

1 **Supplementary Materials**

2 **Alarming nutrient pollution of Chinese rivers as a result of agricultural transitions**

3 Maryna Stokal<sup>1\*</sup>, Lin Ma<sup>2\*</sup>, Zhaohai Bai<sup>2</sup>, Shengji Luan<sup>3</sup>, Carolien Kroeze<sup>1,4</sup>, Oene Oenema<sup>5</sup>, Gerard Velthof<sup>5</sup>,  
4 Fusuo Zhang<sup>6</sup>

5 <sup>1</sup>Environmental Systems Analysis Group, Wageningen University, Droevendaalsesteeg 3, 6708 PB  
6 Wageningen, The Netherlands.

7 <sup>2</sup>Key Laboratory of Agricultural Water Resource, Center for Agricultural Resources Research, Institute of  
8 Genetic and Developmental Biology, Chinese Academy of Sciences, Huaizhong Road 286, Shijiazhuang, Hebei  
9 050021, China.

10 <sup>3</sup>Peking University, University Town, Nanshan District, Shenzhen 518055, China.

11 <sup>4</sup>Department of Science, Faculty of Management, Science & Technology, Open University of the Netherland,  
12 Valkenburgerweg 177, 6419 AT Heerlen, The Netherlands.

13 <sup>5</sup>Alterra, Wageningen University and Research Centre, Droevendaalsesteeg 4, 6708 PB Wageningen, The  
14 Netherlands.

15 <sup>6</sup>Center for Resources, Environment and Food Security, China Agricultural University, Beijing 100193, China.

16 \* Corresponding authors:

17 The first corresponding author: Maryna Stokal ([maryna.stokal@wur.nl](mailto:maryna.stokal@wur.nl)).

18 The second corresponding author: Ma Lin ([malin1979@sjziam.ac.cn](mailto:malin1979@sjziam.ac.cn)).

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20 Supplementary Materials contain:

- 21 1. Methods;
- 22 2. Tables S1-S2;
- 23 3. Figures S1-S12;
- 24 4. References used in the Supplementary Materials.

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## 1 **1. Methods (extended)**

2 This section is divided into two subsections (indicated by italic subheads). First, we present model description  
3 and inputs. We describe the model that we used to quantify total dissolved nitrogen (TDN) and phosphorus  
4 (TDP) inputs to Chinese rivers in 1970, 2000 and 2050. Second, we discuss the performance of the model.

5 *Model description and inputs.* We quantified TDN and TDP inputs to the Chinese rivers for 1970 and 2000 using  
6 two widely accepted models (which was not done before; see Figure S7 for model inputs and outputs): Global  
7 *NEWS-2* (Nutrient Export from WaterSheds) [1] and NUFER (NUtrient flows in Food chains, Environment and  
8 Resources use) [2]. The following rivers are included in this study: the Huang (Yellow River), Huai, Hai, Liao,  
9 Changjiang (Yangtze River) and Zhujiang (Pearl River) (Figure S1). The drainage areas of the three largest  
10 rivers, the Huang, Changjiang and Zhujiang, were divided into 22 sub-basins. Literature was reviewed to identify  
11 the main sub-basins of the Huang [3, 4], Changjiang [5, 6] and Zhujiang [7, 8]. Sub-basins were named after  
12 rivers (or lakes) at their outlets or after hydrological (monitoring) stations at their outlets. The drainage areas of  
13 the Huai, Hai and Liao rivers were not modified, and they are considered as individual sub-basins. The all sub-  
14 basins (25 in total) were delineated using the Topological Simulated Network (STN-30 v6.01) applied in Global  
15 *NEWS-2* [1, 9].

16 We implemented Global *NEWS-2* at the sub-basin scale [1, 10]. We ran the model with direct inputs of animal  
17 manure to rivers (fractions), calculated from provincial data (Figure S2) by the NUFER model [2]. TDN and  
18 TDP inputs to rivers of sub-basins were quantified by source for the years 1970 and 2000. We included point  
19 ( $RS_{pnt_{F,total,j}}$ , kg, see equation 1) and diffuse ( $RS_{dif_{F,total,j}}$ , kg, see equation 2) sources of nutrients in rivers. Point  
20 sources of the nutrients (F: TDN and TDP) include direct discharges of animal manure to rivers ( $RS_{pnt_{F,ma,j}}$ , kg),  
21 and human sewage ( $RS_{pnt_{F,sew,j}}$ , kg). Diffuse sources of the nutrients (F: TDN and TDP) include manure  
22 ( $RS_{dif_{F,ma,j}}$ , kg) and synthetic fertilizers ( $RS_{dif_{F,fe,j}}$ , kg) used in croplands, atmospheric N deposition (only for  
23 TDN;  $RS_{dif_{F,dep,j}}$ , kg), biological N fixation (only for TDN;  $RS_{dif_{F,fix,j}}$ , kg), leaching of organic matter  
24 ( $RS_{dif_{F,lch,j}}$ , kg), and P-weathering (only for TDP;  $RS_{dif_{F,wth,j}}$ , kg).

$$25 \quad RS_{pnt_{F,total,j}} = RS_{pnt_{F,ma,j}} + RS_{pnt_{F,sew,j}} \quad (1)$$

$$26 \quad RS_{dif_{F,total,j}} = RS_{dif_{F,ma,j}} + RS_{dif_{F,fe,j}} + RS_{dif_{F,dep,j}} + RS_{dif_{F,fix,j}} + RS_{dif_{F,lch,j}} + RS_{dif_{F,wth,j}} \quad (2)$$

27 Manure point sources were not included in Global *NEWS-2* [1] before, and are calculated using fractions of  
28 manure directly discharged to rivers from NUFER [2] (see below equations 3 and 4). Nutrient inputs from  
29 sewage and diffuse sources were quantified following Global *NEWS-2* [1] (see below equations 5-13).

30 For the year 2050 we calculate TDN and TDP inputs to rivers by source using equations 1-13 as for 1970 and  
31 2000. But, the fractions of manure directly discharged to rivers ( $fr_{N_{sw,j}}$  and  $fr_{P_{sw,j}}$  in equations 1 and 2) were  
32 used as for 2000.

33 Most model inputs for 1970, 2000 and 2050 were derived from gridded global datasets (0.5 longitude by 0.5  
34 latitude) developed for Global *NEWS-2* [11-13], and most of model parameters are also from this model [1]  
35 (Figure S7). For the year 2050 these Global *NEWS-2* inputs were developed based on scenarios of the  
36 Millennium Ecosystem Assessment [14]. Detailed descriptions of scenarios are introduced in earlier studies [11-  
37 15]. In this study we used inputs of the Global Orchestration (GO) scenario, representing the globalized world  
38 with reactive management of environmental problems [14, 16] (see below a brief description of the scenario).

39 Below we describe sub-basin scale calculations and the globalized scenario with reactive management (GO) for  
40 China.

41 ○ Quantifying nutrient inputs to rivers of sub-basins from the manure point source

42 TDN ( $RS_{pnt_{TDN,ma,j}}$ , kg) and TDP ( $RS_{pnt_{TDP,ma,j}}$ , kg) inputs to rivers of sub-basin j from the manure point sources  
43 were calculated as:

$$44 \quad RS_{pnt_{TDN,ma,j}} = Nexc_j \cdot fr_{N_{sw,j}} \quad (3)$$

$$45 \quad RS_{pnt_{TDP,ma,j}} = Pexc_j \cdot fr_{P_{sw,j}} \quad (4)$$

1 Here  $N_{exc_j}$  and  $P_{exc_j}$  are the N and P animal excretion in each sub-basin (j) (kg), respectively. We used gridded  
 2 Global *NEWS-2* information (0.5 longitude by 0.5 latitude) on animal manure excretion (losses of N during  
 3 storage and housing were taken into account) (see Figure S7) [1]. These values were originally derived from  
 4 IMAGE (Integrated Model to Assess the Global Environment) [17] and prepared by Bouwman, et al. [12] for  
 5 Global *NEWS-2*. IMAGE first calculated country-based manure production based on animal stocks and excretion  
 6 rates, and then spatially allocated this manure production over the grids (0.5 longitude by 0.5 latitude) using land  
 7 use maps (see details in [12, 17]). We used this gridded information on animal manure excretion to calculate  
 8 animal manure for sub-basins (see Figure S8 for values) using ArcGIS functions. We used these values for P  
 9 excretion ( $P_{exc_j}$ ). We calculated N excretion ( $N_{exc_j}$ ) from the available N animal manure from Global *NEWS-2*  
 10 while accounting for N losses to the air (these losses were derived from NUFER, see the next paragraph).

