Supporting information

Multi-scale modeling of nutrient pollution in the rivers of China

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Supporting information consists of the following sections:

S1 Model description

We developed a new modeling approach to simulate the nutrient inputs to rivers on multiple scales (polygon, grid, sub-basin and county) for China. The modeling framework is based on three existing models: the MARINA 1.0 model (*M*odel to *A*ssess *R*iver *I*nputs of Nutrients to se*A*s)[1], the NUFER model (*NU*trient flows in *F*ood chains, *E*nvironment and *R*esources use)[2] and the hydrological model VIC (*V*ariable *I*nfiltration *C*apacity)[3]. The study area is introduced in section S1.1. The multi-scale nutrient model is described in general in section S1.2, followed by descriptions of point sources (section 1.3), diffuse sources (section 1.4), other sources (section 1.5), and model outputs on multiplescales (section 1.6).

S1.1 Study area

Before we go to the details of the model, we first introduce the the study area. The study area of China is presented in Figure S1. The county and provincial maps indicate that the total modeling area covers 87% of the total area of China. Tibet and Taiwan are not included because of data limitation. The sub-basin areas are delineated based on a DDM30 flow direction map of Döll and Lehner[4], which is identical to the flow direction network used in the global hydrology model, VIC[3]. The sub-basins cover 14 main rivers in China (Figure S1A). The bulletin of the ecology and environment of China reports that 30-40% of the Chinese rivers are classified as rivers with poor water quality (i.e. class Ⅳ and below according to Chinese water quality standards, meaning that direct contact with humans is not preferred) over the period of 2000 to 2017[5, 6]. The water quality of Changjiang improved more than that in other rivers. The drainage area with the poor water quality reduced to 15% in the year 2017. The Hai basin is the most polluted basin. Here, the polluted area still accounts for 58% of the total river basin area in 2017. The State Council issued an Action Plan for Prevention and Control of Water Pollution[7]. Measures such as improving the treatment of wastewater, better management of rural agricultural activities and of livestock production are proposed. The action plan remains general on the national level. The Ministry of Agriculture proposed measures in particular for livestock production, aiming towards sustainable resource use and a better environment[8]. For seven regions, specific measures are proposed (Figure S1B). For example, for the BJS region (Beijing, Tianjin, and Shanghai), advanced treatment of the animal manure is proposed because of the limited crop land in that region. The North China Plain (provinces of Hebei, Shanxi, Shandong and Henan) is the main region for crop and livestock production in China. Manure recycling on land through centralized collection and treatment facilities is proposed in this region. For more details we refer to the associated policies[7, 8].

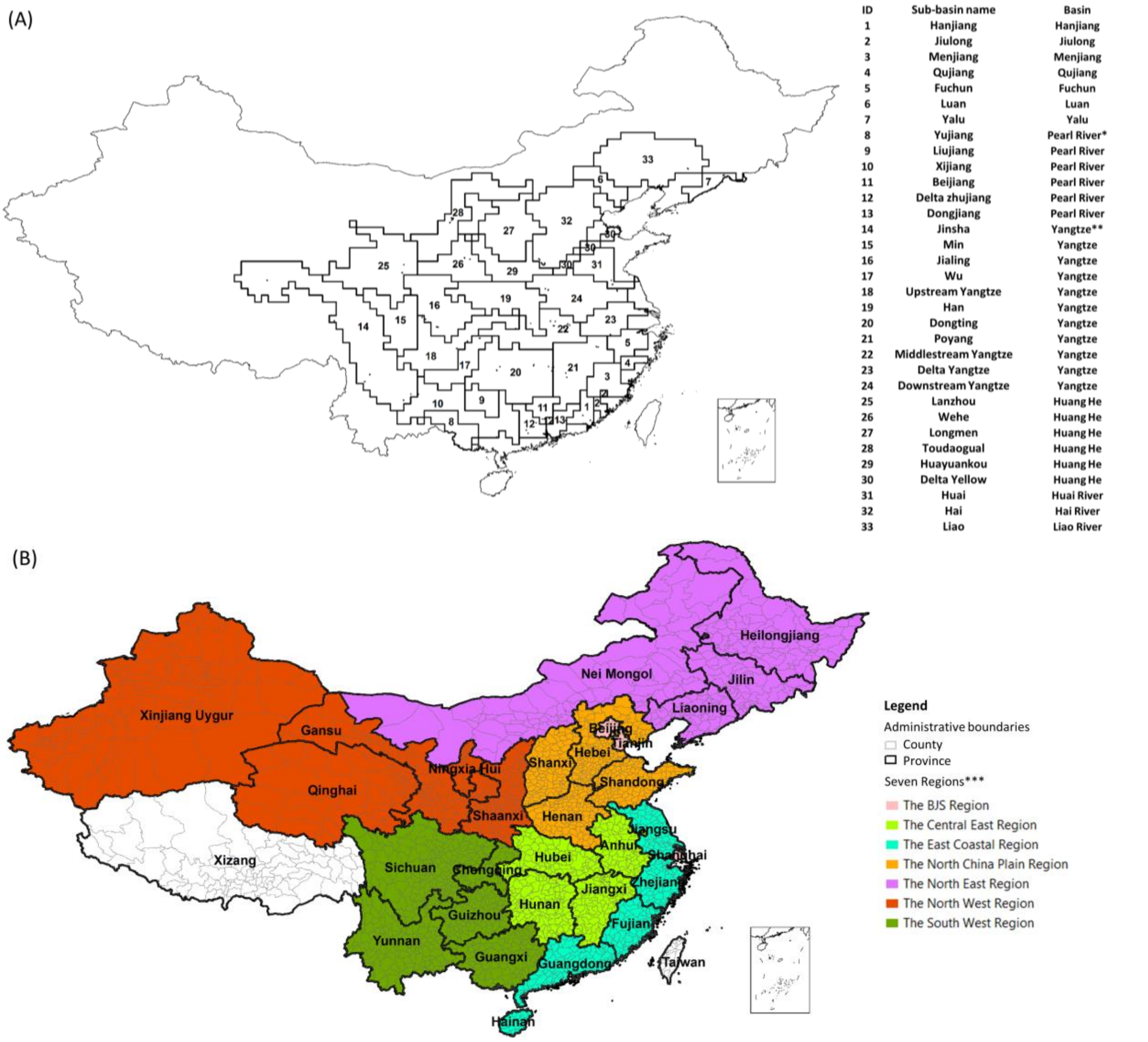

Figure S1. Maps of the study area on multiple scales: (A) Main river sub-basins in China that are included in our study. The border black lines indicate the boundaries of the river sub-basins. (B) The provincial and county map of China. The white area indicates that the detailed input data are not available. Thus, these areas are not included in our study. *The Pearl River is Zhujiang in Chinese. **: The Yangtze River is Changjiang in Chinese. ***:The seven regions is defined as the policy that proposed measures for animal manure management[8].

S1.2 General overview

Figure 1 in the main text presents an overview of our modeling on different scales. Here, we illustrate how different modeling processes are applied on different scales by source category (diffuse, point and others).

Diffuse sources include a number of explicit land-based sources of nutrients in rivers (A) that are modelled for each polygon while accounting for retention processes on land (C), and parameterized export processes (B):

A. Explicit land-based sources:

1) Synthetic fertilizers (for DIN, DIP, DON and DOP);
2) Animal manure applied in the agricultural area (for DIN, DIP, DON and DOP);
3) Human excreta from population that is not connected to the centralized wastewater treatment plants (WWTPs) (for DIN, DIP, DON and DOP)
4) Atmospheric N deposition on agricultural areas (for DIN)
5) Atmospheric N deposition on non-agricultural areas (for DIN)
6) Biological N_2-fixation by agricultural crops (for DIN)
7) Biological N_2-fixation by natural vegetation (for DIN)
8) Nutrient export from agricultural land by crop harvesting and animal grazing (for DIN, DIP, DON and DOP)

Model inputs for (1), (2), (3), (4), (5), (6) and (8) are from the county database for 2012 and NUFER county outputs. These inputs are first quantified on the county scale and then transformed to polygons within counties using a land-use weighted method (see section S1.4). Inputs for (7) are from the gridded datasets of the original Global NEWS-2[9] model (Nutrient Export from WaterSheds) for the year 2000. These inputs are distributed to polygons within grids by a simple area weighted method (see section S1.4).

B. Parameterized export processes:

9) Weathering of P-contained minerals (for DIP)
10) Leaching of organic matter from agricultural and non-agricultural soils (for DON and DOP)

Both (9) and (10) are quantified as a function of runoff, following the Global NEWS-2 export coefficient approach[9]. For runoff, the half-degree hydrology from VIC is used. Polygons within the grid have the same value as the grid to which these polygons belong to(see section S1.4).

C. **Soil retention:** soil retention is calculated as a function of runoff following Global NEWS-2[9]. Runoff on half-degree grids is from VIC[10, 11]. Polygons within the grid are assigned the value of that grid (see section S1.4).

Point sources include:

11) Direct discharges of untreated manure to water bodies (for DIN, DIP, DON and DOP);
12) Human waste from centralized wastewater treatment plants (WWTPs) (for DIN, DIP, DON and DOP);

Direct discharges of manure (11) are based on the county database 2012 and NUFER county outputs. These inputs are, first, quantified on the county scale and then transformed to polygons within the counties using a land-use weighted method (see section S1.3). Data for (12) are based on the WWTPs database that we developed and includes 4204 WWTPs in China. We calculate model outputs for each WWTP and store the calculated information in the polygons according to the XY coordinates of the WWTPs.

Others:

13) Human excreta from the urban and rural population that is discharged to water bodies (DIN, DIP, DON and DOP);

Data for (13) are from the county database 2012, NUFER county outputs and a dataset for the city and county levels from the China Urban-Rural Construction Statistical Yearbook for year 2012 and 2014. These sources are quantified for each county and we used a simple area weighted method to transform to polygons (see section S1.5).

S1.3 Point sources

Point sources of dissolved N and P inputs to rivers ($RS_{\boldsymbol{pnt_F}}$, kg year^{-1}) include direct discharges of manure to rivers ($RS_{pnt\_ma,F}$) and human waste emitted from centralized waste water treatment plants (WWTPs) ($RS_{pnt\_con,F}$). We quantify point sources based on MARINA 1.0 (Eq.S1) as:.

$$\boldsymbol{RS_{pnt_F} = RS_{pnt\_ma,F} + RS_{pnt\_con,F}} \quad \text{(Eq.S1)}$$

For the direct manure discharges ($RS_{pnt\_ma,F}$, kg year^{-1}) the improvements relative to the MARINA 1.0 model include:

(1) We update inputs to the year 2012 from the county database of the NUFER [12, 13]:

$$\boldsymbol{RS_{pnt\_ma,F}(county) = T(E)ma_{dis} \cdot EF_{pnt_{ma,F}}} \quad \text{(Eq.S2)}$$

Where, $T(E)ma$ is the nutrient element E (N or P) in animal manure that is directly discharged to water bodies (kg year^{-1}). The $T(E)ma_{dis}$ is updated with the NUFER outputs on the county scale. It is calculated based on the product of the total manure excretion (quantified based on the county database and literature) and the rate of the direct discharges of manure per county (derived from on-site surveys)[4, 29]. $EF_{pnt_{ma,F}}$ is the fraction of element E (N or P) in animal manure entering surface waters as form F (DIN, DON, DIP, DOP) (0-1). This fraction is taken from the MARINA 1.0 model and it is derived based on literature[1, 14-17]. This fraction is 0.7 for DIN and DIP, and 0.3 for DON and DOP.

(2) We use the locations of animal manure farms to transform county inputs to polygon units. The rural residential area from the land-use data (1×1 km grid)[18] is used as a proxy for locations of animal farms (Figure S2) as in the study of Zhao et al. [19]:

$$\boldsymbol{RS_{pnt\_ma,F}(polygon) = RS_{pnt_{ma},F}(county) * Frac_{farms\_county}} \quad \text{(Eq.S3)}$$

$Frac_{farms\_county}$ is the fraction of animal farms in a county that are located in the polygon. Here, we describe how we use a **land-use weighted method** and farm locations in the distribution of direct manure discharges in three steps (Figure S2):

1. The ArcGIS intersect tool is used to intersect the polygon-based map for China with the locations of animal farms (1×1 km grid);
2. The number of animal farms (i.e. number of 1×1 km grid grids) within each polygon is used to calculate the number of farms within each county in ArcGIS (Figure S3).
3. $Frac_{farms\_county}$, i.e. the fraction of animal farms in a county that are in a polygon is calculated as:

$$Frac_{farms\_county} = \frac{Number\ of\ animal\ farms\ (grids\ of\ 1x1\ km)\ in\ the\ polygon}{Number\ of\ animal\ farms\ (grids\ of\ 1x1\ km)\ in\ that\ county} \quad \text{(Eq.S4)}$$

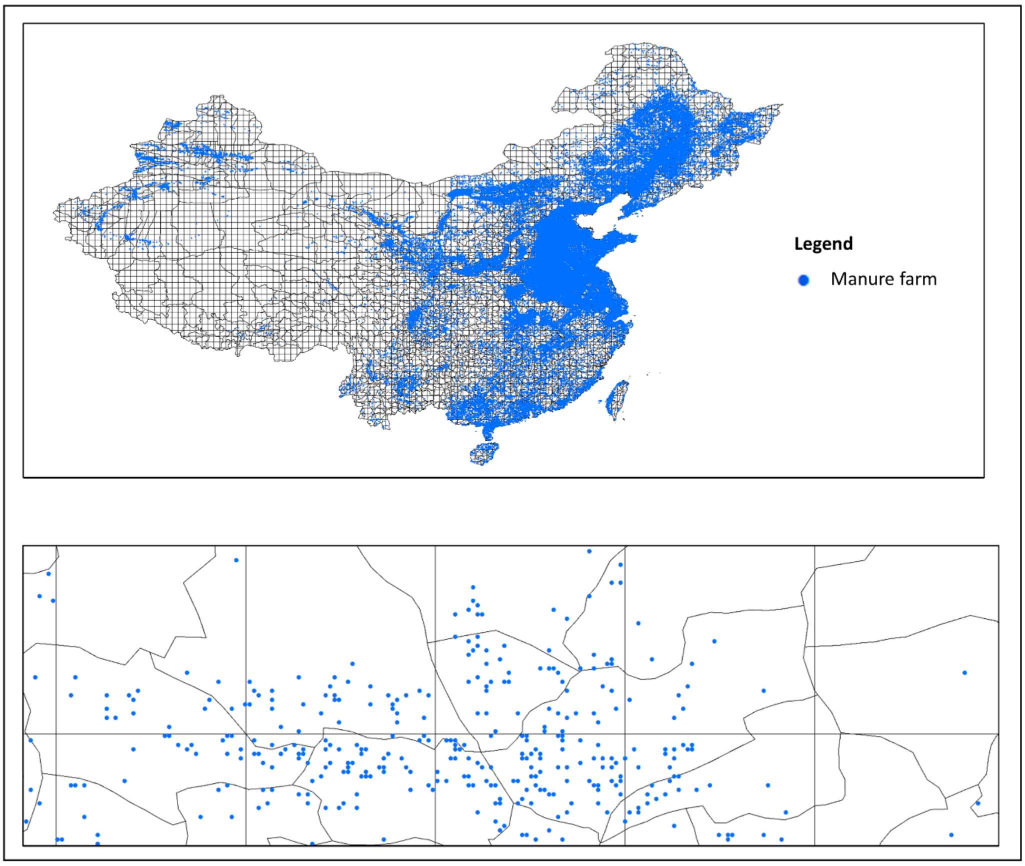

Figure S2. Locations of rural residential areas (a proxy for manure farms) that are intersected with polygons.

