The details for each lake were provided in Table S2 and S3.

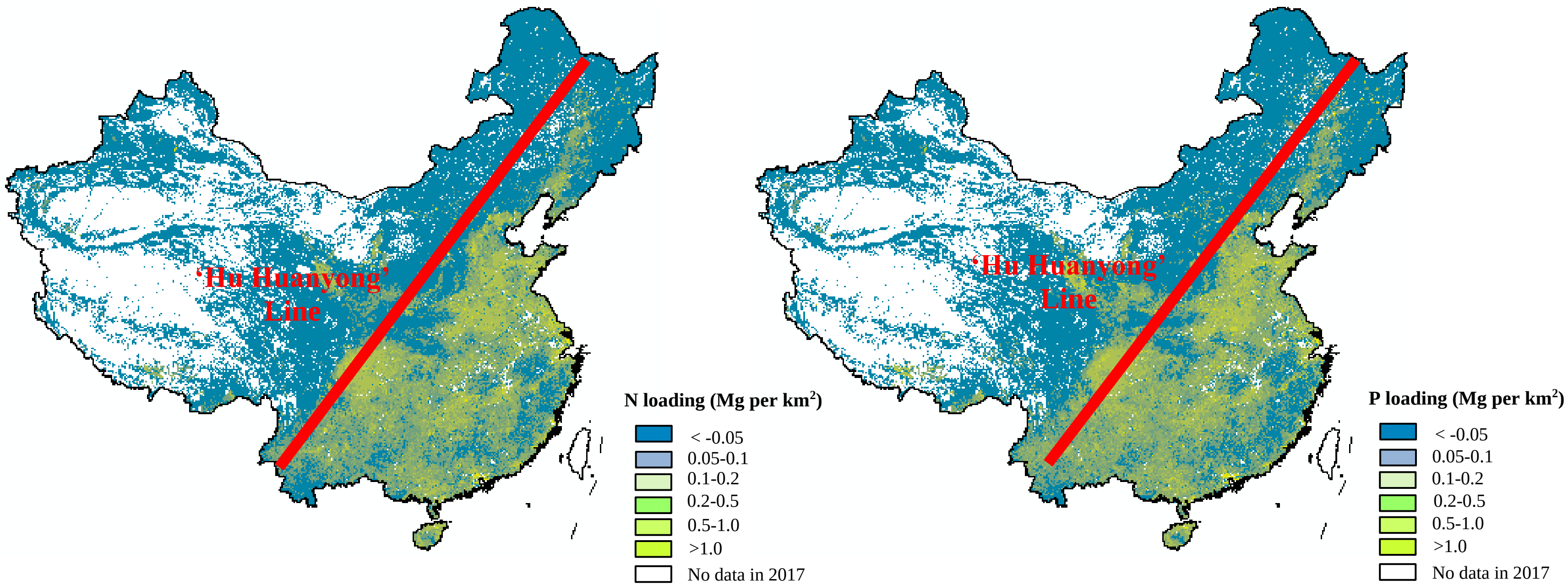