11  $frN_{sw,j}$  and  $frP_{sw,j}$  are the fractions of N and P in animal excretion that are discharged directly to surface waters  
 12 (sw) of each sub-basin (j), respectively. These fractions as well as N losses to the air (indicated as  $frN_{NH_3,j}$  in eq.  
 13 11 below) were calculated for each sub-basin from provincial data by NUFER [2] (Figure S2). The main purpose  
 14 of NUFER is to quantify N and P use efficiencies in food chain (includes crop and animal production, food  
 15 processing and households) for 31 provinces in China for 1980, 2005 and future. Animal production (see details  
 16 on the other aspects of NUFER description in [18]) includes two intensities of animal breeding, reflecting  
 17 traditional and industrial farming systems. The high intensity types of animal breeding are defined for pig  
 18 systems (> 50 heads), dairy cattle (> 5 heads), beef cattle (> 50 heads), layer poultry (>500 heads), and meat  
 19 poultry (> 2000 heads). Manure excretion was calculated based on animal feed from crop and kitchen residues,  
 20 animal stocks (e.g., pigs, layer and broiler poultry, milk and beef cattle, sheep and goat) and excretion rates for N  
 21 and P (see “Model performance” below for sources of information). Discharge of N and P from animal manure  
 22 to surface waters was calculated from manure excretion by correcting for losses during storage and housing, and  
 23 for applications to cropland and to grassland (see details in [18]). The fractions of N and P in animal manure  
 24 discharged to waters, applied to cropland and grassland, and lost to the air during storage and housing are given  
 25 in Figure S2 for Chinese provinces. These fractions were calculated for 1980 and 2005 because NUFER includes  
 26 those years. We calculated these fractions for sub-basins (area-weighted averages) in ArcGIS (Figure S2). Thus  
 27 the manure fraction that is directly discharged to surface waters of a sub-basin is  $\sum_i$  (fraction of area covered by  
 28 province  $i \cdot$  fraction of direct manure discharge for province  $i$ ).

29 ○ Quantifying nutrient inputs to rivers of sub-basins from the other sources

30 TDN and TDP inputs to rivers from other sources (y) in each sub-basin (j) were calculated following the Global  
 31 *NEWS-2* approach: an export-coefficient approach (indicated as EA in equations below) for leaching of organic  
 32 matters and weathering of P-minerals, and a mass-balance approach (indicated as MA in equations below) for  
 33 animal manure (as a diffuse source), synthetic fertilizer use, human sewage effluents (human waste and  
 34 detergents), biological  $N_2$  fixation, atmospheric N deposition. Global *NEWS-2* quantifies inputs of the dissolved  
 35 N and P to rivers in inorganic (DIN, DIP) and organic (DON, DOP) forms. Thus, TDN was calculated as the  
 36 sum of DIN and DON, and TDP was calculated as the sum of DIP and DOP.

37 Nutrient inputs to rivers from leaching (for DON, DOP) over agricultural ( ${}^{EA}RSdif_{F,lch,ant,j}$ , kg) and non-  
 38 agricultural soils ( ${}^{EA}RSdif_{F,lch,nat,j}$ ) were calculated using equations 5 and 6. Nutrient inputs to rivers from P-  
 39 weathering (for DIP) over agricultural ( ${}^{EA}RSdif_{F,wth,ant,j}$ , kg) and non-agricultural soils ( ${}^{EA}RSdif_{F,wth,nat,j}$ ) were  
 40 calculated similarly to leaching using equations 7 and 8.

$$41 \quad {}^{EA}RSdif_{F,lch,ant,j} = f_F(Rnat_j) \cdot EC_{F,lch} \cdot Ag_{fr,j} \quad (5)$$

$$42 \quad {}^{EA}RSdif_{F,lch,nat,j} = f_F(Rnat_j) \cdot EC_{F,lch} \cdot (1 - Ag_{fr,j}) \quad (6)$$

$$43 \quad {}^{EA}RSdif_{F,wth,ant,j} = f_F(Rnat_j) \cdot EC_{F,wth} \cdot Ag_{fr,j} \quad (7)$$

$$44 \quad {}^{EA}RSdif_{F,wth,nat,j} = f_F(Rnat_j) \cdot EC_{F,wth} \cdot (1 - Ag_{fr,j}) \quad (8)$$

45 In Global *NEWS-2*  $f_F(Rnat_j)$  is a function of annual runoff from land to streams of each sub-basin j (meter),  
 46 calculated for these nutrient forms (F: DIP, DON, DOP, not for DIN). This function was empirically calculated  
 47 from annual runoff using calibrated or default constants (details are given in Mayorga, et al. [1]). Runoff values  
 48 were taken from the gridded database [11] prepared for Global *NEWS-2* [1]. Calibrated or default constants are

1 available in Mayorga, et al. [1].  $EC_F$  is the watershed export constant ( $\text{kg N or P km}^{-2} \text{ year}^{-1}$ ). This constant  
 2 represents P weathering for DIP ( $EC_{DIP, \text{wth}} = 26 \text{ kg km}^{-2} \text{ year}^{-1}$ ) and leaching of organic matter for DON  
 3 ( $EC_{DON, \text{lch}} = 280 \text{ kg km}^{-2} \text{ year}^{-1}$ ) and DOP ( $EC_{DOP, \text{lch}} = 15 \text{ kg km}^{-2} \text{ year}^{-1}$ ) according to Mayorga, et al. [1].  $Ag_{fr, j}$   
 4 is the fraction of agricultural areas in a sub-basin  $j$  (0-1). Agricultural areas account for grasslands, cropland,  
 5 wetland rice and legumes in Global *NEWS-2*. These areas were calculated from gridded database of Global  
 6 *NEWS-2* [1, 12] for sub-basins.

7 Nutrient inputs to rivers from use of animal manure and synthetic fertilizers on land (for DIN, DIP, DON, DOP),  
 8 biological  $N_2$  fixation (for DIN) and atmospheric N deposition (for DIN) over agricultural areas ( $^{MA}RSdif_{F, y, \text{ant}, j}$ ,  
 9 kg), and nutrient inputs to rivers from biological  $N_2$  fixation (for DIN) and atmospheric N deposition (for DIN)  
 10 over non-agricultural areas ( $^{MA}RSdif_{F, y, \text{nat}, j}$ , kg) were calculated as follows ( $y$  represents a nutrient source):

$$11 \quad ^{MA}RSdif_{F, y, \text{ant}, j} = WSdif_{E, y, \text{ant}, j} \cdot G_{F, j} \cdot FEWS_{F, j} \quad (9)$$

$$12 \quad ^{MA}RSdif_{F, y, \text{nat}, j} = WSdif_{E, y, \text{nat}, j} \cdot FEWS_{F, j} \quad (10)$$