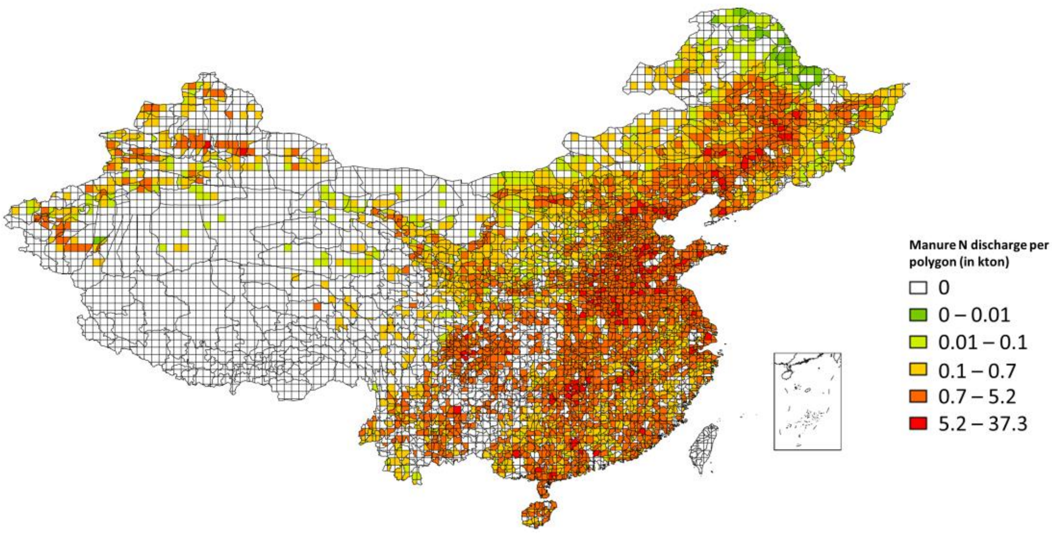

Figure S3. Direct discharges of nitrogen (N) animal manure to rivers by polygon (kton N/year).

For human waste emitted from centralized WWTPs ($RS_{pnt\_con,F}$, kg year^{-1}), we create a unique WWTPs database covering 4204 WWTPs across China (section S3). The database includes the location (longitude and latitude), average daily treatment capacity, treatment technologies and associated treatment efficiencies for individual WWTPs (section S2 and S3). The list of WWTPs is obtained from a national list of operating WWTPs for the year 2014[20] and the National Intensive Monitoring and Control Enterprise List for WWTPs for 2016 [21]. We locate individual WWTPs (XY coordinates) according to the address provided in the list. We estimate the treatment efficiencies for WWTPs based on 46 technologies applied in those WWTPs according to literature and expert opinion (see details in section S2, Table S3). We also update relevant model parameters such as urban and rural population from the county database and treatment rates for city and county levels from the China Urban-Rural Construction Statistical Yearbook for 2012 and 2014 [22, 23] (see an overview of input sources in Table S1).

With the WWTPs database, the nutrient inputs to rivers from individual WWTPs can be quantified following the same approach from Van Drecht, et al.[24] and we adjusted for the individual WWTPs in China as:

$$\boldsymbol{RS_{pnt\_con,F}} = \{(\boldsymbol{1} - \boldsymbol{hw_{frem,E}}) \cdot \boldsymbol{PopCon_{WWT}} \cdot \boldsymbol{E(E)_{pnt}} \cdot \boldsymbol{Frac_{treat}}\} \cdot \boldsymbol{FE_{pnt_F}} + (\boldsymbol{1} - \boldsymbol{Frac_{treat}}) \cdot \boldsymbol{PopCon_{WWT}} \cdot \boldsymbol{E(E)_{pnt}} \cdot \boldsymbol{FE_{pnt_F}} \quad \text{(Eq.S5)}$$

$$\boldsymbol{PopCon_{county}} = \boldsymbol{Pop_{urb}} * \boldsymbol{ConRate_{urb}} + \boldsymbol{Pop_{rur}} * \boldsymbol{ConRate_{rur}} \quad \text{(Eq.S6)}$$

$$\boldsymbol{PopCon_{WWT}} = \boldsymbol{PopCon_{county}} * \boldsymbol{CapacityWeigh_{WWT}} \quad \text{(Eq.S7)}$$

Where, $hw_{frem,E}$ is the removal of nutrient element E (N or P) during treatment in sewage systems (fraction,0-1). $\boldsymbol{E(E)_{pnt}}$ (kg person^{-1} year^{-1}) is the input of nutrient element N or P to watersheds (land) resulting from human excreta ($\boldsymbol{E(E)_{hum}}$for both N and P) and inputs of P detergents from laundry (E^P_{Ldet}) and dishwashers (E^P_{Ddet}) in sewage influents (kg person^{-1} year^{-1}).These are calculated as a function of GDPppp and GDPmer (purchasing power parity based and market exchange rate based GDP in 1995 U.S. dollars person^{-1} year^{-1}) on the province scale according to Van Drecht et al.[24]. $\boldsymbol{FE_{pnt_F}}$ is the fraction of sewage effluents exported to rivers as nutrient form (F) (0-1). $\boldsymbol{FE_{pnt_{DIN}}}$ is directly proportional to the removal rate of N during treatment, while for other nutrient forms it is the calibrated constant from Global NEWS-2. $PopCon_{WWT}$ is the population number connected to the individual WWT. Pop_{urb} and Pop_{rur} refer to the urban and rural population on a county level respectively (people/year). $ConRate_{urb}$ and $ConRate_{rur}$ refer to the fraction of the national urban and rural population that is connected to centralized WWTPs (0-1). The fraction value is 0.739 for urban and 0.018

for rural, respectively[25]. $CapacityWeigh_{WWT}$ is a fraction of the treatment capacity of the individual WWT to the total capacity of the WWTPs located in the county (0-1). By introducing this parameter, we distribute the connected population from county to individual WWTPs. $Frac_{treat}$ is a treatment rate for the centralized WWTPs, i.e. the fraction of the wastewater that is treated in the total wastewater transported to the WWTPs (0-1). $Frac_{treat}$ may vary among different cities. Thus we distinguish this model parameter between urban and rural per city (Table S1, section S3). For WWTPs located in the urban area, we apply the treatment rate for urban area. For WWTPs located in the rural area, we apply the treatment rate for rural area.

Thus, the outputs of dissolved N and P to rivers from individual WWTPs are also included into our final WWTPs database (section S3). The outputs from individual WWTPs are assigned to polygons where the WWTP is located (longitude and latitude). We do this to calculate the total dissolved N and P inputs to rivers by polygon according to Eq. 1 in the main manuscript.

Table S1. Overview of the model inputs to quantify nutrient inputs to rivers from point sources.

Model parameters*	Description	Original input scale	Transformation method to polygons	Final scale	Source
$\boldsymbol{T(E)ma_{dis}}$	Manure discharges to rivers (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{EF_{pnt_{ma,F}}}$	Fraction of N in anima lmanure entering water bodies as form F	National	Same national values for every polygon	Polygon	[1, 14-17]
$\boldsymbol{Frac_{farms\_county}}$	Fraction of animal farms in a county that are located in the polygon	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{Pop_{urb}}$	Urban population	County	Area weighed method	Polygon	[12, 13]
$\boldsymbol{Pop_{rur}}$	Rural population	County	Area weighed method	Polygon	[12, 13]
$\boldsymbol{hw_{frem,E}}$	Treatment efficiencies for N and P for WWTPs	Individual WWTP	-	Individual WWTP	See Section S2
$\boldsymbol{ConRate_{urb/rur}}$	Connection rate of urban population and rural population	National	Same national values for every WWTP	Individual WWTP	[25]

Table S1 (continued)

$\boldsymbol{CapacityWeigh_{WWT}}$	Fraction of the treatment capacity of the individual WWT to the total capacity of the WWTPs located in the county	Individual WWTP	-	Individual WWTP	See Section S3
$\boldsymbol{E(E)_{pnt}}$	The inputs of nutrient N or P to watersheds (land) resulted from human excrements in kg person^{-1} year^{-1}	Calculated according to	-	Polygon	[12, 13, 24]
GDPppp & GDPmer	Purchasing power parity based and market exchange rate based GDP in 1995 U.S. dollars person^{-1} year^{-1}	Province	Same value for WWTPs located in the same province	Individual WWTP	[26, 27]
$\boldsymbol{FE_{pnt_F}}$	Fraction of sewage effluents exported to rivers as nutrient form (F)	-	$\boldsymbol{FE_{pnt_{DIN}}}$ is directly proportional to the removal rate of N during treatment, while for other nutrient forms it is the calibrated constant as Global NEWS-2 (0.14, 1, and 0.01 for DON, DIP and DOP respectively).	Individual WWTP	[1, 9]

Figure S4. The locations of wastewater treatment plants (WWTPs) categorized by the daily average capacity (in 10 thousand m^3) and treatment efficiencies (%) for phosphorus. The examples are for coastal areas and urban area of Beijing. The treatment levels include low (P removal rate <= 40%) , medium (40% - 65%) and high (>=65%). The classification is derived from Van Drecht et al.[24].

S1.4 Diffuse sources

Diffuse sources (RS_{dif_F},kg year^{-1}, Eq. S8-S22) include explicit land-based sources (WS_{dif_F}, kg year^{-1}) which are partly retained by soil (FE_{ws_F}, 0-1) before entering water systems and parameterized export processes ($RSdif_{EC_F}$,kg year^{-1}, Eq. S23).

$$\boldsymbol{RS_{dif_F}} = (\boldsymbol{FE_{ws,F}} \cdot \boldsymbol{WS_{dif_F}} + \boldsymbol{RSdif_{EC_F}}) \quad \text{(Eq.S8)}$$

The **explicit land-based sources** are calculated as following:

$$\boldsymbol{WS_{dif_N}} = \boldsymbol{WSdif_{ant,N}} + \boldsymbol{WSdif_{nat,N}} \quad \text{(Eq.S9)}$$

$$\boldsymbol{WSdif_{nat,N}} = \boldsymbol{WSdif_{dep,nat,N}} + \boldsymbol{WSdif_{fix,nat,N}} \quad \text{(for DIN only)} \quad \text{(Eq.S10)}$$

$$\boldsymbol{WSdif_{ant,N}} = (\boldsymbol{WSdif_{fe,ant,N}} + \boldsymbol{WSdif_{ma,ant,N}} + \boldsymbol{WSdif_{dep,ant,N}} + \boldsymbol{WSdif_{fix,ant,N}} + \boldsymbol{WSdif_{hum,ant,N}}) - \boldsymbol{WSdif_{ex,N}} \quad \text{(for DIN and DON)} \quad \text{(Eq.S11)}$$

$$\boldsymbol{WSdif_{ant,P}} = (\boldsymbol{WSdif_{fe,ant,P}} + \boldsymbol{WSdif_{ma,ant,P}} + \boldsymbol{WSdif_{hum,ant,P}}) - \boldsymbol{WSdif_{ex,P}} \quad \text{(for DIP and DOP)} \quad \text{(Eq.S12)}$$

Where, $WSdif_F$ is the net N or P inputs to land (kg year^{-1}); subscript 'ant' and 'nat' refer to the inputs originate from agricultural area and natural area, respectively. $WSdif_F$ includes synthetic fertilizer use ($\boldsymbol{WSdif_{fe,ant,E}}$, kg year^{-1}), animal manure that is applied on land ($\boldsymbol{WSdif_{ma,ant,E}}$, kg year^{-1}), human waste that is applied on land from rural and urban population disconnected to sewage systems ($\boldsymbol{WSdif_{hum,ant,E}}$, kg year^{-1}), biological N_2 fixation ($\boldsymbol{WSdif_{fix,ant,N}}/\boldsymbol{WSdif_{fix,nat,N}}$, kg year^{-1}), and atmospheric N deposition ($\boldsymbol{WSdif_{dep,ant,N}}/\boldsymbol{WSdif_{dep,nat,N}}$, kg year^{-1}). $\boldsymbol{WSdif_{ex,E}}$ is the nutrient export via crop harvesting and animal grazing (kg year^{-1}). For DON, $\boldsymbol{WSdif_{dep,ant,N}} + \boldsymbol{WSdif_{fix,ant,N}} = 0$. The quantifications of $\boldsymbol{WSdif_{hum,ant,E}}$ are followed the method from MARINA 1.0[1] and we update the inputs for the contemporary year and with more detailed spatial levels (Table S2):

$$\boldsymbol{WSdif_{hum,ant,E}} = \boldsymbol{WSdif_{hum,urb,E}} + \boldsymbol{WSdif_{hum,rur,E}} \quad \text{(Eq.S13)}$$

$$\boldsymbol{WSdif_{hum,urb,E}} = \boldsymbol{E(E)_{hum}} * \boldsymbol{Popuncon_{urb}} * \boldsymbol{F}_{urb\_unc\_land} \quad \text{(Eq.S14)}$$

$$\boldsymbol{WSdif_{hum,rur,E}} = \boldsymbol{E(E)_{hum}} * \boldsymbol{Popuncon_{urb}} * \boldsymbol{F}_{rur\_unc\_land} \quad \text{(Eq.S15)}$$

$$\boldsymbol{Popuncon_{urb}} = \boldsymbol{Pop_{urb}} * (\boldsymbol{1} - \boldsymbol{ConRate_{urb}}) \quad \text{(Eq.S16)}$$

$$\boldsymbol{Popuncon_{rur}} = \boldsymbol{Pop_{urb}} * (\boldsymbol{1} - \boldsymbol{ConRate_{rur}}) \quad \text{(Eq.S17)}$$

Where, $\boldsymbol{WSdif_{hum,ant,E}}$ refers to the human waste of N and P applied on land from urban ($\boldsymbol{WSdif_{hum,urb,E}}$, **kg**) and rural ($\boldsymbol{WSdif_{hum,rur,E}}$,**kg**) population disconnected to sewage systems. $\boldsymbol{E(E)_{hum}}$

is the human excretion of N and P calculated following Van Drecht et al.[16]. $\boldsymbol{Popuncon_{urb}}$ and $\boldsymbol{Popuncon_{rur}}$ are the urban and rural population that is not connected to WWTPs. $F_{urb\_unc\_land}$ and $F_{rur\_unc\_land}$ are the fractions of human excretion that are applied on land from urban and rural population disconnected to sewage systems, respectively (0-1) (Table S2). $\boldsymbol{WSdif_{hum,ant,E}}$ is quantified on the county scale and use the **area weighted method** (see below) to distribute the county values to the polygon scale.