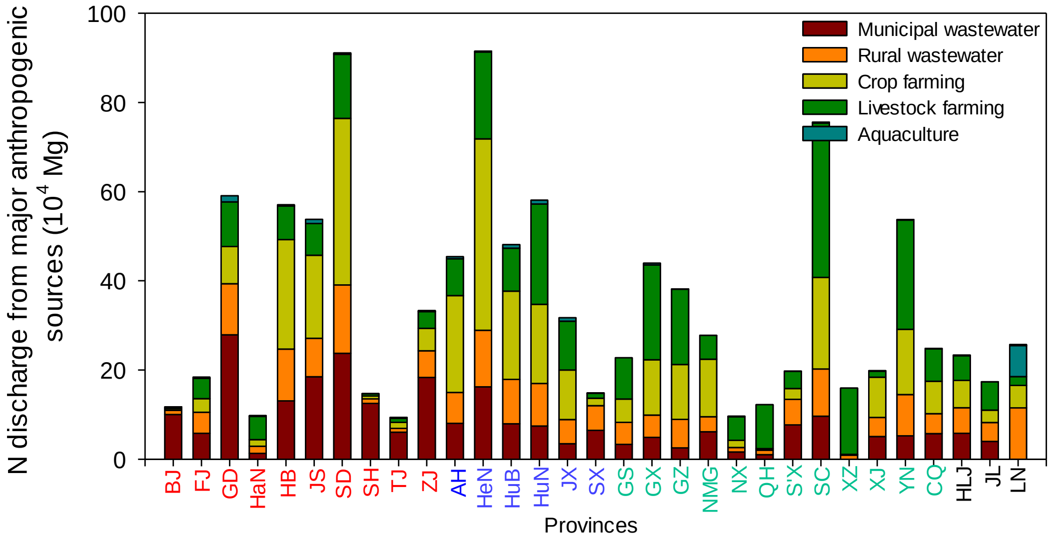
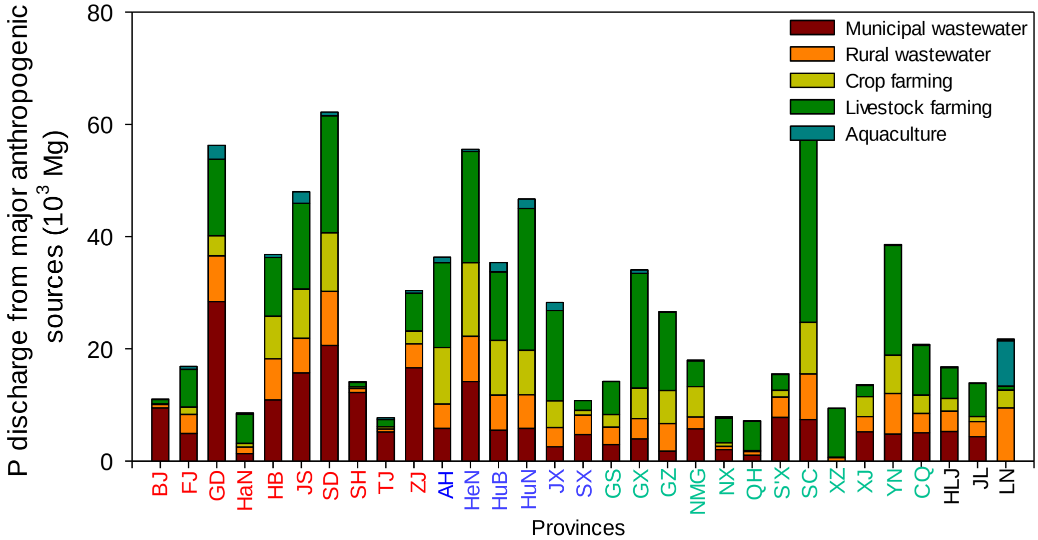