13 In Global *NEWS-2*  $WSdif_{E, y, \text{ant}, j}$  is inputs of element (E: N or P) to agricultural land from source  $y$  in sub-basin  $j$   
 14 (kg).  $WSdif_{E, y, \text{nat}, j}$  is inputs of element (E: N or P) to non-agricultural land (watersheds) from source  $y$  in sub-  
 15 basin  $j$  (kg). Values for synthetic fertilizers, fixation and deposition were taken directly from gridded database of  
 16 Global *NEWS-2* [12] and calculated for each sub-basin (see Figure S8 for fertilizers). Values for N  
 17 ( $WSdif_{N, \text{ma}, \text{ant}, j}$ , kg) and P ( $WSdif_{P, \text{ma}, \text{ant}, j}$ , kg) inputs to land from animal manure were calculated using equations  
 18 7 and 8 (see below; abbreviations of the equations are explained above).  $G_{F, j}$  is the fraction of land-based diffuse  
 19 sources of nutrient forms (DIN, DON, DIP, DOP) that remain in the agricultural soil of each sub-basin  $j$  after  
 20 crop harvesting and animal grazing (0-1). It was calculated for sub-basin  $j$  as:  $1 - (\text{the amount of element (N or P) that is exported from soils by harvesting and grazing (kg) / the total inputs of element (N or P) to agricultural soils from all diffuse sources (kg)})$ . These sources include use of animal manure (for N and P) and synthetic  
 23 fertilizers (for N and P), biological  $N_2$  fixation by agricultural crops (for N) and atmospheric N deposition over  
 24 agricultural areas (for N). Values for harvesting and grazing as well as for the nutrient sources were derived from  
 25 gridded database of Global *NEWS-2* [1].  $FEWS_{F, j}$  is the fraction of nutrient form (F: DIN, DON, DIP and DOP)  
 26 that enter surface waters from diffuse sources (0-1). It was calculated as a function of annual runoff (see details  
 27 in Mayorga, et al. [1]).

$$28 \quad WSdif_{N, \text{ma}, \text{ant}, j} = Nexc_j \cdot (1 - (\text{fr}N_{NH_3, j} + \text{fr}N_{\text{sw}, j})) \quad (11)$$

$$29 \quad WSdif_{P, \text{ma}, \text{ant}, j} = Pexc_j \cdot (1 - \text{fr}P_{\text{sw}, j}) \quad (12)$$

30 Nutrient inputs ( $RSpnt_{F, y, j}$ , kg) to rivers from human sewage effluents (human waste for N and P, detergents for  
 31 P) were calculated as follows:

$$32 \quad RSpnt_{F, y, j} = RSpnt_{E, y, j} \cdot FEpnt_F \quad (13)$$

33 In Global *NEWS-2*  $RSpnt_{E, y, j}$  is inputs of element (N or P) to rivers from source  $y$  (human waste or detergents) in  
 34 sub-basin  $j$  (kg). These inputs were calculated by Global *NEWS-2* taking into account the produced sewage  
 35 influents from human excretion and detergents, population connected to sewage systems and nutrient removal  
 36 (see details in Van Dreht, et al. [13]). Values for  $RSpnt_{E, y}$  were derived from gridded database of Global *NEWS-2*  
 37 [13] and calculated for each sub-basin, except for the Dongjiang sub-basin of the Pearl River for 2000 (see  
 38 Figure S1 for sub-basin locations). We recalculated values of  $RSpnt_{E, y}$  for this sub-basin for 2000 and 2050  
 39 assuming a larger fraction of N and P removal during treatment (0.80 for N and P in 2000, meaning that 80% of  
 40 the nutrients in sewage effluents are removed by treatment; 0.80 for N and 0.90 for P in 2050). This removal  
 41 fraction was 0.10 for both N and P in the original Global *NEWS-2* in 2000 (0.40 for N and 0.47 for P in 2050).  
 42 Urban areas of this sub-basin are highly developed in terms of technology and economy and the removal fraction  
 43 of 0.10 for treatment facilities are thus too low for this sub-basin. The same holds for 2050.

44  $FEpnt_F$  is the fractions of nutrient form (F: DIN, DON, DIP and DOP) that enter surface waters from point  
 45 sources. These fractions are set at 1.0 for DIP, 0.01 for DOP and 0.14 for DON in Global *NEWS-2* [1]. For DIN  
 46 this fraction was calculated by Global *NEWS-2* based on the efficiency of N removal during treatment (see  
 47 details in Mayorga, et al. [1] and Van Dreht, et al. [13]). We recalculated this fraction for the Dongjiang sub-

1 basin of the Pearl River for 2000 and 2050, taking into account N removal fraction mentioned above for 2000  
2 and 2050. For the other sub-basins and nutrient forms we used the fractions provided by Global NEWS-2.

3 ○ Describing the globalized GO scenario for China

4 The GO scenario assumes a globalized world with an economy that will develop rapidly in China (e.g., at least a  
5 10-fold increase in GDP at purchase power parity between 2000 and 2050) (see also Table S1). This is because  
6 of globally connected markets and trends [10, 13]. The total population is assumed to increase slightly because of  
7 better education (e.g., low mortality and fertility rates, and high migration). However, urban population is  
8 assumed to increase fast that might be due to migration of people to cities to find better jobs [13]. For example,  
9 we calculated that for 2050 around 166 urban people per km<sup>2</sup> of the total studied area (Table S1), which is lower  
10 than in 2000 (around 92 urban people per km<sup>2</sup> in 2000, Table S1). Most densely populated urban areas are  
11 projected for the downstream sub-basins with big cities such as Beijing (the Hai sub-basin, 330 urban people km<sup>-2</sup>  
12 <sup>2</sup>), Shanghai (the Changjiang delta sub-basin, 619 urban people km<sup>-2</sup>), Shenzhen and Guangzhou (the Zhujaing  
13 delta sub-basin, 342 urban people km<sup>-2</sup>). The same increasing trends are projected for population with a sewage  
14 connection (Table S1). In this world more people are assumed to be connected to sewage systems because of  
15 investments in better infrastructure. However, sewage treatment may still be low to reduce inputs of nutrients to  
16 rivers from sewage systems [10, 13].

17 In the globalized scenario Chinese agriculture is projected to intensify for food security reasons. Agricultural  
18 areas in China might be slightly reduced between 2000 and 2050 [10] (see also Table S1). A possible reason of  
19 this can be the side-effect of urbanization (more cities requiring more land). However, Strokal, et al. [10]  
20 calculated that applications of, for example, synthetic fertilizers to grow more crops will increase for Chinese  
21 basins between 2000 and 2050 under this globalized scenario. Animal manure production is also projected to  
22 increase (almost double for 16 Chinese basins according to [10]) due to an increase in numbers of animals [12].

23 The drawback of this scenario is that it does not assume direct losses of animal manure to Chinese rivers in the  
24 future. It assumes that all available manure (after correcting for losses during storage and housing) will be  
25 applied on land. Considering the development of industrial farms today and poor manure management it is likely  
26 that direct discharges of animal manure to rivers will occur in the coming years without efficient nutrient  
27 management. Thus, in this study we assumed that these direct discharges of manure will occur in 2050. To this  
28 end, we used the fractions of manure directly discharged to rivers calculated for 2000.

29 *Model performance.* First, we discuss uncertainties in the original Global NEWS-2 and NUFER models because  
30 we used these models as the basis to quantify TDN and TDP inputs to rivers at the sub-basin scale. Next, we  
31 elaborate on uncertainties in our model results.