For most of the explicit land-based sources (Table S2), we use the land-use weighted method to distribute the county inputs to polygons. The locations of arable land from the land-use data (1×1km grid) are used. The steps for this method are similar with the land-use weighted method for manure farms. Here, to allocate county inputs, we use the maps of the arable land (1×1km grid).

For other explicit land-based sources, the **area weighted method** is used to distribute county inputs to polygons as follows:

1. In the area weighted method, we first use the Arc-GIS tool to calculate the area fraction of a polygon that is located in a certain county or (Figure S5) or grid.
2. Then, we distribute the inputs on the county level to polygons according to the associated fractions from step 1. An example is given for atmospheric N deposition below (Eq.S18-S19). The same principle is applied to transform other county values to polygons.

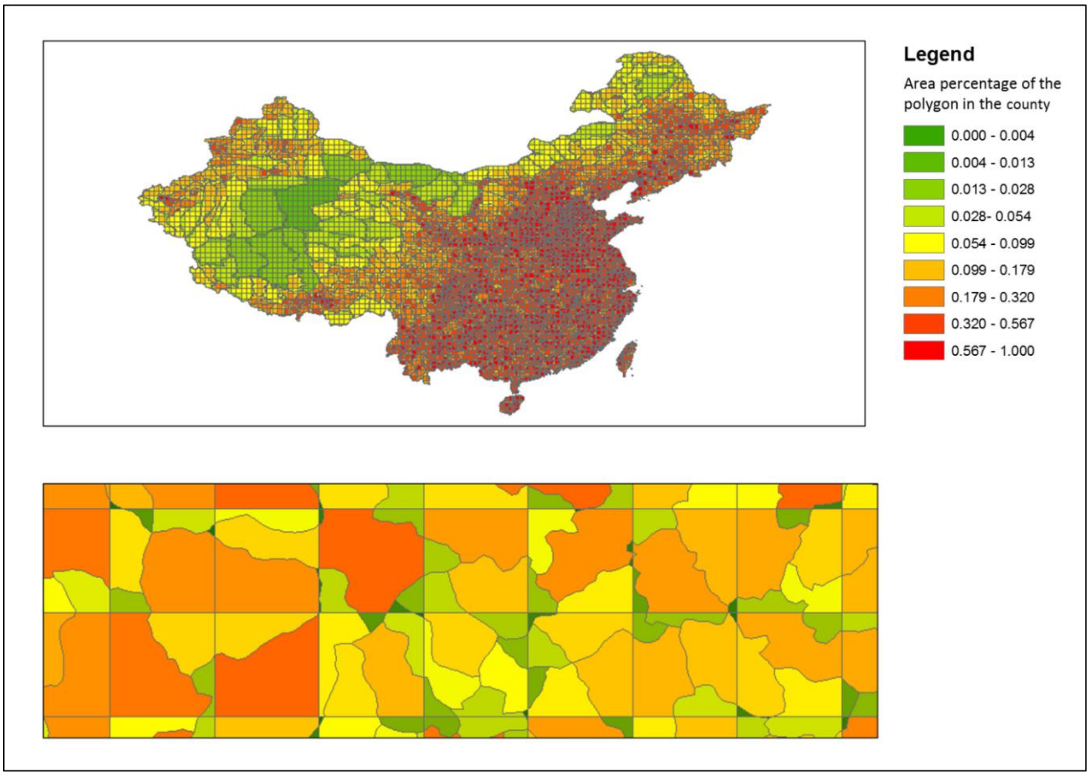

Figure S5. Intersected polygon units and the fraction of their areas in counties.

Biological N_2 fixation by natural vegetation ($\boldsymbol{WSdif_{fix,nat,N}}$) from grid to polygon is calculated as:

$$WSdif_{fix,nat,N,1} = Nfix\_nat_a * Fr_{1\_a} \tag{Eq.S18}$$

$$Fr_{1\_a} = \frac{Area_1}{Area_a} \tag{Eq.S19}$$

Where, the subtitle of the letters (e.g. a) refers to the county or grid (in this example the original input is on the grid, see Table S2). The subtitle of the numbers (e.g. 1) refers to the polygon. $WSdif_{fix,nat,N,1}$ refers to the biological N_2 fixation by natural vegetation in polygon 1 (kg $year^{-1}$). $Nfix\_nat_a$ is biological N_2 fixation (kg $year^{-1}$) in natural areas in a certain grid (kg $year^{-1}$). $Fr_{1\_a}r$ refers to the fraction of the area in polygon 1 and in grid a which is located in that polygon. $Area_1$ and $Area_a$ are the total areas of polygon 1 or grid a in this example. Above equations are the examples of the area-weighted method for $Nfix\_nat_a$. This input in (Eq.S18) can be replaced with other inputs provided on the county or grid scales.

$$\boldsymbol{FE_{ws,F}} = f_F(\text{Rnat}) * \text{eF} \tag{Eq.S20}$$

$$f_F(\text{Rnat}) = \text{Rnat}^{aF} \qquad \text{for DIN, DON and DOP} \tag{Eq.S21}$$

$$f_F(\text{Rnat}) = \frac{1}{(1+\frac{\text{Rnat}}{aF})^{-bF}} \quad \text{for DIP} \qquad \text{(Eq.S22)}$$

Where, $\boldsymbol{FE_{ws,F}}$ is the export fraction of nutrient form (F) entering rivers (0-1). The $FE_{ws,F}$ is calculated as a function of the total runoff ($Rnat$, m^3 $year^{-1}$) on the polygon scale. The total runoff is the average natural total runoff from 1970 to 2000 on the 0.5°x0.5° grid scale from VIC (Table S2). We apply the same value for each polygon located in the grid. The calibrated coefficients in the function (eF, aF, bF) are directly from the Global NEWS-2 model[9] that are also applied in MARINA 1.0[1] for China.

The weathering of P-contained minerals (for DIP), and leaching of organic matter from agricultural and non-agricultural soils (for DON and DOP) are calculated according to the export-coefficient approach according to Global NEWS-2[9] for each polygon. This approach is also applied in MARINA 1.0[1] for China.

The **parameterized export processes** ($RSdif_{EC_F}$,kg $year^{-1}$) includes the weathering of P-contained minerals (for DIP), and leaching of organic matter (for DON and DOP) and are calculated by the export-coefficient approach of MARINA 1.0 for polygons. For DIP, DON and DOP:

$$\boldsymbol{RSdif_{EC_F}} = f_F(\text{Rnat}) * \text{EC}_F \qquad \text{(Eq.S23)}$$

Where, $f_F(\text{Rnat})$ is identical to the $f_F(\text{Rnat})$ runoff function used in modeling the $\boldsymbol{FE_{ws,F}}$ terms (see Eq.S21-S22). EC_F is the calibrated constant of nutrient form (F).

Table S2. Overview of the model inputs to quantify nutrient inputs to rivers from diffuse sources and from 'others' sources.

Abbreviation of model inputs*	Description	Original spatial scale	Method to transform model inputs to polygons	Spatial scale of this study	Reference
Explicit land-baed sources					
$\boldsymbol{WSdif_{fe,ant,E}}$	Synthetic fertilizers applied on land (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{WSdif_{ma,ant,E}}$	Animal manure applied on land (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{WSdif_{dep,ant,E}}$	Atmospheric N deposition** on agricultural areas (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13, 28, 29]
$\boldsymbol{WSdif_{fix,ant,E}}$	Biological N_2-fixation by agricultural crops (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{WSdif_{ex,E}}$	Nutrient export via crop harvesting and animal grazing (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13]
$\boldsymbol{WSdif_{hum,ant,E}}$	Human excreta applied on land from unconnected population to sewage (kg year^{-1})	County	Area weighted method	Polygon	NA***
$\boldsymbol{WSdif_{dep,nat,N}}$	Atmospheric N deposition** on natural areas (kg year^{-1})	County	Land-use weighted method	Polygon	[12, 13, 28, 29]
$\boldsymbol{WSdif_{fix,nat,N}}$	Biological N_2-fixation from natural area (kg year^{-1})	Grid	Area weighted method	Polygon	[9, 30]

Table S2 (Continued)

$\boldsymbol{F_{urb\_unc\_land}}$	Fraction of human excretion applied on land from urban population that is not connected to sewage	National	National value is applied (12.6% for N and P) to all polygons	Polygon	[1, 12, 13]
$\boldsymbol{F_{rur\_unc\_land}}$	Fraction of human excretion applied on land from rural population that is not connected to sewage	National	National value is applied (53% for N and P) to all polygons	Polygon	[1, 2, 31]
$\boldsymbol{Rnat}$	Total runoff from land to streams (m^3 $year^{-1}$)	Grid	The same grid values are applied for polygons that are located within the grids	Polygon	[109, 110]
$\boldsymbol{eF}$	Model coefficients to quantify $\boldsymbol{FE_{ws,F}}$	Basin	The same values are applied for all polygons following the Global NEWS-2 and MARINA 1.0 approaches (0.94, 0.010, 0.29 and 0.010 for DIN, DON, DIP and DOP, respectively).	Polygon	[9]
$\boldsymbol{aF}$	Model coefficients to quantify $\boldsymbol{FE_{ws,F}}$	Basin	The same values are applied to all polygons following the Global NEWS-2 and MARINA 1.0 approaches (1, 0.95, 0.85 and 0.95 for DIN, DON, DIP and DOP, respectively).	Polygon	[9]
$\boldsymbol{bF}$	Model coefficients to quantify $\boldsymbol{FE_{ws,F}}$	Basin	The same values are applied for all polygons following the Global NEWS-2 and MARINA 1.0 approaches (2 for DIP).	Polygon	[9]

Table S2 (Continued)

Parameterized export processes					
EC_F	Calibrated model coefficients to quantify $\boldsymbol{RSdif}_{EC_F}$	Basin	The same values are applied to all polygons following the Global NEWS-2 and MARINA 1.0 approaches (1, 0.95, 0.85 and 0.95 for DIN, DON, DIP and DOP, respectively).	Polygon	9
Others					
$\boldsymbol{EF}_{pnt_{hum\_uncon,F}}$	Fraction of element E entering rivers as form F	National	The same values are applied for all polygons following the Global NEWS-2 and MARINA 1.0 approaches (0.7 for DIN and DIP; 0.3 for DON and DOP).	Polygon	1, 9
$\boldsymbol{F}_{\boldsymbol{urb\_unc\_dis}}$	Fraction of human excretion that is directly discharged to rivers (untreated) from urban population that is not connected to sewage	National	National values are applied (63.4% and 87.4% for N and P) to all polygons	Polygon	1, 2, 31, 32
$\boldsymbol{F}_{\boldsymbol{rur\_unc\_dis}}$	Fraction of human excretion that is directly discharged to rivers (untreated) from rural population that is not connected to sewage	National	National values are applied (23% and 47% for N and P) to all polygons	Polygon	1, 2, 31
$\boldsymbol{Popuncon}_{\boldsymbol{urb}}$	Urban population that is not connected to WWTPs	County	Area weighed method	Polygon	12, 13

Table S2 (Continued)

$\boldsymbol{Popuncon_{rur}}$	Rural population that is not connected to WWTPs	County	Area weighed method	Polygon	[12, 13]

*Subscript E refers to nutrient element N or P. **Atmospheric N deposition includes both wet and dry deposition. Based on Xu el al[29] and Wang et al[12], the dry and wet N deposition rates are derived for the year 2012 (national average is 18.7 and 18.2 kg/ha, respectively) in China. *** It results from the calculation of different parameters.