Figure S3. National N and P discharges in aggregated anthropogenic sources in 2017 (with a spatial resolution of 1 km × 1 km grid). The aggregated anthropogenic sources include municipal wastewater, rural wastewater, livestock farming, crop farming and aquaculture. The "Hu Huanyong" Line is used as a geographic boundary between the high-developed and densely-populated Eastern region and the less-developed and sparsely-populated Western region in China. The details about the calculations were provided in Supplementary Text S1.



(A)



(B)

Figure S4. N (A) and P (B) discharges by different anthropogenic sources in 31 provinces in China in 2017. Yearly changes of N and P discharges in each sector in 31 provinces are provided in the Supplementary Data Set S4 and S5.

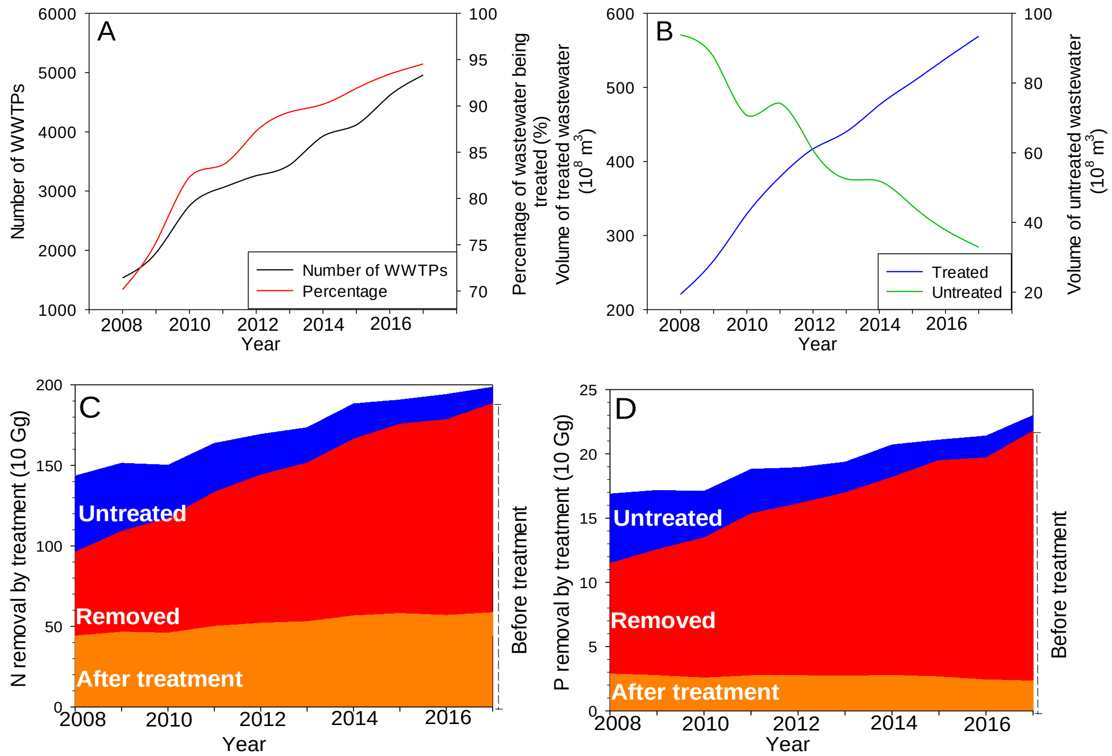


Figure S5. Summary about the advances in municipal wastewater treatment during 2008–2017 in China. The details in each province for each year were provided in Supplementary Data Set S2.

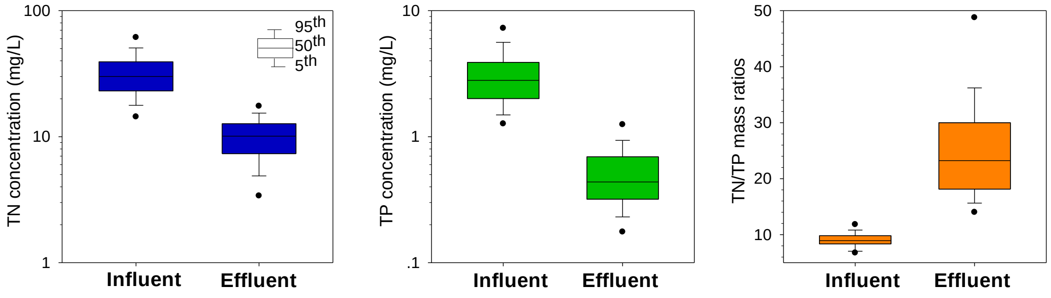


Figure S6. TN and TP concentrations and their mass ratios in the influent and effluent of WWTPs on the national scale in 2017.

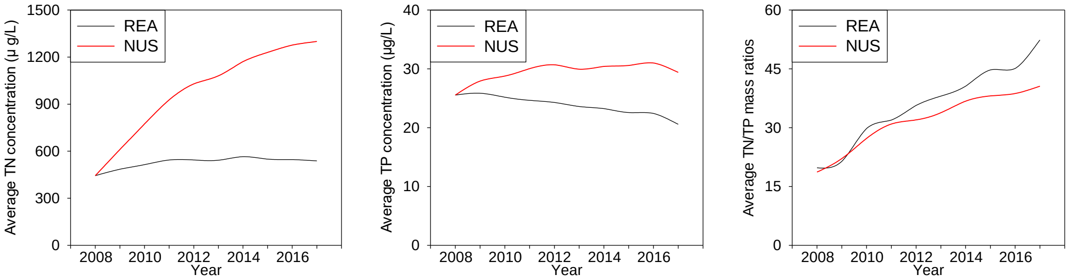


Figure S7. Temporal changes of the average TN, TP concentrations and TN/TP mass ratios in lakes during 2008–2017 under REA and NUS. The details were provided in Supplementary Text S2.