32 ○ Evaluating the original Global NEWS-2 and NUFER

33 The uncertainties in the original Global NEWS-2 model were addressed in earlier studies [1, 11, 12, 16, 19].  
34 Mayorga, et al. [1] calibrated and validated the model for large world basins. Model parameters (e.g., parameters  
35 used to calculate annual runoff from land to streams and EC<sub>F</sub>, see equations 3 and 4 above) were calibrated using  
36 observations at the river mouths. Values for model parameters were selected based on the Nash-Sutcliffe model  
37 efficiency (R<sub>NSE</sub><sup>2</sup>) [20]. The model was validated by comparing results with the other set of observations at the  
38 river mouths. The validation results of dissolved N and P forms indicate an acceptable model performance  
39 according to R<sub>NSE</sub><sup>2</sup> [20]: R<sub>NSE</sub><sup>2</sup> of DIN is 0.54, R<sub>NSE</sub><sup>2</sup> of DIP is 0.51, R<sub>NSE</sub><sup>2</sup> of DON is 0.71, and R<sub>NSE</sub><sup>2</sup> of DOP is  
40 0.90. The original Global NEWS-2 has been applied in many studies to quantify river export of nutrients for past  
41 (1970, 2000) and future (2030, 2050) years and their impacts on coastal waters for African [21] and South  
42 American [22], European [23], Indonesian [24, 25] and Chinese [10, 26, 27] basins, for the Black Sea [28] and  
43 Bay of Bengal [29, 30] regions. Global NEWS-2 is thus widely accepted model to analyze nutrient pollution of  
44 aquatic systems at global [1, 15], continental [19, 21, 22] and basin [23, 31, 32] scales. Furthermore, the model  
45 was further validated for the Chinese rivers such as the Changjiang [32], Zhujiang, Huang, Liao (covering the  
46 study area) [10], using three indicators: Pearson's coefficient of determination (R<sub>P</sub><sup>2</sup>), R<sub>NSE</sub><sup>2</sup>, and Model error  
47 (ME). Yan, et al. [32] validated the model for DIN export by the Changjiang River for the period of 1970-2002.  
48 Their results indicate a good performance of the model (R<sub>P</sub><sup>2</sup> = 0.93). Strokal, et al. [10] validated the model for

1 DIN and DIP export by large Chinese rivers for 2000. The results indicated a satisfactory performance of the  
2 model ( $R_p^2 = 0.96$ ;  $R_{NSE}^2 = 0.42$ ;  $ME = 18\%$ ).

3 The NUFER model was developed for China. NUFER is a widely accepted model because it has been applied in  
4 many studies to quantify N and P use efficiencies in the food chain at national [18, 33-35] and provincial [2]  
5 scales over time (e.g., the period of 1980-2010 [2, 35]). This model is also used to assess management options to  
6 improve nutrient management and thus to increase N and P use efficiencies in the food chain (covering crop and  
7 animal production, food processing, households) [36, 37]. The NUFER model was developed based on three  
8 reliable sources of information [18]: statistical data (e.g., to obtain animal numbers) [38, 39], field surveys of  
9 about 50 thousand farms during 1999-2008 (e.g., to obtain sizes of farms, manure management information) [40]  
10 and literature (e.g., to obtain N and P excretion per animal category, see details in Ma, et al. [18]). These sources  
11 of information give us confidence of using information from NUFER for direct manure discharges.

12 ○ Evaluating the sub-basin model results

13 We addressed the uncertainties in our modeling results in different ways. *First*, we validated modeling results by  
14 comparing them with collected available information from open literature. We compared model results to  
15 observations available in the literature. However, not many studies report average concentrations of N and P in  
16 Chinese rivers at the sub-basin scale. Concentrations of nutrients are often measured at monitoring or  
17 hydrological stations (e.g., located at the outlets of sub-basins). Various studies presented nutrient concentrations  
18 in the estuaries of the Huai, Hai, Liao [41, 42], Huang [41-46], Changjiang [41, 43, 45-49], and Zhujiang [41, 43,  
19 45, 46, 50-54]. Several studies analyzed trends in N and P concentrations from individual stations of rivers,  
20 mainly the Changjiang River [55], [47, 56-59]. For example, empirical studies by Wang [56], and Li, et al. [59]  
21 indicate large increases in N (approximately a 5-fold) and P (around 5-10-fold) inputs to surface waters of the  
22 Changjiang River between 1960s-2000. Tao, et al. [55] indicated an increase in nutrient fluxes at the downstream  
23 stations of the Huang River since the 1970s, in particular for nitrogen. These reported trends are in line with our  
24 calculations showing large increases in the total N and P inputs in these rivers (Figure 3; Figure S8; Table 1).  
25 Xu, et al. [60] indicated that today Chinese rivers are polluted, with higher concentrations of nutrients in  
26 northern than in southern rivers. This is in line with our model results. We calculate that N and P inputs in the  
27 northern rivers (e.g., Huai, Huang) increase faster than in the central and southern rivers (e.g., Changjiang,  
28 Zhujiang). Furthermore, the northern downstream rivers of the Huai, Huang delta, Hai sub-basins have generally  
29 higher calculated loads of N and P than, for example, upstream rivers.

30 Some studies quantified N and P transport to waterbodies of the large Chinese rivers [61], in particular of the  
31 Changjiang River [32, 62-66]. A few studies focused on sub-basin analyses of the Changjiang River [62, 63, 66].  
32 But these studies do not include direct discharges of animal manure to surface waters. Nevertheless, we  
33 compared these measurements with our results. We calculate inputs to rivers of around 5.3 Tg N, 1 Tg N and 1.7  
34 Tg N in the Changjiang, Huang and Zhujiang, respectively in 2000 (the sum of the total dissolved N inputs to  
35 their sub-basins; see Figure S8). Xing, et al. [61] quantified around 3.8 Tg N, 0.8 Tg N and 0.8 Tg N transported  
36 to waterbodies of the Changjiang, Huang and Zhijiang, respectively in 1995. Their values are lower than ours,  
37 which could be because they ignore direct manure discharges to the rivers. Furthermore, Xing, et al. [61] used  
38 an export coefficient approach to calculate inputs of N to the rivers from land without considering sub-basin  
39 characteristics. Liu, et al. [62] and Bao, et al. [63] quantified around 2 Tg N transported to waterbodies of the  
40 Changjiang River in 1980, which is close to our estimate of 1.7 Tg TDN (the sum of inputs to rivers of the sub-  
41 basins) in 1970. For the year 2000 Liu, et al. [62] quantified approximately 4.5 Tg of N transported to  
42 waterbodies of the Changjiang River, where around 0.4 Tg N is from animal manure use in agriculture. We,  
43 however, calculated 5.3 Tg TDN, where 2.2 Tg TDN from direct discharges of animal manure and 0.4 Tg N  
44 from animal manure use (as the sum of inputs to surface waters of the sub-basins of this river; Figure S8). This  
45 comparison shows that our results are close to the results of Liu, et al. [62] and Bao, et al. [63] for 1970, but  
46 higher than the results of Liu, et al. [62] and Xing, et al. [61] for 2000. A possible reason is that those studies do  
47 not account for direct discharges of animal manure, which was limited in the 1970s (Figures 1 and 2), where our  
48 estimates are comparable with their estimates. Ti, et al. [67] calculated an N budget for main land of China. This  
49 study adopted a 25% value for livestock waste discharged to waterbodies to quantify N export by the  
50 Changjiang, Zhujiang and Huang for the 2000s. We, however, demonstrate that this percentage is much higher  
51 for 2000 and ranges between 30% and 45% for N (Figure 2; Figure S3).

1 *Second*, we compared our model inputs with an independent county-based dataset. We used global gridded  
2 datasets to derive most of the model inputs for sub-basin modeling of nutrient inputs to rivers (see Figure S7).  
3 These gridded datasets were developed using, for example, national assessments (e.g., FAO) [11-13]. We  
4 analyzed the differences between our gridded data and independent county data (Figure S9). County data are  
5 largely based on local statistics provided by Data Center for Resources and Environmental Sciences of Chinese  
6 Academy of Sciences [68]. We compared the following model inputs: N and P synthetic fertilizers, N and P  
7 animal excretion, biological N fixation by agricultural crops, human population and sub-basin areas. These  
8 model inputs were selected based on the availability of county data. The county data were aggregated to sub-  
9 basins in ArcGIS and then used for comparison (see Figure S9 for details). Results of this comparison show a  
10 good agreement between our sub-basin inputs derived from the gridded datasets and sub-basin inputs derived  
11 from the county dataset. For example,  $R_p^2$  is in between 0.73 and 0.98 for the studied rivers for N and P animal  
12 manure and synthetic fertilizers. For the other model inputs of Figure S9  $R_p^2$  is above 0.90, except for biological  
13  $N_2$  fixation by crops in the central and southern rivers ( $R_p^2$  is 0.56).