S1.5 Others

Human waste that is directly discharged (untreated) to rivers from population without sewage connections (RS_{others_F}) is quantified following MARINA 1.0[1]. These sources are categorized as 'others' because they are directly discharges to nearby water systems and not retained from soil and nor emitted in a concentrate way like WWTPs. We quantify direct discharges of human waste to rivers for the year 2012 using the county inputs as:

$$RS_{others_F} = RS_{others\_urb\_unc,F} + RS_{others\_rur\_unc,F} \quad \text{(Eq.S24)}$$

$$RS_{others\_urb,F} = \boldsymbol{E(E)_{hum}} \cdot \boldsymbol{Popuncon_{urb}} \cdot \boldsymbol{F}_{urb\_unc\_dis} \cdot \boldsymbol{EF}_{pnt_{hum\_uncon,F}} \quad \text{(Eq.S25)}$$

$$RS_{others\_rur,F} = \boldsymbol{E(E)_{hum}} \cdot \boldsymbol{Popuncon_{rur}} \cdot \boldsymbol{F}_{rur\_unc\_dis} \cdot \boldsymbol{EF}_{pnt_{hum\_uncon,F}} \quad \text{(Eq.S26)}$$

Where, $RS_{others\_urb\_unc,F}$ and $RS_{others\_rur\_unc,F}$ refer to the human waste that is directly discharged to rivers (untreated) from urban and rural population without sewage connections (kg year^{-1}). $\boldsymbol{E(E)_{hum}}$, $\boldsymbol{Popuncon_{urb}}$ and $\boldsymbol{Popuncon_{rur}}$ are described in the Eq.S14-17. $F_{urb\_unc\_dis}$ and $F_{rur\_unc\_dis}$ are the fractions of direct discharges of human waste to rivers from urban and rural population that is not connected to sewage systems, respectively (Table S2). $EF_{pnt_{hum\_uncon,F}}$ is the fraction of element E (N or P) entering rivers as form F (DIN, DON, DIP, DOP) (0-1).

S1.6 Model outputs at multiple scales

Our model is able to generate model outputs at multiple scales. This implies that every source and related processes are pre-processed and quantified on its most appropriate scales. Polygons serve as a storing carrier for the information and finally as a bridge to generate model results at multiple scales.

Total inputs of N and P to rivers are quantified by Eq.1 in the main text. Inputs of N and P to rivers from all source categories (point, diffuse and others) are stored in the polygons and aggregated according to Eq.1 on the polygon scale. For the total inputs of N and P to rivers, the polygon-scale results can be aggregated to the grid, sub-basin or county according to the spatial relation. We do this using ID codes of polygons. These codes are unique for polygons and are related to counties and grids. In this way, we can identify which polygons belong to which grid or county. For example, we would like to aggregate polygon results to counties (e.g., for local policy-makers). In this situation, we identify polygons with the same county ID as for polygons. Then, we aggregate the results of the polygons with the same county ID to get the county scale results.

For the **source attribution** on different scales, the different sources are first aggregated to the associated spatial levels and follow the principle as described below. The share of inputs of N and P to rivers from each source is quantified by the ratio of each river source term (RS, e.g. $\boldsymbol{RS_{pnt\_ma,F}}$ ***or*** $\boldsymbol{RS_{dif_F}}$) in the equations above and the total inputs to rivers ($\boldsymbol{RStotal_F}$).

For a detailed source attribution of explicit land-based sources from diffuse sources on agricultural land (i.e. level 2 of the source attribution in Global NEWS-2), the special fraction G_F is applied:

$$G_F = 1 - \frac{WSdif_{ex,E}}{WSdif_{gross,F}} \qquad \text{(Eq.S27)}$$

Where, $WSdif_{ex,E}$ is the nutrient export of element E (N or P) via crop harvesting and animal grazing (kg year^{-1}). $WSdif_{gross,F}$ is the sum of inputs of N or P to agricultural land from all sources (kg year^{-1}). For DIN, these sources are atmospheric N deposition, biological N_2 fixation by crops (kg year^{-1}), N synthetic fertilizers (kg year^{-1}), N animal manure (kg year^{-1}) and N human waste (kg year^{-1}). For DON, DIP and DOP, these sources are P synthetic fertilizers (kg year^{-1}), P animal manure (kg year^{-1}) and P human waste (kg year^{-1}). For example, the share of synthetic fertilizer inputs of DIN in rivers to the total inputs of DIN from all sources is calculated as:

$$Share_{DIN\_src\_fe} = \frac{\boldsymbol{WSdif_{fe,ant,N}} \cdot G_{DIN} \cdot FE_{ws,DIN}}{RStotal_{DIN}} \quad \text{(Eq.S28)}$$

Where, $Share_{DIN\_src\_fe}$ is the share of synthetic fertilizers that are applied on land to the total inputs of DIN to rivers (0-1). $\boldsymbol{WSdif_{fe,ant,N}}$ is the N synthetic fertilizers that are applied on land (kg year^{-1}). G_{DIN}, $FE_{ws,DIN}$, $RStotal_{DIN}$ are described above.

For the export-coefficient based diffuse sources (i.e. DIP inputs to rivers from weathering and DON and DOP inputs to rivers from leaching of organic matter ($\boldsymbol{RSdif_{ec,F}}$ **kg year^{-1}**)), they are calculated as the same principle for other source categories, i.e. using directly the river source term ($\boldsymbol{RSdif_{ec,F}}$) divided the total inputs to rivers ($\boldsymbol{RStotal_{F}}$) . See details in Global NEWS-2[9].

S2 Treatment efficiencies of wastewater treatment technologies

Table S3. Overview of the treatment efficiencies of the different wastewater treatment technologies to remove total nitrogen (TN) and total phosphorus (TP) in wastewater based on the existing studies (removal rates are in %). WWTPs are the wastewater treatment plants.

Treatment technologies	Ranges for TN removal rates among individual WWTPs (0-1)	TN removal rates (0-1)**	Ranges for TP removal rates among individual WWTPs (0-1)	TP removal rates (0-1)*	Reference
Biological Speed Seperation Filter	75.2%	75%	30%~40%	35	33-35
Ultrafiltration (UF) / Reverse Osmosis (RO)	76%~95%	80%	86%~95%	90%	36-40
Combined process of constructed wetland and ecological pond	21.9%~69.7%	45%	43.33%~95.5%	60%	41
Biolak	34%~73%	55%	80%~90%	85%	42, 43
Biofilm	33%~57.4%	45%	71.8%~99%	80%	44, 45
Bio-filter	82%~91%	70%	41.2%~92%	45%	46
Biological fluidized bed	56.35%~72.05%	65%	28.96%~36.98%	30%	47
Biological contact oxidation process	50%~80%	65%	37.52%~89.06%	60%	48-50
Biological treatment + UF/RO	90%	90%	98%	95%	37, 51, 52
Biological treatment + Bio-filter	NA*	80%	NA*	90%	44, 53
Biological treatment	45%~75.4%	50%	51%~64.5%	60%	24, 53
Active sludge process	10%~57%	30%	30%~55%	45%	54-56
Active sludge process + Advanced oxidation Process	NA*	80%	NA*	90%	39, 40, 57
Hrdrolysis Acidification + A/O	65.3%~72.3%	70%	57.5%~86.4%	75%	58
Oxidation Ditch+ Bio-filter	NA*	80%	NA*	90%	39, 40
Oxidation Ditch	30%~40%	35%	35%~50%	45%	39, 40, 59
Modified Oxidation Ditch	55%~90%	55%	55%~75%	65%	39, 40, 59
Modified Orbal Oxidation Ditch	59.5%~93%	70%	71.2%~78.8%	75%	60, 61
Modified Carrousel Oxidation Ditch	38.77%~91.9%	60%	71.37%~94%	85%	62-64
Improved/modified SBR	69.6%~94.3%	75%	77.4%~96.4%	80%	65-67
Modified CASS	52.6%	50%	78.4%	80%	68, 69
Modified A²/O	56%~90%	75%	67%~90%	80%	70-73
Fine Bubble-aerated Oxidation Ditch	43%~50%	45%	62.5%~80%	65%	74-76
Bardenpho process	72.2%~89%	80%	91.7%~93%	90%	77, 78
Orbal Oxidation Ditch	60.5%~90%	70%	73.8%	75%	79, 80
Reverse Osmosis (RO)	90%	90%	98%	95%	39, 40, 81, 82
Carrousel Oxidation Ditch	50%~69.3%	55%	75%~77%	75%	83-86
Chemical and biological treatment	NA*	80%	NA*	90%	39, 40
Secondary biological treatment	NA*	45%	NA*	55%	39, 40
UASB+CASS	NA*	80%	NA*	90%	39, 40, 87
SBR	47.8%~75%	60%	42%~95.3%	80%	65, 88, 89
MSBR+ABF	44.55%~80%	70%	89.7%~94.3%	90%	39, 40, 90

Table S3 (continued)

MSBR	78.6%~89%	85%	90%~93.8%	90%	91, 92
MBR	75%~85%	80%	89%~90%	90%	93, 94
MBBR	46.3%~48.2%	45%	65.5%~79.2%	70%	95-97
HyBAS + Advanced treatment	NA*	80%	NA*	90%	39, 40
DE Oxidation Ditch	63.6%~70.72%	65%	82.73%~93.4%	85%	98, 99
CAST	20.7%~80%	50%	80%~84.5%	80%	67, 100, 101
CASS	51.9%~55.1%	50%	85.7%~91.4%	80%	102-104
BAF	30%~90%	40%	46.5%~87.9%	45%	105, 106
A^2/O+Biofilm	55%	55%	87.89%	85%	107
A^2/O+MBR	76%	75%	69.81%~95%	80%	108, 109
A^2/O	40%~83.2%	55%	72%~97%	75%	48, 56, 70, 73, 110, 111
A/O+MBR	48.5%~77%	55%	58%~90%	80%	112-115
A/O	20%~66%	45%	62%~98%	65%	56, 116, 117

*: For some combinations of the technologies which lack of data, we derived the removal rate from the study of Van Drecht et al[24]: the removal rates of N and P during treatment of the wastewater are categorized into three general categories depending on technologies: primary (average 10% removal rate for both N and P), secondary (35% for N and 45% for P) and tertiary (80% for N and 90% for P).

**: The removal rate is the average of the values from the literature studies for each technology. When the range of the values is large, we exclude the extremes (outliers) and calculate the average of the values without extremes. The average numbers are rounded off.

S3 Centralized wastewater treatment plants (WWTPs) database

Table S4. Description of the variables (column names) that are used in the newly created wastewater treatment plants (WWTPs) database. The database can be found in the 'FinalWWTPdatabase.xlsx' file in the supplementary information. N and P are nitrogen and phosphorus, respectively.

Column names	Description	Source
FID	ID for each WWTP	-
OBJECTID	ID for each county where individual WWTPs are located	Processed*;[13]
County	County name in Chinese	Processed*;[13]
City	City name in Chinese	Processed*;[13]
Province	Province name in Chinese	Processed*;[13]
Organization code	Organization code of an enterprise that operates the WWTPs that are registered in the State Administration for Industry and Commerce of the People's Republic of China	[21]
ProjectName	Project name of WWTPs in Chinese	[20, 21]
Longitude	Longitude	This study**
Latitude	Latitude	This study **
Address	Address in Chinese	Processed***;[20, 21]
Capacity	Average treatment capacity (10^4 m^3/day)	[20]
Nrevoval	Removal rate for N (0-1)	See Table S3
Premoval	Removal rate for P (0-1)	See Table S3
Treatment technology	Treatment technology	[20]
Treatment rate for urban	The fraction of the wastewater treated in the total wastewater transported to the urban WWTPs ($Frac_{treat}$)	Processed*;[22, 23]
Treatment rate for rural	The percentage of the wastewater treated in the total wastewater transported to the rural WWTPs ($Frac_{treat}$)	Processed*;[22, 23]
DIP	Inputs of dissolved inorganic phosphorus to rivers from WWTPs (in kton $year^{-1}$)	Modelled****
DIPrm	Dissolved inorganic phosphorus that is removed from WWTPs during treatment (in kton $year^{-1}$)	Modelled****
DOP	Inputs of dissolved organic phosphorus to rivers from WWTPs (in kton $year^{-1}$)	Modelled****
DOPrm	Dissolved organic phosphorus that is removed from WWTPs during treatment (in kton $year^{-1}$)	Modelled****
DIN	Inputs of dissolved inorganic nitrogen to rivers from WWTPs (in kton $year^{-1}$)	Modelled****
DINrm	Dissolved inorganic nitrogen that is removed from WWTPs during treatment (in kton $year^{-1}$)	Modelled****
DON	Inputs of dissolved organic nitrogen to rivers from WWTPs (in kton $year^{-1}$)	Modelled****
DONrm	Dissolved organic nitrogen that is removed from WWTPs during treatment (in kton $year^{-1}$)	Modelled****

*: Linked with the associated sources via spatial locations of WWTPs; **: The original lists of WWTPs include the name of WWTPs in Chinese and we need to first search for the address of 4000s WWTPs and locate the exact latitude and longitude according to the address. ***: Some addresses are not easily available. Thus we obtained the addresses according to the information that was possible to find with the project name. ****: Model outputs; interpret with caution and contact the author.

S4 Model evaluation

This section is an extended description of the model evaluation in the manuscript. We evaluated our model by (1) model inputs, (2) modeling approaches, (3) model parameters and (4) comparisons with others. The model uncertainties (5) are associated with model inputs, modeling approaches and model parameters. We recognize the uncertainties and discuss them in the end of this section.

S4.1 Model inputs

The NUFER and VIC models provide important model inputs for this study. These models were evaluated and are widely used in existing studies. NUFER was developed to quantify the N and P flows and losses in the food chain in China and was applied on national, provincial and county scales[12, 118]. VIC has been validated using daily grid-based (0.5°x0.5°) observed records of the streamflow for 1,557 river monitoring stations worldwide from the Global Runoff Data Centre. A realistic representation of the observed conditions was found with a normalized bias in the simulated streamflow of less than ┃0.25┃for half of the stations worldwide [10, 11]. The statistical year books for the Chinese counties are known as the most reliable data source in China and used widely in existing studies[12, 18]. In this study, the unique WWTPs database is created based on the official government documents, exact locations of WWTPs (longitude and latitude), literature reviews and expert knowledge (see section S3). We use the land-use information that is at 1km x 1km for the contemporary year 2010 from the Data Center for Resources and Environmental Sciences[18]. We consider our model inputs reliable and of the satisfying quality considering the fact that the data sources provide most recent and complete information that represents local characteristics.