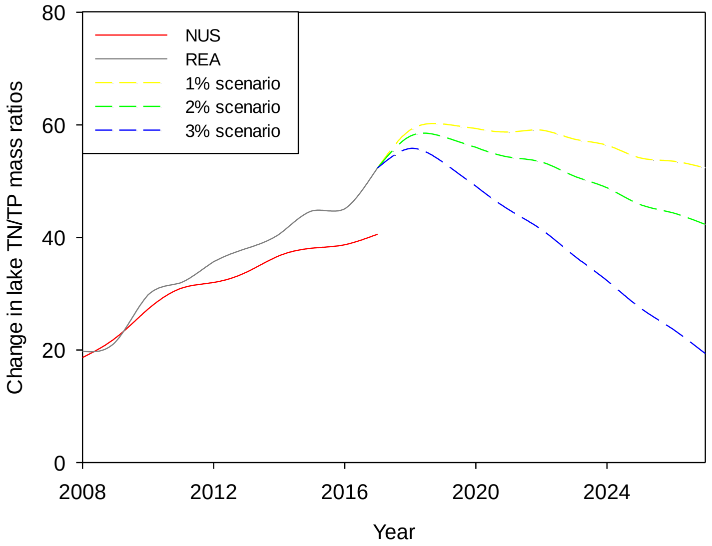


Figure S8. Response in lake TN/TP mass ratios under future scenarios with different improvements in TN removal efficiencies by WWTPs. The details about three scenarios were provided in the Supplementary Text S3.

Table S1. Limits of TN and TP concentrations based on China's water quality standards (GB3838-2002)

GB 3838-2002	Grade					
	I	II	III	IV	V	>V
TN concentration (μg/L)	≤200	≤500	≤1000	≤1500	≤2000	>2000
TP concentration (μg/L)	10	25	50	100	200	>200

Table S2. Summary about temporal trends in TN, TP concentration and TN/TP mass ratios in 46 lakes with continuous monitoring data during 2008–2017

Category	Number of lakes	Surface areas (km^2)	Water volumes (10^8 m^3)	Population density (10^4 people/km^2)	Wastewater discharge (10^6 m^3)	Change rate (10^{-3} $month^{-1}$)
TN concentration						
Increasing	22	138(25–2346)	29(5–214)	249(27–542)	58(1–1009)	6(1–34)
No change	12	50(13–2403)	13(2–132)	134(12–524)	3(0–199)	–
Decreasing	12	228(6–1222)	17(1–98)	131(1–685)	15(0–1663)	7(3–28)
TP concentration						
Increasing	8	62(11–342)	17(1–29)	95(21–247)	2(0–14)	6(2–14)
No change	16	143(31–1374)	40(6–146)	178(6–438)	28(0–416)	–
Decreasing	22	118(23–2380)	14(3–215)	250(20–716)	24(1–2433)	7(3–26)
TN/TP mass ratio						
Increasing	24	126(25–2380)	24(3–214)	304(101–717)	133(19–2449)	9(2–42)
No change	19	158(9–1207)	23(4–101)	37(1–459)	2(0–93)	–
Decreasing	3	242(92–354)	25(3–31)	228(112-251)	1(0–14)	4(2–10)

Table S3. Temporal changes about TN, TP concentrations and TN/TP mass ratios in 46 lakes during 2008–2017