14 *Third*, we checked some model parameters with local information and experts. For example, we used a  
15 parameter for nitrogen and phosphorus removal during treatment in the Dongjiang basin based on expert  
16 knowledge. This basin drains into Hong Kong. The areas experience high economic development and thus high  
17 potentials for efficient treatment of sewage influents. Another example are the fractions of nutrients in animal  
18 manure that are directly discharged to surface waters. We used information for these fractions from NUFER. We  
19 discussed these fractions with experts from local Chinese institutions (e.g., Chinese Academy of Sciences,  
20 Chinese Agricultural University, Peking University) to increase our confidence in the magnitude of manure  
21 pollution of water bodies in China.

22 We consider our model appropriate for modeling N and P inputs to the Chinese rivers at the sub-basin scale for  
23 three main reasons (see details in Supplementary Methods). *First*, our modeled nutrient pollution levels in rivers  
24 are generally in line with observations (see references above). This pollution today might be underestimated in  
25 existing modeling studies because of ignoring direct discharges of animal manure. We argue that the impact of  
26 direct discharges of animal manure from livestock production on river quality is much larger than the impact of  
27 over-fertilization of soils by synthetic fertilizers. Our model indicates that the northern rivers suffer from  
28 manure-related pollution more than the central and southern rivers because of the dry northern environment  
29 making point sources main contributors to nutrient pollution. This is in line with experimental studies reporting  
30 on severe pollution of the northern rivers. Our model captures the increasing trends in dissolved N and P inputs  
31 to the Chinese rivers since 1970, which is in agreement with other studies (see above references). We also  
32 illustrated that the river pollution may increase fast in the future if the management of animal manure stays as  
33 during the ongoing transitions. *Second*, the comparison of model inputs with independent county data (Figure  
34 S9) convincingly shows that our model input for sub-basin is of good quality (e.g.,  $R_p^2$ : 0.73-0.98 for studied  
35 rivers N and P synthetic fertilizers and animal manure). *Third*, we verified relevant model parameters with  
36 experts and local information (e.g., nutrient removal during treatment in the Dongjiang basin, fractions of direct  
37 manure discharges from NUFER).

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1 **2. Tables S1-S2**

2 **Table S1. Main drivers of nutrient inputs to the studied rivers in 1970, 2000 and 2050.** The year 2050 is  
 3 based on Global Orchestration (GO) of the Millennium Ecosystem Assessment scenarios and represents the  
 4 globalized scenario with reactive environmental management [14, 15]. The range in brackets shows minimum  
 5 and maximum values of the sub-basins. These drivers were derived from gridded Global *NEWS-2* database [1,  
 6 15]. See the Supplementary Materials for model description and inputs.

<b>Drivers</b>	<b>1970</b>	<b>2000</b>	<b>2050 GO</b>
Income, GDP at purchase power parity (1995 US\$ 1000 inh <sup>-1</sup> ) <sup>a</sup>	0.46 <sup>*</sup>	3.6 <sup>*</sup>	38.5 <sup>*</sup>
Total human population (inh km <sup>-2</sup> ) <sup>b</sup>	159 (22-443)	224 (34-680)	245 (34-682)
Population connected to sewage systems (inh km <sup>-2</sup> ) <sup>b</sup>	9 (0-96)	51 (0-347)	133 (0-522)
Urban population (inh. km <sup>-2</sup> ) <sup>b</sup>	28 (0-224)	92 (0-445)	166 (0-671)
Agricultural area (fraction of the total area, 0-1) <sup>c</sup>	0.49 (0-0.91)	0.76 (0.48-0.99)	0.71 (0.26-0.95)

7 <sup>\*</sup>No range. <sup>a</sup>Income per inhabitant was calculated as income from all population (sum of people from all sub-  
 8 basins) divided by this total population. <sup>b</sup>the total human population per km<sup>-2</sup> of the studied area was calculated  
 9 as population from all sub-basins (people) divided by the total area of all sub-basins. The same holds for  
 10 population connected to sewage systems and urban population. <sup>c</sup>The fraction of agricultural area was calculated  
 11 as the sum of agricultural areas of all sub-basins (km<sup>2</sup>) divided by the total area of these sub-basins.

12



1 **Table S2. Area-weighted averaged total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to**  
2 **Chinese rivers in 2050** (kg km<sup>-2</sup> of basin year<sup>-1</sup>, and share of the animal manure point source in %). The range in  
3 brackets shows minimum and maximum values of the sub-basins. The year 2050 is based on Global  
4 Orchestration (GO) of the Millennium Ecosystem Assessment scenarios and represents the globalized scenario  
5 with reactive environmental management [14, 15]. See the Supplementary Materials for model description and  
6 inputs. Other diffuse sources include biological N<sub>2</sub> fixation, atmospheric N deposition, weathering of P-  
7 contained minerals, and N and P leaching from organic matter.

<b>Nutrient sources in rivers</b>	<b>TDN</b>	<b>TDP</b>
Animal manure (point)	2180 (685-6378)	457 (138-1436)
Animal manure (diffuse)	340 (1-1277)	10 (0-46)
Synthetic fertilizers (diffuse)	826 (1-3422)	48 (0-236)
Other diffuse sources for agricultural areas	564 (3-2300)	5 (0-18)
Other diffuse sources for non-agricultural areas	368 (1-2104)	2 (0-10)
Sewage effluents (point)	408 (0-1697)	114 (0-473)
Total	4687 (900-10900)	636 (161-1913)
<i>The share of point animal manure (%)</i>	<i>47 (22-90)</i>	<i>72 (51-91)</i>