S4.2 Modeling approaches

Table S4 presents a comparison of our modeling approach with the approaches of the selected existing nutrient models. The selected models cover the main basin areas in China (see Figure S1) and are applied at different scales (e.g. sub-basin, grid). Our modeling approaches for diffuse sources are largely based on the modeling approaches of Global NEWS-2 and MARINA 1.0. The nutrient balances on land are first quantified and multiplied with the parameter presenting the nutrient retentions in soils. This structure to quantify the nutrient inputs from diffuse sources to rivers is also adopted in the IMAGE-GNM model. In our study, we improve the modeling approaches, in particular, for point sources.

Table S5. Comparison of the main characteristics between our model and the selected nutrient models.

Main characteristics	Global NEWS-2[9]	MARINA 1.0[1, 119]	IMAGE-GNM[120]	SWAT[121, 122]	The model of this study
Spatial scale					
Calculation unit	Basin	Sub-basin	Grid (0.5° × 0.5°)	Sub-basin /Hydrological response unit	Multiple scales: polygon/Grid (0.5° × 0.5°) /Sub-basin /County
Spatial extent of model outputs	World	China	World	Multiple regions	China
Point source manure discharges*					
Manure discharges to rivers as a point source	Not included	Included	Not included	Not included	Included
Locations of animal farms	No location specified	No location specified	Not location specified	No location specified	Location specified
Point source of human waste from WWTPs					
Locations of WWTPs	No locations	No locations	Modelled distribution*	Locations are included	Locations are included
Measurement data requirement for modeling	No measurement data required	No measurement data required	No measurement data required	Required measurements	No measurement data required
Model inputs for diffuse sources					
Source	Mostly national statistics	Mostly national statistics	Mostly national statistics	Regional/ catchment data	Mostly county statistics
Spatial resolution of model inputs	Grid data (distributed according to national values) and aggregated to basins	Grid data (distributed according to national and provincial values) and aggregated to sub-basins	Gridded data (distributed according to national values)	Catchment scale	Modelled polygon data (distributed according to county values)
Model outputs					
Nutrient forms	Dissolved inorganic and organic forms for N and P; Particulate N and P	Dissolved inorganic and organic forms for N and P	Total N and total P	Nitrate, organic N; Soluble P, organic P	Dissolved inorganic and organic forms for N and P
Nutrients inputs to rivers/to sea	to rivers & to sea	to rivers & to sea	to rivers & to sea	to rivers & to sea	currently only to rivers
Computational requirements					
Level	Low	Low	Medium	Medium	Medium

*: The model allocates the national population of China that is connected to sewage over grids. This is done by ranking, starting from a grid with highest total population density until the total population equals to the population connected to sewage; see details in Morée et al.[32].

S4.3 Model parameters

We updated most of the model parameters for point sources and diffuse sources with the contemporary information for more detailed spatial levels (see details in section S1). Some parameters taken from the MARINA 1.0 model are from expert opinion and literature. The rest of model parameters are directly taken from the Global NEWS-2 model. We perform a sensitivity analysis to quantify the uncertainties in important model parameters.

We perform the sensitivity analysis by adopting the local one-factor-at-a-time (OAT) method[123, 124]. We select this method considering the computational costs and the relatively simple relationship among parameters and inputs. We use the elementary effects (EE) defined by Moris[123] as an indicator to present the level of the sensitivity of how the model responses to changes in the selected parameters and inputs as:

$$S = \frac{\frac{\boldsymbol{M}(\boldsymbol{e_1}, \cdots, \boldsymbol{e_i} + \Delta\boldsymbol{e_i}, \cdots, \boldsymbol{e_p}) - \boldsymbol{M}(\boldsymbol{e_1}, \cdots, \boldsymbol{e_i}, \cdots, \boldsymbol{e_p})}{\boldsymbol{M}(\boldsymbol{e_1}, \cdots, \boldsymbol{e_i}, \cdots, \boldsymbol{e_p})}}{\frac{\Delta\boldsymbol{e_i}}{\boldsymbol{e_i}}} \quad \text{(Eq.S29)}$$

Where, M is the model output, $\boldsymbol{e_i}$ refers to the different model parameters, and $\Delta\boldsymbol{e_i}$ refers to the perturbation in the selected single model parameter. We selected important model parameters and inputs covering both point and diffuse sources. These parameters include: runoff, synthetic fertilizer applied on land, animal manure applied on land, direct discharges of manure, connection rates of urban and rural population to WWTPs, treatment rates of WWTPs, treatment efficiencies for N and P and locations of manure farms.

The selected parameters (except for locations of manure farms) are increased and decreased by 10% and 50% and the average EE is calculated for each parameter in order to evaluate the response of model outputs (i.e. the nutrient inputs to rivers by nutrient forms) to changes in model inputs and parameters (Table S6). We design another setting for quantifying the elementary effects of locations for animal farms. We randomly select 20% of the polygons and increase the percentage of the animal farms by 10% of those polygons and quantify the elementary effect of the selected polygons on nutrient inputs to rivers by nutrient forms. Table S6 shows the range of the elementary effects and their average EEs for each nutrient form.

The results of the sensitivity analysis indicate that the model is relatively robust. The average EEs of the selected parameters and inputs are less than 1. This implies that 10% of the perturbation in the model parameters and inputs results in changes in the model outputs (nutrient inputs to rivers) by less than 10%. This percentage in most cases is substantially smaller. The model outputs are aggregated on the national level to calculate the EEs. And for each polygon, the sensitivity (i.e. the EE) varies (e.g. for locations of manure farms), depending on the dominant source of nutrients in rivers of the polygons.

Table S6. The results of the sensitivity analysis for selected model parameters and inputs.

Parameter and input	Description	Average elementary effects (EE) (0-1)			
		DIN	DON	DIP	DOP
$\boldsymbol{R_{nat}}$	Runoff	71.6%	22.3%	17.2%	6.3%
$\boldsymbol{WSdif_{fe,ant,E}}$	Fertilizer applied on land	59.5%	4.3%	15.6%	3.2%
$\boldsymbol{WSdif_{ma,ant,E}}$	Manure applied on land	9.6%	0.7%	5.0%	1.0%
$\boldsymbol{T(E)ma_{dis}}$	Manure discharges to rivers	19.6%	55.7%	64.2%	76.4%
$\boldsymbol{ConRate_{urb/rur}}$	Connection rates of urban and rural population	1.6%	8.9%	3.9%	11.1%
$\boldsymbol{Frac_{treat}}$	Treatment rates of WWTPs	1.6%	8.9%	3.9%	11.1%
$\boldsymbol{hw_{frem,N}}$	Treatment efficiencies for N	2.6%	4.7%	0.0%	0.0%
$\boldsymbol{hw_{frem,P}}$	Treatment efficiencies for P	0.0%	0.0%	15.2%	0.4%
-	Locations of manure farms	25.6% (0% ~ 99.0%)	37.4% (0% ~97.9%)	43.3% (0%~98.9%)	47.3% (0%~97.7%)

S4.4 Comparison of model outputs with other modeling studies

In this section, we compare our results with the selected models and other studies for main basins in China (Figure S6, Table S8). Below, we discuss the differences and similarities between our results and other of these existing models and studies.

Comparisons with the selected models

The selected models cover the main basin areas in China (Figure S1, Figure S6) at different scales (e.g. sub-basin, grid) and with different modeling approaches (Table S5). We compare our estimates for 2012 with the results of the MARINA 1.0 [1, 119] and IMAGE-GNM models for the year 2000 for the main basin areas in China[120, 125] and for year 2010 for Changjiang[126]

(Table S8). We model 21.9 Tg of the total dissolved nitrogen (TDN) and 2.2 Tg of the total dissolved phosphorus (TDP) emitted to the Chinese rivers in 2012. Point sources are responsible for 71% of TDP inputs to rivers and 21% of TDN (Figure S6).

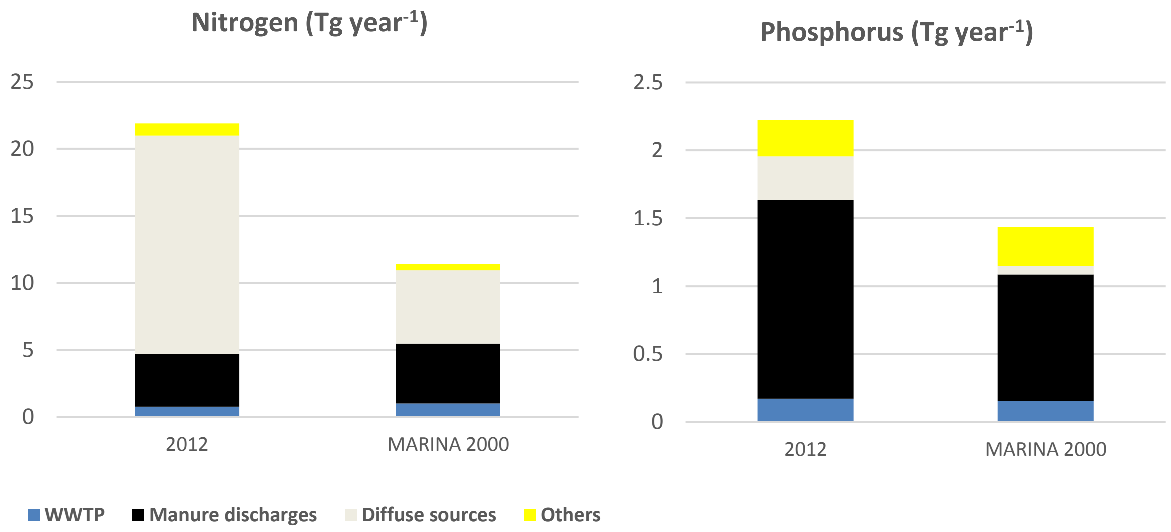

Figure S6. Nitrogen (N) and phosphorus (P) inputs to rivers and their source attribution according to this study in 2012 and according to MARINA 1.0 in 2000 (Tg year^{-1}). MARINA 1.0 is short for the Model to Assess River Inputs of Nutrients to seAs[1]. This study and MARINA 1.0 present inputs of the total dissolved nitrogen and total dissolved phosphorous from point sources (human waste from WWTPs, direct discharges of animal manure to rivers), diffuse sources (synthetic fertilizer use, manure applied on land, biological N_2 fixation by agricultural crops and by natural vegetation, atmospheric N deposition, leaching of organic matter, and weathering of P-contained minerals from agricultural and non-agricultural soils, human waste that stays on land) and 'Others' sources (human waste from urban and rural population that is not connected to sewage systems, but discharged directly to rivers).

Compared to model results of 2000 from the MARINA 1.0 model, TDN increased by 92% and TDP increased by 52% by 2012. The increase for TDN is higher than for TDP because of the different increase rates for diffuse sources and point sources. Diffuse sources dominate in TDN inputs to rivers while point sources dominate in TDP. For diffuse sources, the increase is the combined effect of an increase in nutrient inputs to rivers from land activities (e.g. fertilizer applied) and a decrease in N retention in soils (i.e. increased $\boldsymbol{FE_{ws,F}}$ for N). Point sources account for 48% of TDN inputs and 76% of TDP inputs in 2000. The manure N discharges decreased from 4.46 to 3.91 Tg (14%), while manure P discharges increased from 0.93 to 1.46 Tg (57%) between 2000 (MARINA 1.0) and 2012 (our model). This difference is the combined effect of (1) the lower discharge rate of manure in 2012 compared to 2000 and (2) different N and P excretion rates to estimate the animal manure excretion between the NUFER model (provided for us manure discharges data[12]) and the Integrated Model to Assess the Global

Environment (IMAGE, provided the manure excretion data for the MARINA 1.0 and Global NEWS models[30]).

Results of IMAGE-GNM for the main basins in China (Figure S1) for the year 2000[120, 125] are closed to MARINA 1.0 and are generally lower compared to our study for the year 2012. IMAGE-GNM results are the total N and total P inputs to rivers while we model different forms of N and P. IMAGE-GNM includes diffuse sources (fertilizers, manure applied on land, biological N_2 fixation, atmospheric N deposition) via surface runoff, shallow and deep groundwater, soil erosion, N and P deliveries from flooded areas and point sources (human waste discharged from WWTPs). Additional sources in IMAGE-GNM are P weathering, wastewater from aquaculture, allochthonous organic matter, N deposition on water bodies. The recent model estimates from IMAGE-GNM by Liu et al.[126] for the year 2010 are lower than ours (Table S8); they modelled 8148 kton of TN (vs 11627 kton of TDN in this study) and 729 kton of TP (vs 1077 kton of TDP in this study). For N, both models agree that diffuse sources dominate. Since the share of particular N is small[127, 128] and the modelled soil retentions are comparable, the differences mainly result from N nutrient budget (our 19417 vs 14200 kton in IMAGE-GNM). This is mainly due to different input sources (county vs provincial statistics, NUFER vs IMAGE). Moreover, the direct manure discharges have the limited share in the total inputs. Therefore neglecting this source has a limited influence on the total N inputs. For P, particulate P could be up to 40% of TP[128]. We consider this difference could result in neglecting manure discharges in their estimates (Table S8) because manure discharges are dominant sources of P.

Our results are higher than in the MARINA 1.0[1, 120] and IMAGE-GNM[120, 125] models because we calculate for 2012 whereas the other models for 2000. Table S7 summarizes the nutrients loads to coastal seas for the main basins based on measurement data for 2000s and 2012s. It shows the increasing trend in nutrient pollution between 2000s and 2012s according to the measurements. This justifies why we have higher values in 2012 compared to 2000 from the other models. Our results for the source attribution are comparable with the results of the sub-basin approach from the MARINA 1.0 model. This confirms our model approach is somewhat robust: we model on polygon units while our aggregated results are in line with the sub-basin model. However, our results for the source attribution differ from the results of IMAGE-GNM. A possible reason is that we account for manure point sources whereas IMAGE-GNM does not.

Comparisons with the other studies for main basins in China

We compare our results with existing studies estimating nutrient inputs for main Chinese basins in Table S8. We compare our results with these studies in terms of nutrient inputs to rivers and their sources for the major river basins in China.