Lake	Province	Precipitation/mm	Temperature/°C	Area/km^2	Volume/$10^8 m^3$	Population density /person/km^2	Number of sampling	K -TN	P -TN	K -TP	P -TP	K -NP	P -NP
Bailienhe	Hubei	1578.06	15.74	32.75	12.5	254.9	70	0.0038	0.118	-0.0173	0	0.0208	0
Baiyangdian	Heibei	558.42	13.74	366	0.6	227.8	114	-0.00008	0.94	0.0033	0.007	-0.0035	0.027
Bangongcuo	Xizang	107.45	-1.42	332.93	47	0.9	18	-0.028	0.014	-0.0245	0.003	0.0119	0.425
Beier	Neimenggu	171.54	1.74	608.78	37.5	0.0	34	-0.0155	0.009	0.0014	0.895	-0.0169	0.148
Caizi	Anhui	1653.94	16.90	71.47	3	448.1	72	-0.0065	0.001	-0.0132	0	0.0067	0.031
Chaerseng	Neimenggu	376.35	1.32	45.32	9.4	13.2	42	0.003	0.114	0.0061	0	-0.0029	0.198
Chao	Anhui	1408.17	16.89	786.91	20.7	624.3	115	-0.0005	0.76	-0.0029	0.001	0.0022	0.05
Chenghai	Yunnan	851.26	17.05	74.75	31.2	99.4	71	0.0043	0	0.0139	0	-0.0097	0
Daguangba	Hainan	1375.14	25.01	35.16	17.1	99.6	71	-0.0164	0	-0.026	0	0.0096	0.008
Dianchi	Yunnan	1044.84	15.60	298.34	4.2	480.2	120	-0.0072	0	-0.0075	0	-0.0004	0.677
Dongjiang	Hunan	1754.42	17.64	130.58	92	306.6	67	0.0339	0	0.0001	0.954	0.0335	0
Dongting	Hunan	1597.15	17.26	2432.5	220	300.3	120	0.0028	0	-0.0028	0	0.0046	0
Erhai	Yunnan	835.91	13.61	242.26	25.3	253.6	120	0.0013	0.003	0.0029	0	-0.0022	0.003
Fuxian	Yunnan	992.67	17.02	215.37	140.6	266.2	66	0.0033	0	-4E-16	0.06	0.0033	0.001
Fushui	Hubei	1768.44	16.53	56.81	17.3	162.0	71	-0.0061	0.001	-0.005	0.002	-0.0015	0.413
Gaoyou	Anhui	1036.47	15.94	703.14	55.5	546.2	60	0.0268	0	-0.0185	0	0.0453	0
Geheyan	Hubei	1290.11	13.27	40.9	34	90.2	64	0.0333	0	0.0007	0.78	0.0326	0
Hongze	Jiangsu	918.44	15.88	1374.36	135	608.5	120	-0.0039	0	-0.0058	0	0.0026	0.002
Huanglongtan	Hubei	825.94	14.91	12.7	11.6	90.6	71	0.0167	0	0.0061	0.022	-0.0022	0.456
Jingpo	Heilongjiang	570.69	3.96	73.44	35.6	15.0	82	0.002	0.019	0.0014	0.418	-0.0009	0.643
Laoshan	Shandong	479.51	12.93	1.71	0.5	657.4	119	-0.0026	0.001	-0.0061	0	0.0031	0.001
Lianhua	Heilongjiang	579.12	3.27	0.7	42	10.1	40	0.0023	0.346	-0.0047	0.339	0.007	0.2
Liangzi	Hubei	1594.51	17.98	302.36	22.5	233.3	72	0.0059	0	0.0071	0	-0.0025	0.072
Longgan	Hubei	1765.97	17.59	302.31	16.6	456.8	72	0.0049	0.018	0.0048	0.062	0.0001	0.968
Lugu	Yunnan	499.95	7.75	50.03	25.6	36.7	70	-0.0001	0.954	0.0019	0.021	-0.002	0.349
Luoma	Jiangsu	737.34	15.13	247.63	15	750.4	71	0.0066	0	-0.0048	0	0.0121	0