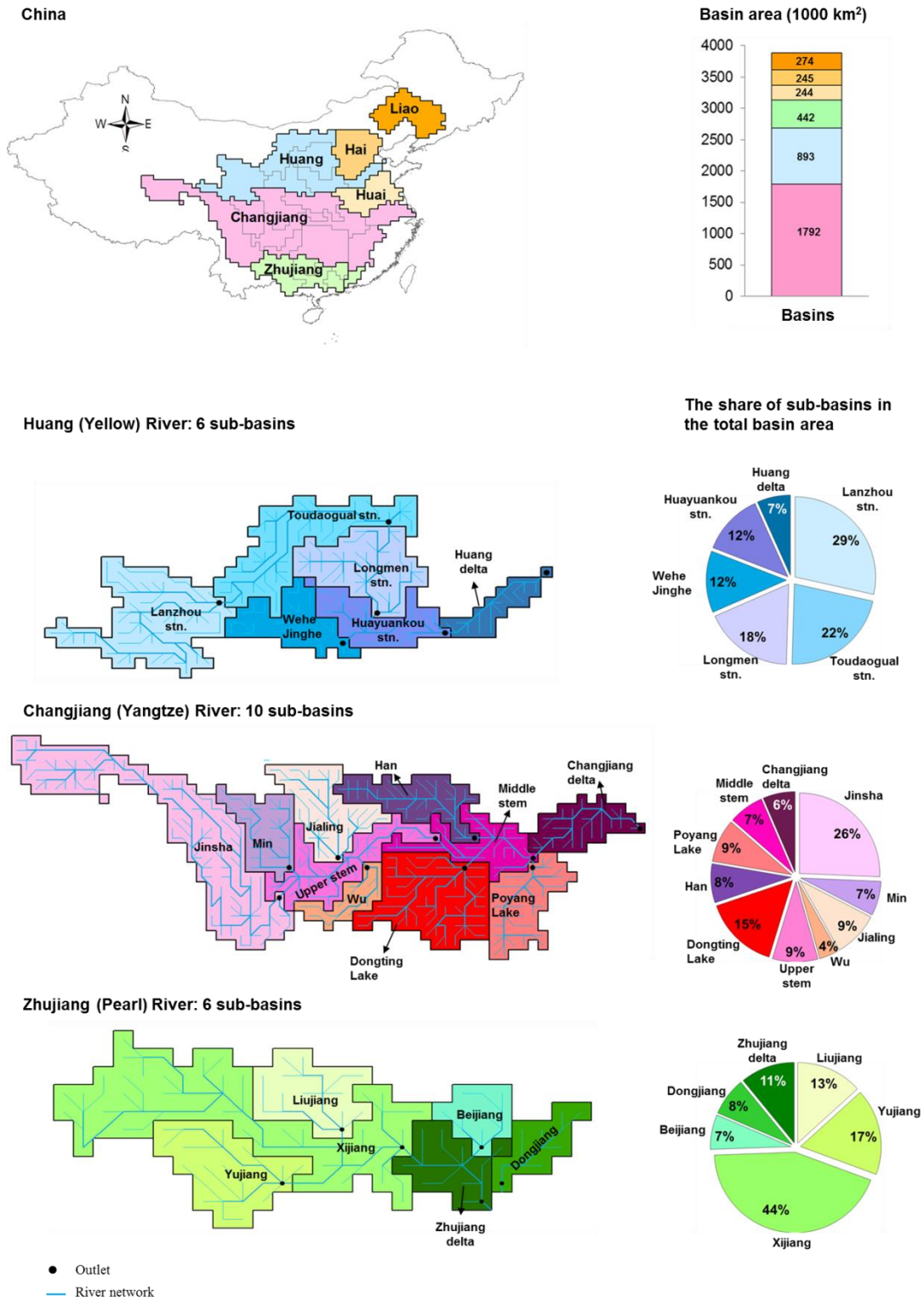
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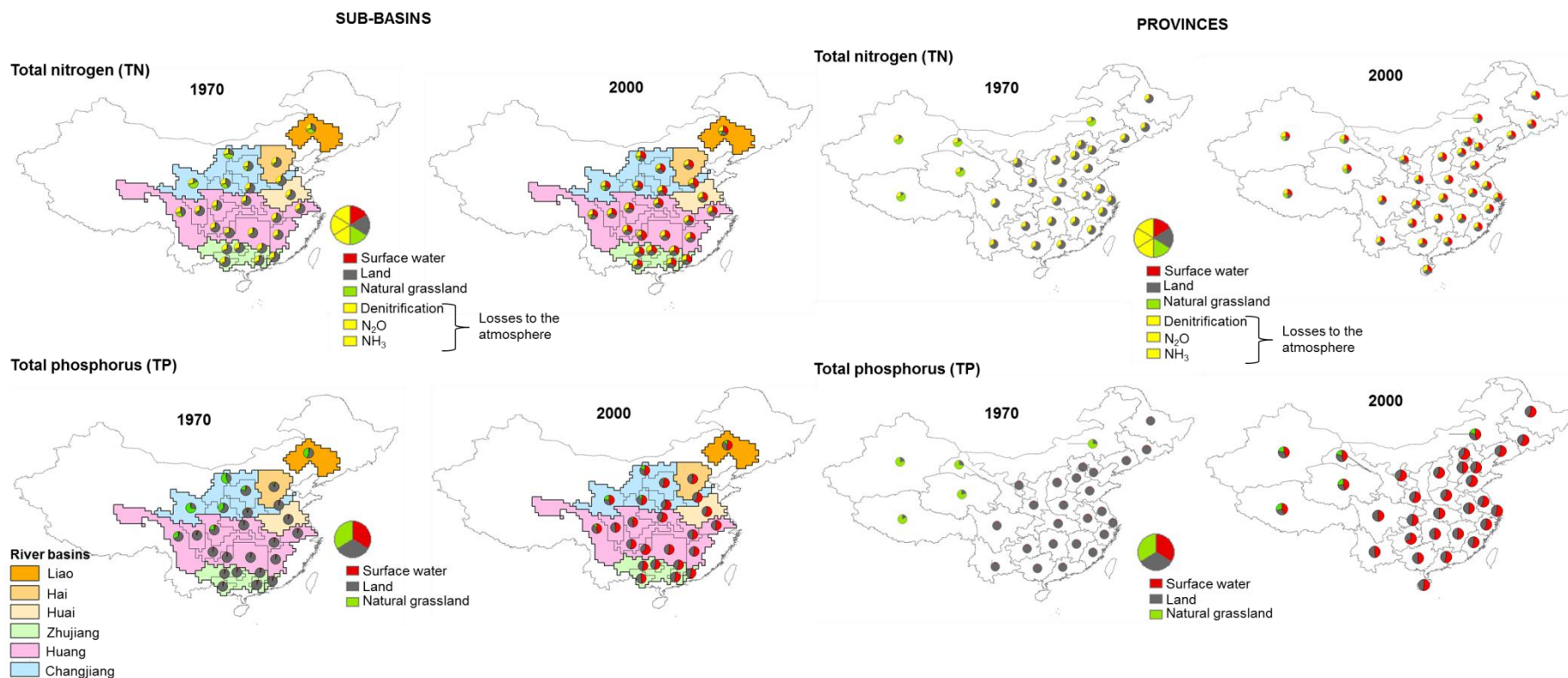
1 **3. Figures S1-S9**

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4 **Figure S1. Locations (maps) and areas (bar and pie graphs) of the six major rivers and their sub-basins in**  
 5 **China.** Sub-basins are delineated based on the Topological Simulated Network (STN-30 v6.01), applied to the  
 6 Global *NEWS-2* model [1, 9]. Various sources were used to delineate the main sub-basins of the Huang [3, 4],  
 7 Changjiang [5, 6] and Zhujiang [7, 8] rivers. Sub-basins are named after rivers (or lakes) at their outlets or after  
 8 hydrological (monitoring) stations at their outlets.

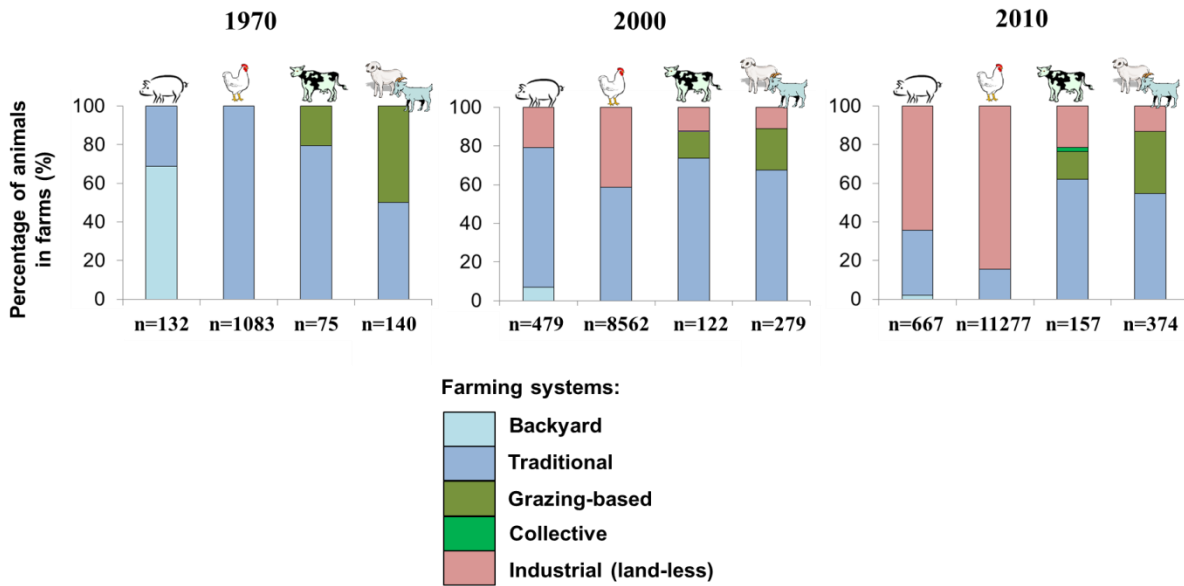


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2 **Figure S2. Fate of total nitrogen (TN) and phosphorus (TP) in animal excretion by sub-basin and by province in 1970 and 2000.** The pies show the fractions of the  
 3 nutrients in animal excretion that are directly discharged to surface waters, applied to land and natural grassland, and lost to the atmosphere due to housing and storage (only  
 4 for N) for sub-basins of major rivers in China in 1970 (close to 1980) and 2000 (close to 2005). The fractions for sub-basins are area-weighted averages, calculated using the  
 5 provincial information in ArcGIS (Figure S7). The provincial information was calculated by the NUFER model [2, 18, 37] on the basis of various farming surveys and  
 6 statistical yearbooks as the basis [2, 18, 37]. The maps show that more than half of TN in animal manure and over two-thirds of TP in animal manure were applied to cropland  
 7 in 1970 in the sub-basins expect for several upstream sub-basins of the Huang River and Liao River. Very small part of the nutrients in animal manure discharged directly to  
 8 rivers in 1970. In 2000 application of manure to land decreased compared to 1970 and about 30-70% of the TN and TP in animal manure were discharged directly to rivers in  
 9 the sub-basins in 2000.