For **nutrient inputs to rivers**, Xu et al.[129] quantified around 730 and 100 kton of TN and TP inputs to the **Pearl basin** in 2012**,** respectively. Ti et al.[130] estimated 770 kton of TN in the Pearl River in 2010. We quantify around 3145 and 200 kton of TDN and TDP inputs to the Pearl basin, which are around 4 and 2 times for N and P compared to the existing study, respectively. We argue that other studies may underestimate the nutrient inputs to rivers because these values are even lower than the measured concentrations at the river mouth. Tong et al.[131] published the monthly measured concentrations of TN and TP at the river mouth of the eight major rivers in China from 2006 to 2012. In 2012, the estimated TN and TP loads at the river month (according to the concentrations and river discharges) were around 806 kton and 34 kton, respectively. The nutrient inputs to rivers should be much higher because of nutrient retentions during the export towards the river month. The differences between our results and the existing studies could be associated with different data sources (county vs province and city statistics) and model structures. For example, the direct discharges of animal manure as point sources are only accounted partly (the untreated manure from biogas project) in the study of Xu et al.[129]. The rest studies consider manure mainly as diffuse sources (retained by soil). In the study of Ti et al.[130], the model structure for quantifying nutrient inputs to rivers from diffuse sources is different (e.g. the manure discharges are accounted in their diffuse sources and manure applied on land is not included in the diffuse sources).

For the **Changjiang River** basin, Ti et al.[130] quantified around 3206 kton of TN for 2010 while we quantify around 3 times higher of TDN inputs (12730 kton) to rivers. Liu et al.[132] quantified 4500 kton of TN entering surface waters in 2000. Measurements showed that the TN concentrations in Changjiang at the river mouth increased 2 fold from 2000 to 2012 (Table S7). Since nutrient inputs to rivers strongly correlate to river exports, we consider our estimates are more in line with the increasing measured trend of river exports. Han et al.[133] used the measurement data for nutrient concentrations in soil and water to estimate the TN (2073 kton) and TP (477 kton) inputs to the **Huanghe** basin for 2005. The estimates are higher than our estimates for TDN (1462 kton) and TDP (207 kton) for 2012. This may due to the fact that we do not account for particulate forms while dissolved forms take only part of the proportion according to their measurements.

We also compare our estimates with other studies by **source**. For **diffuse sources**, two more formalized approaches (NANI and N budget approaches) for quantifying the nutrient inputs to rivers from **diffuse sources** are summarized in Table S8. These two approaches are, in principle, similar and can be explained by the two main steps. First, nutrient inputs to land from human activities (e.g., fertilizer applied on land) are quantified. Second, how much of these inputs to land entering rivers is quantified (soil retentions are accounted for via parameters, summarized as 'soil retention parameter' in Table S8).

The NANI approach quantifies the total net anthropogenic N inputs to rivers (see details in Table S8) and applies an empirical coefficient to represent the percentage of these N inputs entering the rivers. This empirical relationship established originally using the data from regions draining to the coastal zone of the North Atlantic Ocean (Howarth et al.[134]). Later, this relationship was found applicable to other world regions in the temperate zone. Studies in Table S8 adopted the NANI approach and were mainly based on the uniformed empirical coefficient from other regions or studies. They were applied to the Chinese basins to quantify inputs of nutrients to rivers on the basin scale.

The nutrient budget (N inputs deducted N exports) is another commonly used method to quantify inputs of nutrients to rivers (see details in Table S8). This method is used in Global *NEWS*-2, MARINA 1.0 and IMAGE-GNM. The soil retention parameter is quantified as a function of annual runoff from land to streams using calibrated (in Global *NEWS*-2) or un-calibrated (in IMAGE-GNM) parameters (e.g. runoff, slope). Our study is based on the approach from Global *NEWS*-2 and MARINA 1.0. We quantify the nutrient budget based on the county statistics and the NUFER county outputs. We quantify the soil retention parameter as a function of the total runoff from the VIC hydrological model on the half-degree grid scale to account for the variability within the basin. When we re-aggregate the results and calculate the soil retention parameter on the basin scale, then we model higher percentages of nutrients entering the rivers compared to the Global *NEWS*-2 results for 2000. However, the relative ratio of these percentages among the basins are comparable. This somehow agrees with the studies, which suggest that the capacity in the watershed to retain N can be diminished due to the increasing N inputs from human activities[135-137]. For example, for the Changjiang basin, the average soil retention for DIN (from $\boldsymbol{FE_{ws,F}}$ parameter: 49%) on the basin scale captures the increasing trend of $\boldsymbol{FE_{ws,F}}$ (increased from 0.11 to 0.61 from 1970 to 2003)[138].

For direct **manure discharges**, Table S8 indicates that there are still limited studies to account for this source of nutrients in rivers. However manure discharges are major sources of nutrients in many Chinese rivers [12, 118, 119, 139-142]. Ma et al. [118] quantified that 30% of N and 45% of P from manure were discharged to surface waters in 2005 at the national scale and Wang et al.[12] showed that the manure discharges contributed still substantially to nutrient pollution in 2012 with the national average discharge rate of 35% for N and 54% for P[12].

For **human waste discharges from WWTPs**, our estimates are generally lower compared with other studies and with 2000 (Table S8). Compared to 2000, we estimate lower nutrient discharges from WWTPs to rivers in 2012. This is in line with the studies and reports that show decreasing trends in nutrient inputs from sewage systems[143-147]. A possible reason is the improved treatment of the wastewater in WWTPs. Our averaged N and P removal rates during treatment on the national scale is 61% and 74% respectively, which are close to the average value of the most commonly used technologies in WWTPs in China (TN ranging from 55% to 59% and TP ranging from 70% to 78%)[143, 144]. Some studies adopted the method from Van Drecht et al.[24]. We consider our inputs are more up-to-date (e.g. treatment efficiencies) and spatially detailed (e.g. county inputs for treatment rate). For example, Ti et al. [130] adopted the approach and inputs from Van Drecht et al. [24]. This could result in potential over-estimations of nutrient inputs to rivers from sewage systems compared to our study. This is because the treatment removal efficiencies of Van Drecht et al. [24] are generally lower than in our study. Some studies estimated nutrient discharges to rivers from WWTPs using the uniform nutrient concentrations of human waste and industrial waste. For example, Zhang et al. [144] estimated 146 kton of TN discharged to the Huai River from WWTPs (including human waste and industrial discharges) on average over the period of 2003-2012. Our estimate is 37 kton TDN inputs to Huai in 2012 from human waste discharged from WWTPs. This is in line with the First National pollution consensus [142] that most of the discharges (around 60%) from WWTPs are industrial discharges.

In summary, we conclude that we apply the approach to quantify the nutrient budget of diffuse sources from validated models (MARINA 1.0 and Global NEWS-2). And the parameters representing retention processes for diffuse sources are in acceptable ranges. There are still limited studies considering manure discharges in their estimates in the nutrient inputs to rivers. For human waste discharges from WWTPs, we have more up-to-date model inputs and our estimates are in line with the development of WWTPs and the decreasing measurement trend.

Table S7. Nitrogen (N) and phosphorus (P) loadings into coastal seas by three main rivers for the years 2000 and 2012 (kton $year^{-1}$).

	N load (kton $year^{-1}$)	**P load** (kton $year^{-1}$)	**References**		**N load** (kton $year^{-1}$)	**P load** (kton $year^{-1}$)	**References**
Zhujing (or Pearl in English)							
2000s*	443 (370~554)	9 (5~17)	[148-151]	**2012s***	550 (327 ~806)	21 (9~34)	[131, 152-154]
Changjiang (Yangtze in English)							
2000s*	812 (560~1201)	19 (2~28)	[148-150, 155, 156]	**2012s***	2572 (2046~3000)	216 (196~235)	[131, 157, 158]
Huanghe (or Yellow in English)							
2000s*	88 (15~163)	0.46 (0.04~0.82)	[149, 150, 155, 159-162]	**2012s***	99 (22~186)	0.62 (0.23~1.26)	[131, 133, 162, 163]

*:2000s include the years below 2004. 2012s include the years after 2006.

**: We estimated mean N loads from different studies to compare the general trend between the years. N and P loads are estimated using measured concentrations at the river mouth and water discharges at the river mouth. Measured concentrations differ among nutrient forms and the existing studies. Some studies focus on the total N loads and others on N specific species (NH_4-N, NO_3-N or NO_2-N). Some studies focus on the total P and some on phosphate.

Table S8. Overview of the existing studies for nutrient inputs to the Chinese rivers and comparison of the main aspects with our study. N and P are nitrogen and phosphorus, respectively. WWTPs is wastewater treatment plants.

		Year	**N and P inputs or balance* for diffuse sources**	**Soil retention parameter (% of inputs to land)****	**Manure discharges to rivers**	**WWTPs discharges to rivers**	**Total inputs to rivers**	**Measured load (after retentions during river exports) *****
Zhujiang (Pearl)	Xu et al. [129]	2012 (2000 - 2030)	N inputs[a] 2050 kton P inputs[a]: 70 kton	TN: 15%; TP: 5% (Uniformed factor from literature for the whole basin)	TN:50 kton; TP:21 kton (Treated after biogas fertilizer production and discharge)	TN[b]:310 kton TP[b]:35 kton (human waste discharges)	TN: 730 kton TP:100 kton	Year 2012: TN: 806 kton TP: 34 kton Source: Tong et al.[131]
	Ti et al.[130]	2010 (1980 – 2010)	N inputs[c]: value not explicit P inputs: -	TN: 5.14% for fertilizer	TN: value not explicit (22% of the total manure excretion discharges to water)	TN[d]: 379 kton (human waste discharges)	TN: 770 kton	
	This study	2012	N balance[e]: 3837 kton P balance[e]: 271 kton	DIN[f]: 66% DON[f]: 0.7% DIP[f]: 11.5% DOP[f]:0.7%	TDN[g]:317 kton TDP[g]:112 kton	TDN: 91 kton TDP: 20 kton (human waste discharges)	TDN: 3146 kton TDP: 203 kton	
Changjiang (Yangtze)	Liu et al.[132]	2000	N inputs[h]: 14250 kton	TN: 25% (Uniformed factor from literature for whole basin)	Not included	Not included	TN: 4500 kton	Year 2012 N load: 2572 kton Sources: Tong et al. [131]
	Yan et al. [135]	2003	N balance: 13140 kton	DIN[i]: 50%	Not included	DIN: 560 kton (human waste and industrial discharges)	DIN: 7130 kton	

								Jiang et al.[157] Li et al. [158]

Table S8 (continued)

	Ti et al.[130]	2010 (1980 – 2010)	N inputs[c]: value not explicit P inputs: Not applicable	TN: 5.14% for fertilizer	TN: value not explicit (22% of total manure excretion discharged to water)	TN[d]: 1339 kton (human waste discharges)	TN: 3206 kton	
	Liu et al.[126]	2010 (1900-2010)	N balance: 14200 kton P balance: 1700 kton	TN: 51% TP: 35%	Not included	TN: 505 kton TP: 64 kton (human waste and industrial discharges)	TN: 8148 kton TP: 729 kton	
	This study	2012	N balance[e]: 19417 kton P balance[e]: 2159 kton	DIN[f]: 50% DON[f]: 0.6% DIP[f]: 9% DOP[f]:0.6%	TDN[g]: 1878 kton TDP[g]: 718 kton	DIN: 270 kton DON: 59 kton DIP: 79kton DOP:0.79 kton (human waste discharges)	DIN: 11627 kton DON: 1077 kton DIP: 867 kton DOP: 282 kton	
Huanghe (Yellow)	Han et al.[133]	2005	NA	NA	NA	NA	TN[j]: 2073 kton TP[j]: 477 kton	-
	My study	2012	N balance[e]: 6176 kton P balance[e]: 518 kton	DIN[f]: 12% DON[f]: 0.15% DIP[f]: 1% DOP[f]:0.16%	TDN[g]: 482 kton TDP[g]:147 kton	TDN: 92 kton TDP: 20 kton (human waste discharges)	TDN: 1462 kton TDP: 207 kton	
Hai & Huai	Chen et al.[164] (Hai)	2008-2012 (average)	NANI[k]: 4216 kton P inputs: Not applicable	NA	NA	NA	NA	-
	My study (Hai)	2012	N balance[e]: 3062 kton N inputs[l]: 4293 kton	DIN[f]: 14% DON[f]: 0.17% DIP[f]: 1% DOP[f]:0.17%	TDN[g]: 468 kton TDP[g]:177 kton	TDN: 112 kton TDP: 24 kton (human waste discharges)	TDN: 1126 kton TDP:237 kton	
	Zhang et al.[144] (Huai)	2003-2012 (average)	NANI[k]: 7194 kton P inputs: Not applicable	NA	NA	TN[n]: 146 kton (human waste and industrial discharges)	NA	
	My study (Huai)	2012	N balance: 1446 kton N inputs: 2077 kton	DIN[f]: 33% DON[f]: 0.38% DIP[f]: 4% DOP[f]:0.36%	TDN[g]: 231 kton TDP[g]:104 kton	TDN: 37 kton TDP: 8 kton (human waste discharges)	TDN: 812 kton TDP:138 kton	

*: Nutrient inputs and nutrient balances are different. Nutrient balances (surplus) refer to inputs minus outputs (i.e. crop exports). Some studies use nutrient inputs to estimate the export of nutrients from diffuse sources to water bodies while some use nutrient balances.
**: This parameter represents the percentage of nutrient inputs from land entering water bodies after retentions (different among studies).
***: We use measured N and P concentrations in the river mouth and multiplied them with the discharges at the river mouth to obtain the loads. The inputs to rivers should be lower than measured load due to the retentions in rivers (e.g. in-stream retention, dams or reservoirs retention) during river exports.

a: N and P inputs to rivers from diffuse sources include fertilizer, biogas fertilizer from manure, manure and crop residues;
b: This could be potentially over-estimated because it assumes that human waste generated by the total population are emitted via WWTPs instead of connected population like this study. See details in Section S1.3.
c: N inputs to rivers include manure and rural human waste that is directly discharged to rivers, N losses via leaching and runoff from arable land and natural area, N losses from urban area.
d: Adopted method from Van Drecht et al.[39]: assumed urban population are fully connected to WWTPs and no connection of rural population; and the N removal rates were not explicit and may followed the categories from Van Drecht et al.[39] which are not up-to-date; These could result in over-estimation of wastewater discharges.
e: N and P balances refer to inputs minus outputs (crop exports). For more details please go to Section 2.1 Model description in the manuscript.
f: We re-aggregate the results and calculate the value on the basin scale. The original calculation is on the polygon scale and calculated as the function of runoff.
g: Manure discharge rates to the total manure excretion range from 22% to 56% for N and from 30% to 76% for P. It differs among counties. The national average is 35% and 54% for N and P, respectively.
h: N inputs to rivers refer to anthropogenic reactive N, including synthetic fertilizers, atmospheric deposition and biological N_2 fixation.
i: it is calculated as a function of annual total runoff and adjusted against the measurement data; see details in the indicated literature.
j: The estimation is based on the TN concentration in soil and water. Dissolved N takes a small proportion (NH_4-N and NO_3-N takes around 4% of TN).
k: NANI (Net Anthropogenic N Input) includes fertilizer, atmospheric N deposition, biological N_2 fixation and the net food and feed import.
l: N inputs equal to N balances plus N exports. We derive this value to compare with other studies.
n: the TN discharges to rivers from point sources are the product of human waste and industrial discharges and the volume of wastewater from human waste and industries. It assumed a full connection of urban population and no connection of rural population to sewage systems; and applied the uniformed average N concentration in human waste and industrial sewage effluent.