Miyun	Beijing	555.67	8.72	121.66	43.8	430.9	94	0.009	0	0.0024	0.197	0.0052	0.031
Mopanshan	Heilongjiang	578.66	5.01	25.15	3.2	121.7	64	0.0084	0	-0.0068	0.016	0.0152	0
Nansi	Shandong	665.23	14.63	1097.6	47	719.6	120	-0.0026	0.016	-0.0089	0	0.0054	0
Nanyi	Anhui	1663.34	16.76	145.01	6.8	297.5	72	0.011	0	0.0026	0.314	0.0084	0.033
Nierji	Heilongjiang	474.04	2.70	458.53	86.1	23.9	24	0.0291	0.001	0.0197	0.06	0.009	0.452
Poyang	Jiangxi	2121.59	18.97	2933	163.5	269.4	111	0.0022	0	-0.0017	0.237	0.0034	0.003
Qiandao	Zhejiang	2269.80	16.73	424.57	216.3	164.5	119	0.0009	0.043	-0.0091	0	0.0085	0
Shenjin	Anhui	2086.33	17.61	77.93	4.7	222.9	71	0.007	0.035	-0.004	0.001	0.0109	0.002
Shimen	Shannxi	1462.33	17.82	55.72	11	243.6	105	0.006	0	-0.0073	0.002	0.013	0
Songhua	Jilin	581.53	4.62	193.3	91.7	74.8	69	0.0011	0.221	-0.0011	0.453	0.0022	0.138
Taiping	Anhui	1885.88	16.76	57.55	26.4	324.7	70	0.0056	0.004	-0.0038	0.259	0.0102	0.009
Wabu	Anhui	1197.76	16.67	140.94	5.3	395.7	72	-0.0014	0.426	0.0004	0.797	-0.0018	0.359
Wangyao	Shannxi	416.49	9.19	9.73	2	47.8	66	-0.0078	0.048	0.0077	0.029	-0.0115	0.07
Wulungu	Xinjiang	156.50	1.52	854.89	68.4	8.1	42	-0.0074	0.001	-0.0041	0.101	-0.0017	0.594
Wuchang	Anhui	1937.47	18.05	46.62	2.5	443.0	70	0.0035	0.209	-0.0069	0.003	0.0104	0.003
Xiaoxingkai	Heilongjiang	617.43	4.25	157.63	4.1	54.0	48	-0.0048	0.034	-0.0079	0	0.0027	0.341
Xingkai	Heilongjiang	628.86	2.52	4380	181	18.7	63	-0.0003	0.846	-0.0044	0.004	0.0031	0.139
Yangzonghai	Yunnan	1121.47	16.11	30.55	12	195.7	48	0.0084	0.008	-0.0037	0.001	0.0076	0
Zhanghe	Hubei	1015.46	16.90	55.72	20.3	33.7	60	0.0084	0.084	-0.0001	0.972	0.0052	0.246
Changtan	Zhejiang	1551.62	13.83	23.27	6.9	193.0	60	-0.0007	0.784	-0.0122	0.001	0.0115	0.005

Table S4. Best linear models determined by BIC for each response variable

Response	Best model by BIC	R^2
TN	$0.0079*TN_{MW} + 0.00027\ TN_{LS}$	0.40
TP	$0.00081*TP_{MW} + 0.00023*TP_{LS} + 0.000064*TP_{CF} + 0.00068*TP_{RW}$	0.42
TN/TP	$3.829 * NP_{MW} – 18.906$	0.17

[1]Nutrient discharge intensity refers to nutrient discharge in watersheds normalized by the volume of lakes (Mg per 10^8 m^3 per year).

[2]TN_{MW}: N discharge intensity by municipal wastewater; TN_{LS}: N discharge intensity by livestock; TP_{MW}: P discharge intensity by municipal wastewater; TP_{LS}: P discharge intensity by livestock; TP_{CF}: P discharge intensity by crop farming; TN_{RW}: P discharge intensity by rural wastewater; NP_{MW}: TN/TP mass ratio in municipal wastewater discharge.