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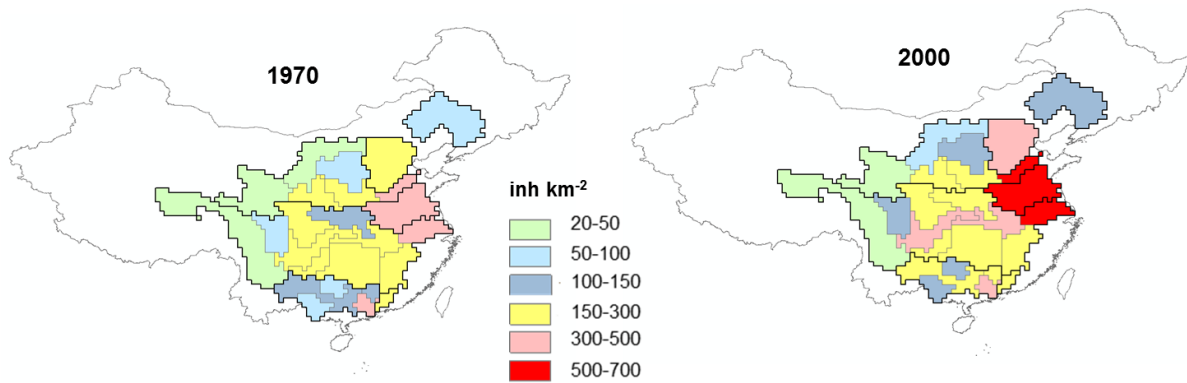


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3 **Figure S3. Percentage of animals in different farming systems in China for 1970, 2000 and 2010.** Animals  
 4 include pigs, poultry (layers and broilers), cattle (dairy and beef), sheep and goat (n, million heads). Information  
 5 was derived from [33, 69-71]. In 1970, animal production was dominated by traditional (and backyard for pigs)  
 6 farms. That time industrial farms did not exist. Traditional farms combine crop with animal production.  
 7 Recycling of animal manure is a common practice in these farms. The number of animals is usually below 50 for  
 8 pigs, 500 for layer poultry, 2000 for broiler poultry, 9 for dairy cattle, 50 for beef cattle, and 1000 for sheep and  
 9 goat. Industrial animal farms started to emerge since the 1980s (examples here for 2000 and 2010). Industrial  
 10 farms are different from traditional. These farms are land-less and thus disconnected from crop production. The  
 11 large part of animal manure is discharged to surface waters because of poor manure management. Small part of  
 12 animal manure is sometimes exported to outside vegetable and fruit farms. The animal number for industrial  
 13 farms with medium size is usually about 50-3000 for pigs, 500-10000 for layer poultry, 2000-50000 for broiler  
 14 poultry, above 200 for dairy cattle, above 50 for beef cattle and above 100 for sheep and goat. The animal  
 15 number for industrial farms with large size is above 3000 for pigs, 10000 for layer poultry and 50000 for broiler  
 16 poultry [70, 71].

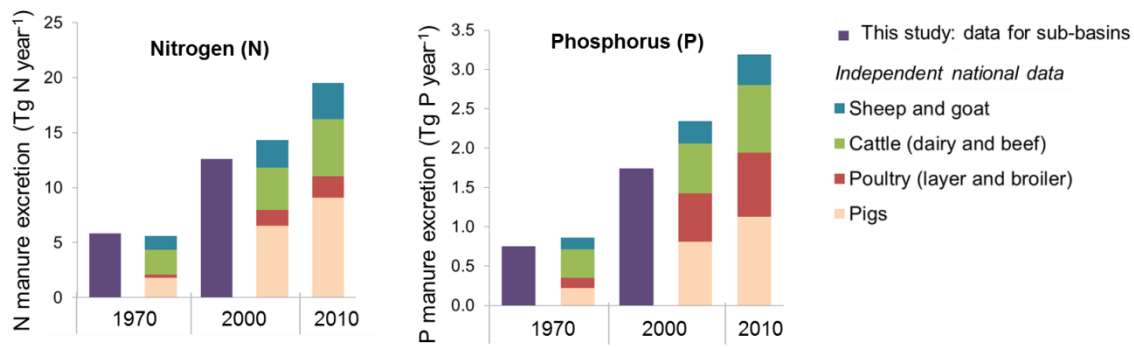
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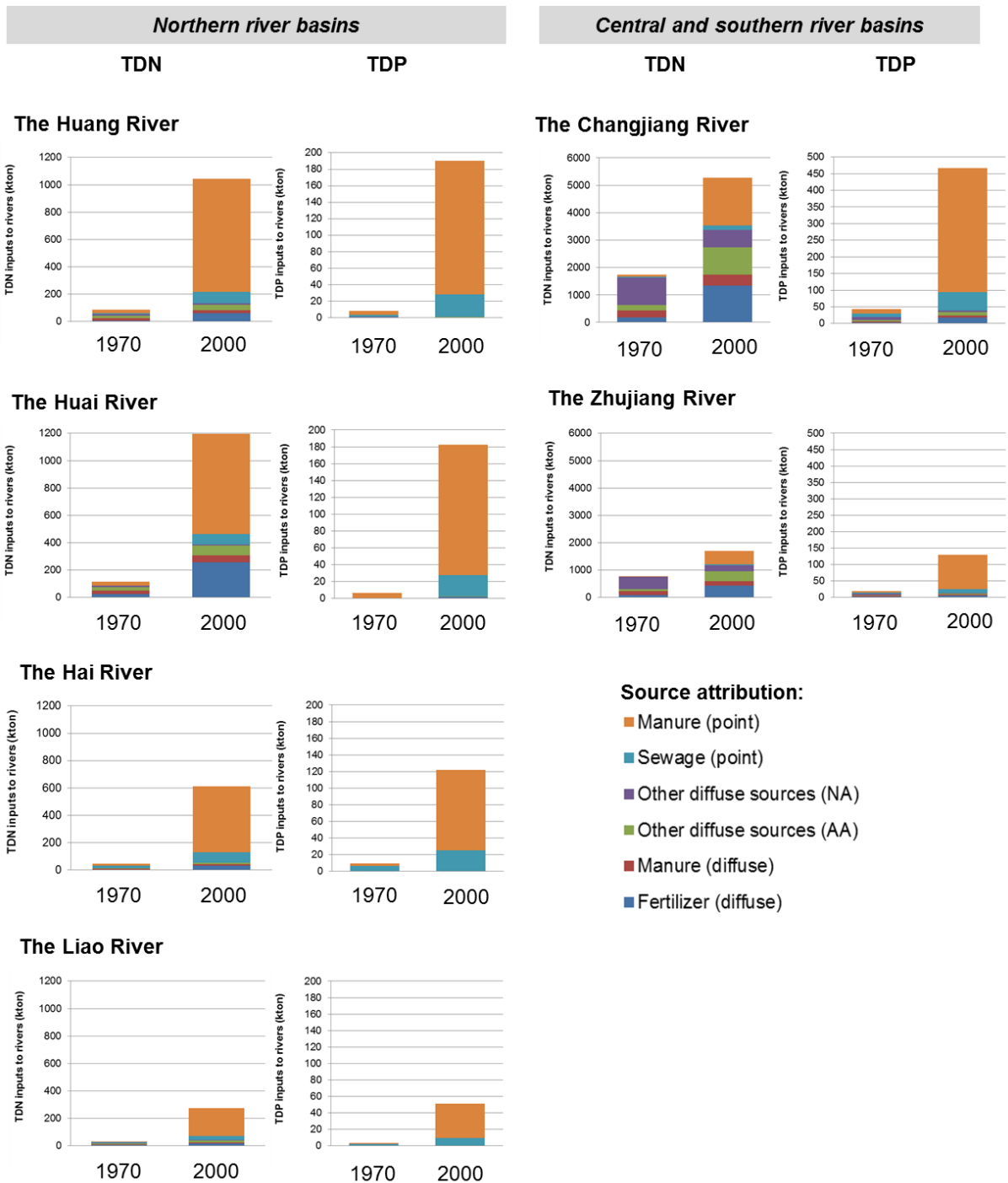
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**Figure S4. Human population density (inh km<sup>-2</sup>) of the sub-basins in 1970 and 2000.** Information is from the Global *NEWS-2* model [1] (see Figure S7). The results show that human population in sub-basins increased largely between 1970 and 2000. The most populated areas (500-700 inh. km<sup>-2</sup>) are located close to the coastal waters and occupy downstream sub-basins of the Huang, Changjiang and Huai (see Figure S1 for locations of the rivers).