S4.5 Uncertainties

Here, we address the uncertainties in our study. For instance, our model includes some calibrated coefficients from Global NEWS-2[9]. Applying these to other scales than the basin scale introduces uncertainty. This holds for the equation for soil retention ($\boldsymbol{FE_{ws,F}}$) and particular for DIN (the elementary effect is 72%.) Nevertheless, our retention for DIN on the basin scale such as Changjiang (49%) captures the increasing trend of $\boldsymbol{FE_{ws,F}}$ for DIN (increased from 0.11 to 0.61 from 1970 to 2003[138]). This agrees with studies that suggest the capacity in the watershed to retain N can be diminished due to the increasing N inputs from human activities[135-137]. For DIP, Harrison et al.[165] also applied $\boldsymbol{FE_{ws,F}}$ on grid, with a satisfactory model performance. Another source of uncertainty is the 30 years average (1970 – 2000) that we use for the total runoff, ignoring annual variability. Although the runoff is not changing dramatically for the current year, this will also influence the estimation of diffuse sources. For manure discharges, there are uncertainties inherited in the discharge rate for each county, which are derived from the field surveys and expert knowledge [12, 166]. We use the rural residential areas as the proxy for locations of manure farms and distribute the manure discharges from the county scale to each polygon based on the area weighted method. This could introduce uncertainties because: (1) they are not real locations of industrial manure farms; (2) the area weighted distribution does not represent the animal numbers for each farm. We recognize the uncertainties and try to quantify them by randomly selecting 20% of the polygons and changing the percentage of manure farms by 10%. The mean elemental effect for each nutrient form is less than 50% (Table S6). These uncertainties in distributing manure discharges to polygons are within the individual counties. Zhao et al.[19] also adopt the same locations as the proxy for manure farms and evenly allocated the NH_3 emissions from manure to these locations. Recognizing the uncertainties, we consider that we made the best use of the information that we could find. Other uncertainties may also lie in the distribution of the diffuse sources. We do not include the grassland and consider only the arable land. However, this simplification could be negligible since the fertilizers applied on grassland in2014 only take less than 3% and 6% of the total fertilizer use for N and P, respectively[167].

Despite the uncertainties, we consider our model appropriate to analyse inputs of nutrients to rivers from diffuse and point sources for four main reasons. First, the model inputs are considered of the acceptable quality. NUFER and VIC provide important inputs for our study and are both widely applied in their own fields. We use statistics year books for the Chinese counties, which are known as the most reliable data source in China[12, 13]. The unique wastewater treatment plants database is created based on the official governmental documents, exact locations with XY coordinates, literature reviews and expert knowledge (section S3). Second, we compare our model approaches with other models performing on different scales. We consider our model mimic the approach of the MARINA 1.0 and Global NEWS-2 models for diffuse sources. We improved the method for point sources (Table S5). Third, the model responses to the sensitivity analysis suggest that the model is fairly robust, with the elementary effects of smaller than 1 (and in most of cases substantially smaller) (Table S6). Fourth, the model comparisons (Figure S6,Table S8) cover different approaches (sub-basin and grid). Compared to other studies for the main basins in China, there are still limited studies considering manure discharges in their estimates of nutrient inputs to rivers. We provide a better representation for WWTPs. All this builds our confidence in using our model to quantify nutrient inputs to rivers by source.

S5 Supplementary results

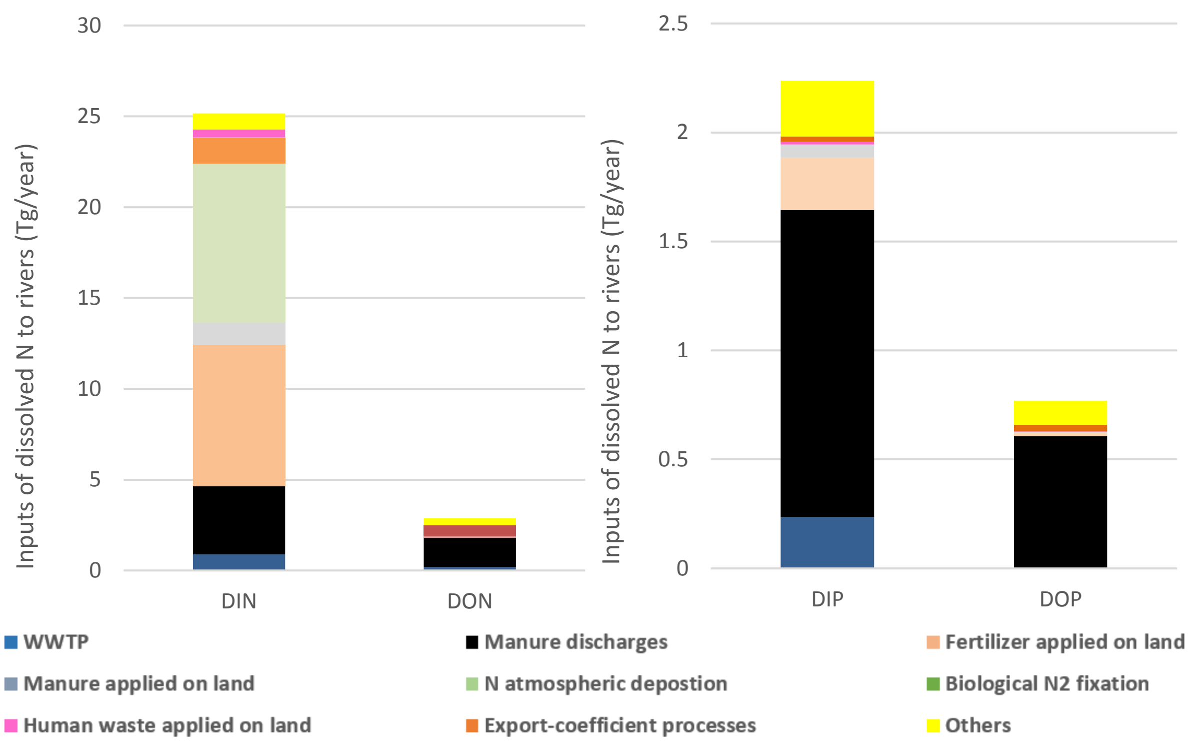

Figure S7. Nitrogen (N) and phosphorus (P) inputs to rivers in China by form: dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), dissolved inorganic phosphorous (DIP), dissolved organic phosphorus (DOP) and by source (Tg/year)). Point sources include human waste from wastewater treatment plants (WWTPs) and direct discharges of animal manure to rivers. Diffuse sources include synthetic fertilizer use, manure applied on land, biological N_2 fixation by agricultural crops and by natural vegetation, atmospheric N deposition, human waste from population unconnected to sewage systems that stays on land, and export-coefficient processes. For DON, the export-coefficient processes refer to leaching of organic matter. For DIP and DOP, it refers to the weathering of P-contained minerals from agricultural and non-agricultural soils. "Others" refer to the human waste from urban and rural population that are unconnected to sewage systems, but discharged directly to rivers.

Figure S8. Locations of animal farms and wastewater treatment plants (WWTPs). The names of the provinces in China are presented on the maps.

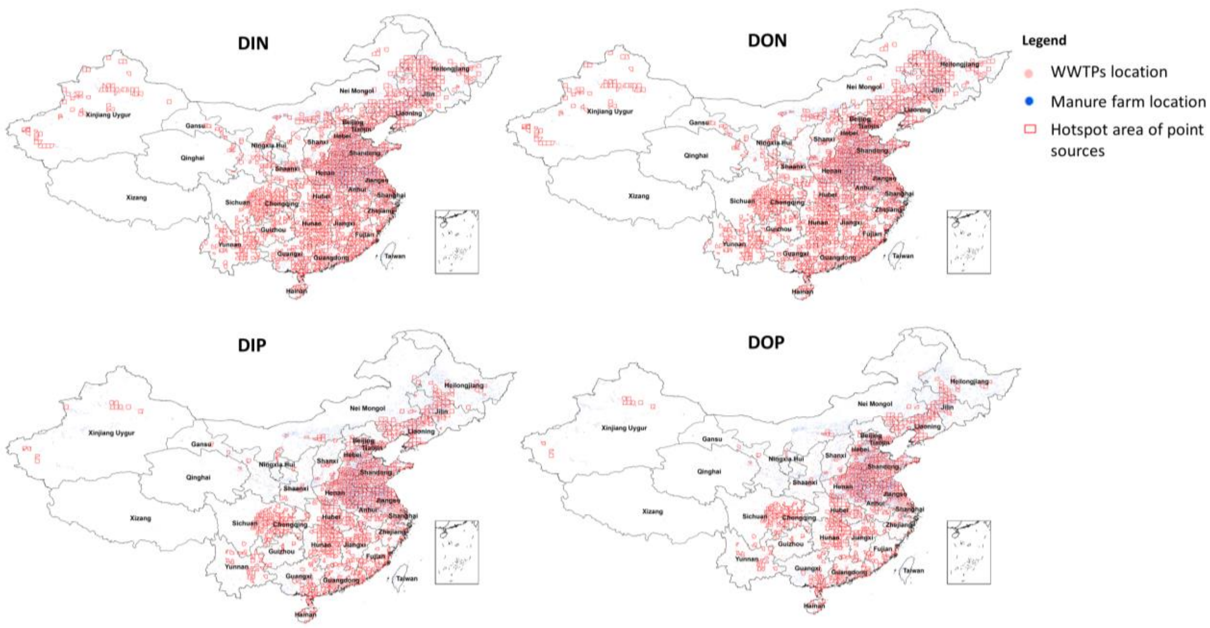

Figure S9. Hotspot areas of point sources (manure discharges and human waste emitted from wastewater treatment plants (WWTPs)). Nutrients are dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) and the locations of the WWTPs and animal farms. Hotspots are defined as the areas where point sources contribute to 50% of the total DON, DIP and DOP inputs to the rivers. For DIN, the hotspots are the areas where point sources contribute to 18% of the total DIN inputs to the rivers.

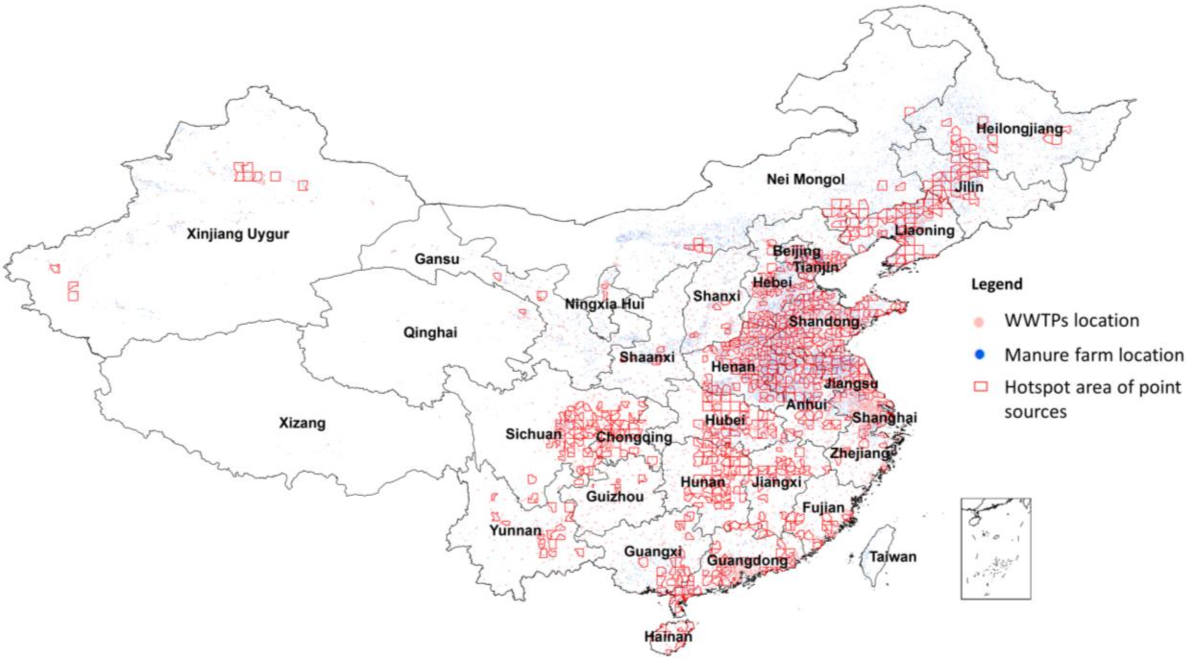

Figure S10. Hotspot areas of point sources (direct discharges of animal manure and human waste emitted from wastewater treatment plants (WWTPs)) inputs to rivers for all the nutrient forms (dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P)); These hotspot areas are determined by overlapping hotspot areas for individual nutrient forms (Figure S9). Hotspots are defined as areas where point sources contribute to 50% of the total DON, DIP and DOP inputs to the rivers. For DIN, the hotspots are the areas where point sources contribute to 18% of the total DIN inputs to the rivers.