Table S5. Summary of ANOVA test results for REA and NUS scenarios

Source of Variation	SS	*df*	*MS*	*F*	*P-value*	*F crit*
TN						
Between Groups	48.1	1	48.1	33.9	0.00	3.85
Within Groups	1303.4	918	1.4			
Total	1351.4	919				
TP						
Between Groups	0.007	1	0.007	4.2	0.05	3.85
Within Groups	1.5	918	0.002			
Total	1.5	919				
TN/TP mass ratio						
Between Groups	6234	1	6234	18.9	0.00	3.85
Within Groups	295469	898	329			
Total	301703	899				

Table S6. Summary of nutrient concentrations and their mass ratios in lakes for REA and NUS

Groups	Count	Sum	Average	Variance
TN concentration				
TN_REA	460	242.5	0.53	0.55
TN_NUS	460	452.8	0.98	2.29
TP concentration				
TP_REA	460	10.9	0.024	0.001
TP_NUS	460	13.5	0.029	0.002
TN/TP mass ratio				
NP_REA	450	16722	37.2	432.0
N/P_NUS	450	14353	31.9	226.0

1. Supplementary Data Set S1 (separate file)

Annual average nutrient data (including TN, TP concentrations and TN/TP mass ratios), geographic locations, surface areas, water volumes for the 111 investigated lakes in 2017;

2. Supplementary Data Set S2 (separate file)

Details about the investigation results of WWTPs in 31 provinces during 2008–2017, including number of WWTPs, the volumes of treated and untreated wastewater, TN and TP concentrations in influent and effluent, N and P discharges through the treated and untreated wastewater in each province;

3. Supplementary Data Set S3 (separate file)

Anthropogenic activity data in 31 provinces during 2008–2017 and values for all the parameters in calculating N and P discharges in each province;

4. Supplementary Data Set S4 (separate file)

Temporal changes of N discharges in each sector (including municipal and rural wastewater, livestock operation, crop farming and aquaculture) in 31 provinces during 2008–2017;

5. Supplementary Data Set S5 (separate file)

Temporal changes of P discharges in each sector (including municipal and rural wastewater, livestock operation, crop farming and aquaculture) in 31 provinces during 2008–2017;

References

1. S. H. Cui, Y. L. Shi, P. M. Groffman, W. H. Schlesinger, Y. G. Zhu, Centennial-scale analysis of the creation and fate of reactive nitrogen in China (1910–2010). *Proc. Natl. Acad. Sci. U.S.A.* **110**, 2052-2057 (2012).
2. X. Liu et al., Intensification of phosphorus cycling in China since the 1600s. *Proc. Natl. Acad. Sci. U.S.A.* **113**, 2609-2614 (2016).
3. Y. D. Tong *et al*., Decline in Chinese lake phosphorus concentration accompanied by shift in sources since 2006. *Nat. Geosci.* **10**, 507-511 (2017).
4. T. Ma *et al.*, China's improving inland surface water quality since 2003. *Sci. Adv.* **6**, eaau3798 (2020).
5. S. C.Chapra *et al.*, Climate change impacts on harmful algal blooms in U.S. freshwaters: A screening-level assessment. *Environ. Sci. Technol.* **51**, 8933-8943 (2017).
6. Y. D. Tong *et al.*, Impacts of sanitation improvement on reduction of nitrogen discharges entering the environment from human excreta in China. *Sci. Total Environ.* **593-594**, 439-448 (2017).
7. Ministry of Agriculture, China. Generation and emission coefficient manual for the first Chinese Pollution Source Census. (*Ministry of Agriculture, China, 2009*).
8. W. J. Yan, E. Mayorga, X. Y. Li, S. P. Seitzinger, A. F. Bouwan, Increasing anthropogenic nitrogen inputs and riverine DIN exports from the Changjiang River basin under changing human pressures. *Global Biochem. Cy.* **24**, GB0A06 (2010).
9. B. J. Gu, X. T. Ju, S. X. Chang, Y. Ge, J. Chang, Nitrogen use efficiencies in Chinese agricultural systems and implications for food security and environmental protection. *Reg. Environ. Change* **17**, 1217-1227 (2017).
10. Ministry of Environmental Protection of the People's Republic of China. Livestock and poultry breeding pollution control engineering specifications (*China Environmental Science Press*, *2009*)