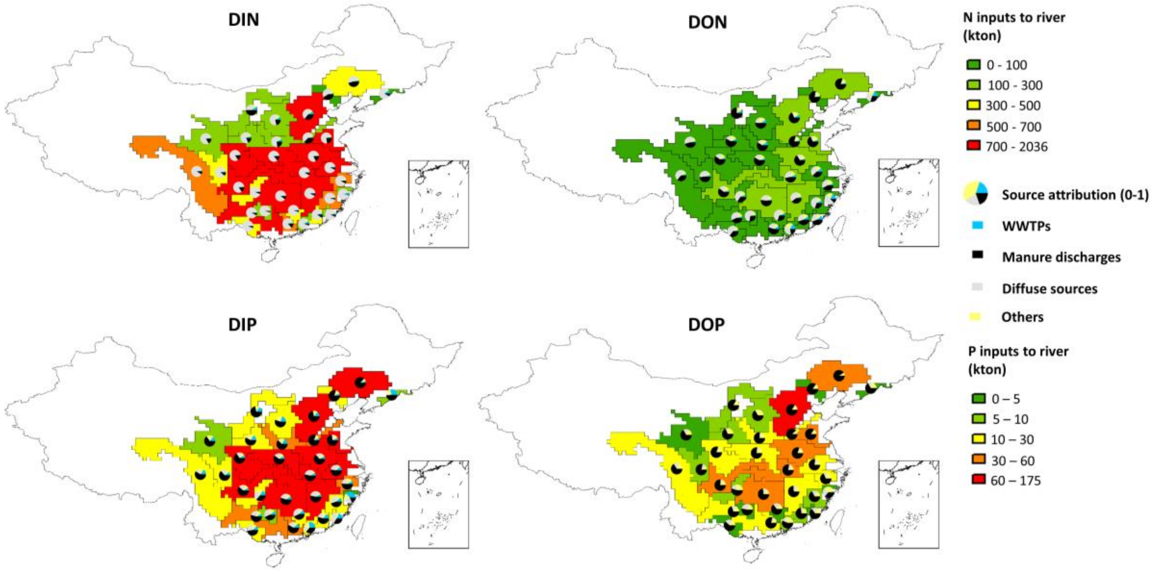

Figure S11. Dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) inputs to rivers by sub-basin (in kton year^{-1}). Pie charts show the share of different sources in inputs of nutrients to rivers (0-1). WWTPs refers to human waste emitted from wastewater treatment plants. Manure discharges refer to the manure directly discharges to rivers. Diffuse sources include synthetic fertilizer use, manure applied on land, biological N_2 fixation by agricultural crops and by natural vegetation, atmospheric N deposition, leaching of organic matter, and weathering of P-contained minerals from agricultural and non-agricultural soils, human waste that is applied on land from population unconnected to sewage systems. "Others" sources include the human waste from urban and rural population that is not connected to sewage systems and discharged to rivers.

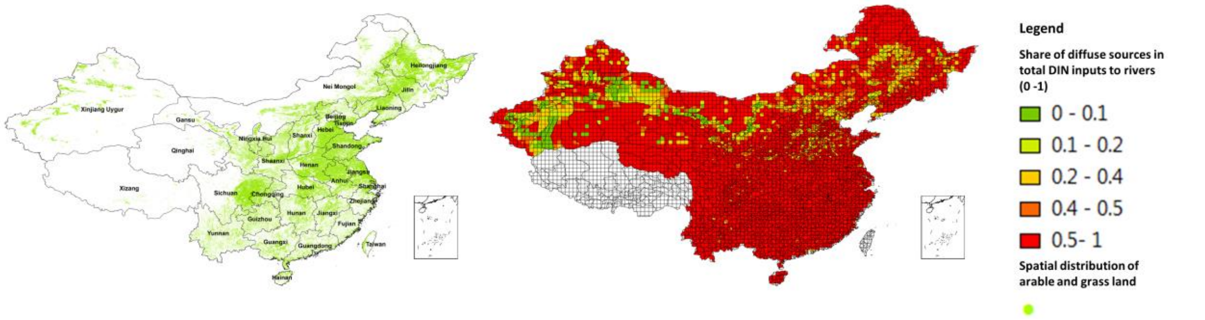

Figure S12. Share of the diffuse sources in the total dissolved inorganic nitrogen (DIN) inputs to rivers (0-1) and the spatial distribution of the arable and grass land (1×1km grid). Diffuse sources for DIN include use of synthetic fertilizers, manure and human waste from unconnected population on land, biological N_2 fixation, atmospheric N deposition, P weathering and leaching of organic matter.

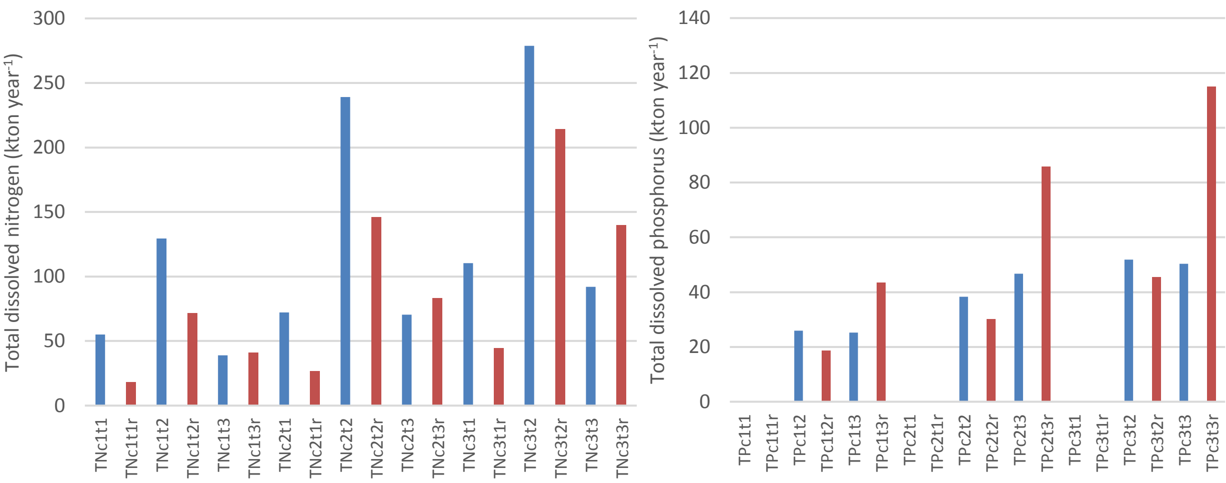

Figure S13. Total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP) inputs to all Chinese rivers from wastewater treatment plants (WWTPs) (kton year^{-1}). The wastewater treatment plants are categorized by the treatment efficiency and capacity. Both treatment efficiencies (fractions) and capacities (m^3/day) are categorized into three classes. The capacities (104 m^3/day) are grouped into small (c1, capacity <1.4), medium (c2, capacity <3.1) and large (c3, capacity > 3.1). The treatment efficiencies are grouped into primary (t1, treatment efficiencies <0.35 for N and <0.4 for P), secondary (t2, treatment efficiencies <0.55 for N and <0.65 for P) and tertiary (t3, treatment efficiencies >0.55 for N and >0.65 for P). For example, TNc1t1 refers to TDN emitted from WWTPs with capacity class 1 (small capacity) and treatment class 1 (primary treatment for N). TNc1t1r refers to the TDN removal by the WWTPs with capacity class 1 and treatment class 1.

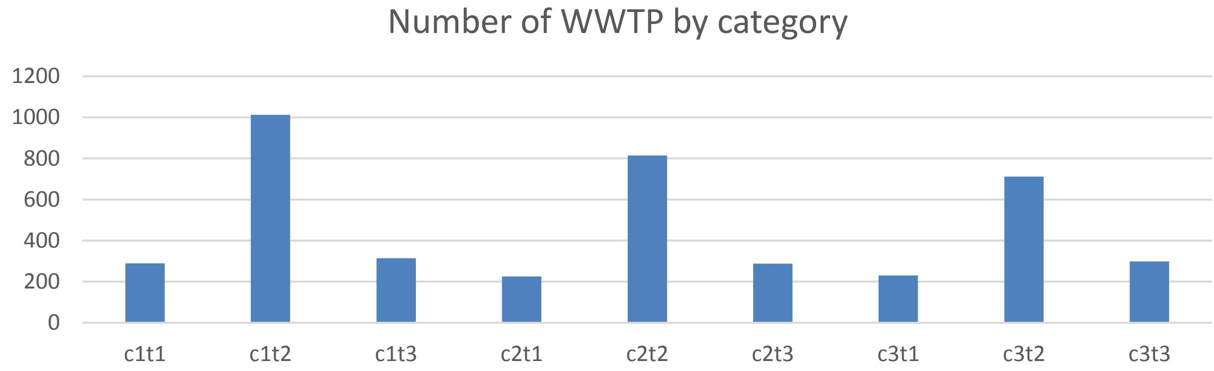

Figure S14. The number of wastewater treatment plants for nine categories. The wastewater treatment plants are categorized by the treatment efficiency and capacity. For both treatment efficiency and capacity they are categorized into three classes. The capacities are grouped into small (c1), medium (c2) and large (c3). The treatment efficiencies are grouped into primary (t1), secondary (t2) to tertiary (t3). See details for grouping in Figure S13. For example, TNc1t1 refers to TDN emitted from WWTPs with capacity class 1 (small capacity) and treatment class 1 (primary treatment for N). TNc1t1r refers to the TDN removal by the WWTPs with capacity class 1 and treatment class 1.

Figure S13 shows nutrient inputs from 4,204 WWTPs by grouping them according to the treatment efficiencies (primary, secondary, and tertiary) and capacities (small, medium and large). Treatment plants with secondary treatment efficiencies account for more than 60% of TDN inputs to rivers from WWTPs. However for TDP, WWTPs with larger capacities and relatively higher treatment efficiencies (medium and large) contribute most. They contribute 42% to TDP pollution from WWTPs while account for 24% of the total number of WWTPs (Figure S13).

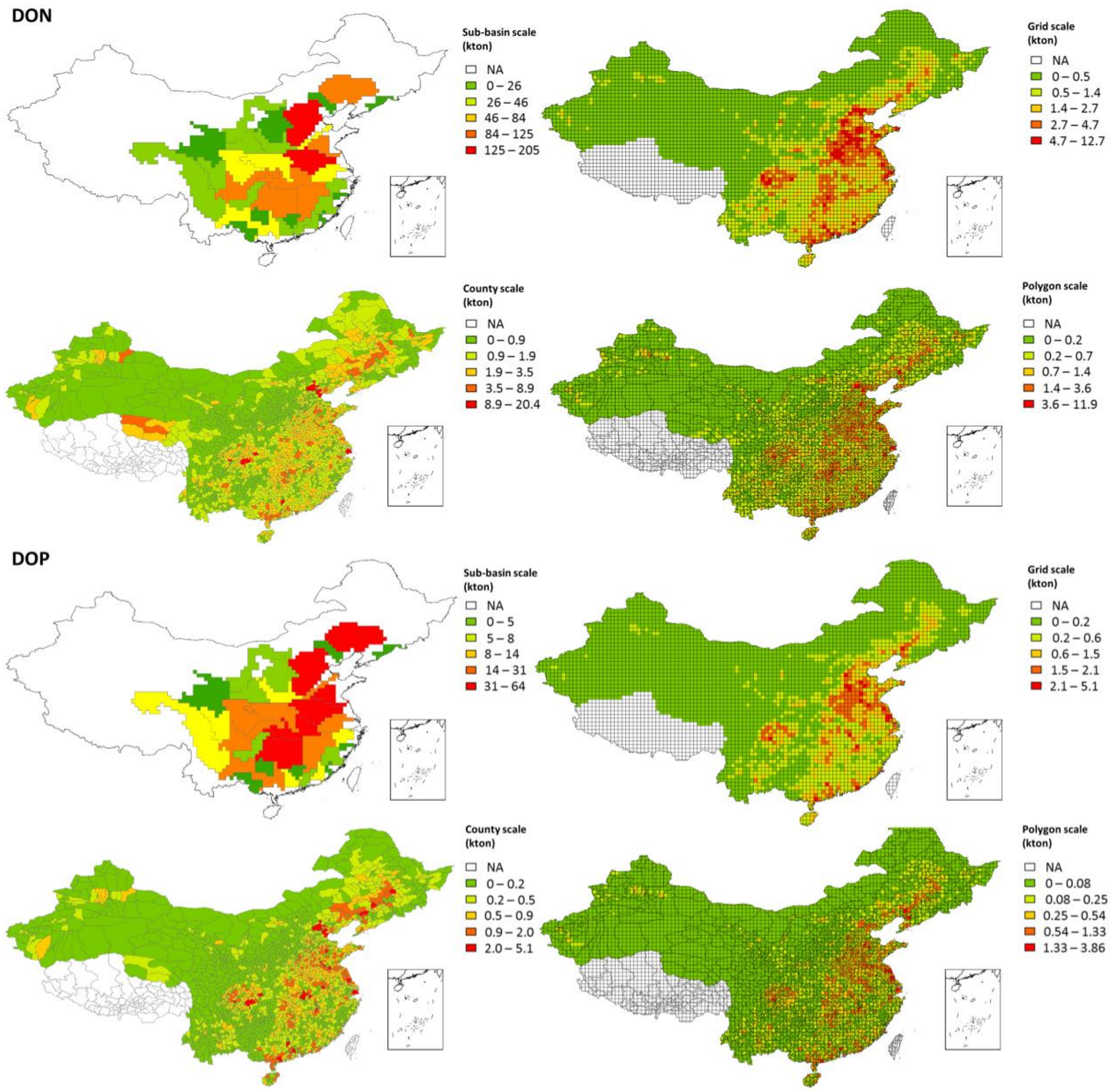

Figure S15. Dissolved organic nitrogen (DON) and phosphorus (DOP) inputs to rivers at multiple scales (kton year^{-1}) in 2012 from all sources. Multiple scales include sub-basin, grid, county and polygon. White areas indicate Tibat and Taiwan provinces, which have no detailed input data.

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