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2 **Figure S5: The total nitrogen (N) and phosphorus (P) excretion by animals in 1970, 1980, 2000 and 2010**  
3 **(Tg year<sup>-1</sup>).** This study includes 25 sub-basins (Figure S1) covering the majority of agricultural areas in China  
4 with animal production [69]. Manure excretion of this study is from Global *NEWS-2* [1] (see Methods and  
5 Figure S7). Information from national data for 1970, 2000 and 2010 is for the entire China. N and P animal  
6 excretion for China was calculated from animal numbers derived from Bai, et al. [69], and FAO [70], multiplied  
7 by N and P excretion rates derived for each animal category from Ma, et al. [2]. The graphs show an increasing  
8 trend in production of animal manure between 1970 and 2010. The reasons for this increase are increasing  
9 number of animals[33, 69] and transitions in farming systems from traditional-dominated in the 1970s to  
10 industrial-dominated in 2010 (Figure S3). Pig and poultry (broilers and layers) manure accounts for over half of  
11 the total manure production in 2010. Over two-thirds of pigs and poultry are kept today in industrial farms,  
12 which lack facilities to treat or recycle animal manure [72]. As a result, considerable amounts of manure are  
13 discharged directly to surface waters (Figure S2; Figures 1-3 in the main text).

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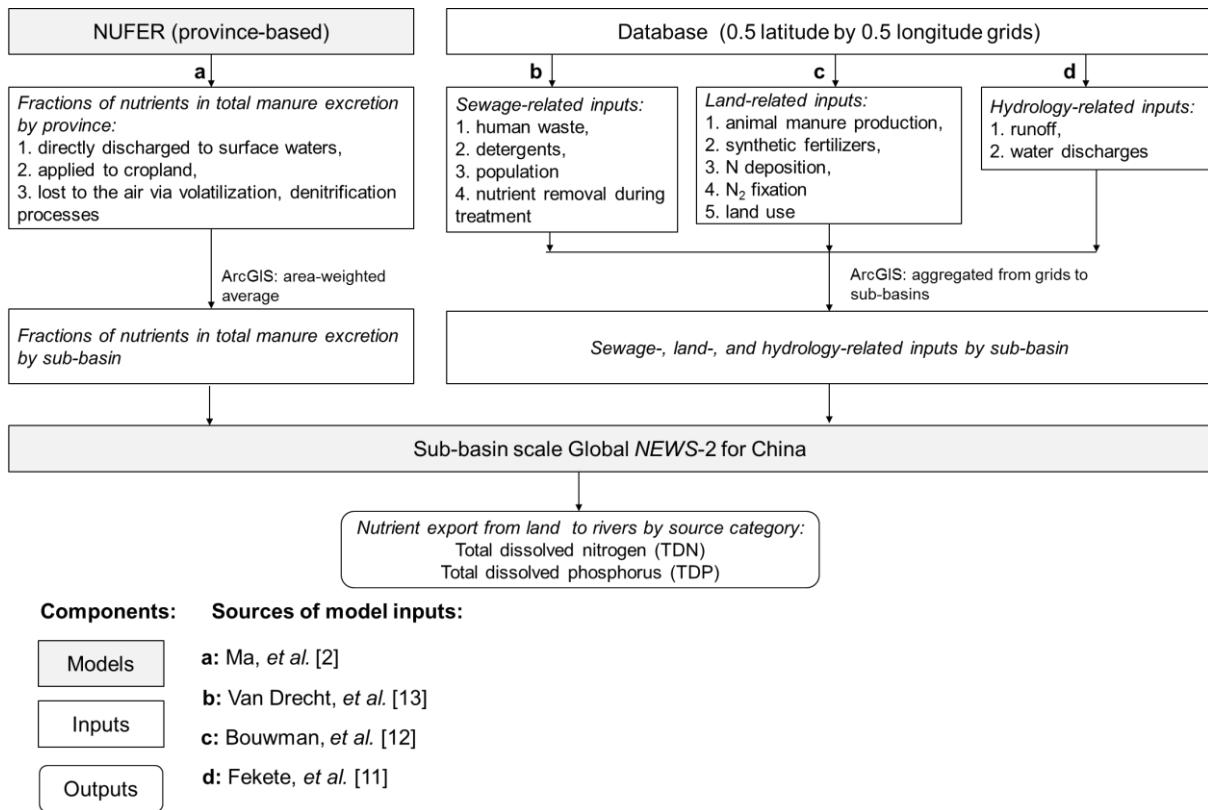
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2 **Figure S6. Total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to the Chinese rivers by source**  
3 **in 1970 and 2000 (kton).** For large rivers, the Huang, Changjiang and Zhujiang, this is the sum of the TDN and  
4 TDN and TDP inputs to rivers of their sub-basins (Figure S1). AA and NA are agricultural and non-agricultural areas,  
5 respectively. For TDN other diffuse sources (AA, NA) include biological N<sub>2</sub> fixation, atmospheric N deposition  
6 over and leaching of organic matters from agricultural (AA) and non-agricultural (NA) areas; for TDP these are  
7 weathering of P-contained minerals and leaching of organic matters from agricultural (AA) and non-agricultural  
8 (NA) areas. TDN and TDP inputs were quantified based on a sub-basin modeling approach, applied to Global  
9 NEWS-2 (see Methods and Figure S7). Results show that TDN and TDP inputs to the Chinese rivers increased  
10 by a factor of 2 to 30 between 1970 and 2000. Animal manure became a dominant point source of these nutrients  
11 in rivers in 2000. The year 2000 reflects the changes in agriculture since the 1980s. For the central and southern  
12 rivers (Changjiang and Zhujiang) the share of point manure source to TDN inputs in 2000 is lower than for the  
13 other rivers. This is the net effects of several factors such as hydrology and differences in soil retention between

1 N and P. For example, runoff influences transport of nutrients from land to rivers. The drainage areas of the  
2 Changjiang and Zhujiang are characterized by higher runoff than the drainages areas of the other northern rivers  
3 [10]. As a result, more nutrients enter rivers from diffuse sources in the central and southern rivers than in the  
4 northern rivers. Furthermore, N retention in soils is generally lower than P retention because P has a strong  
5 ability to accumulate [10, 73].

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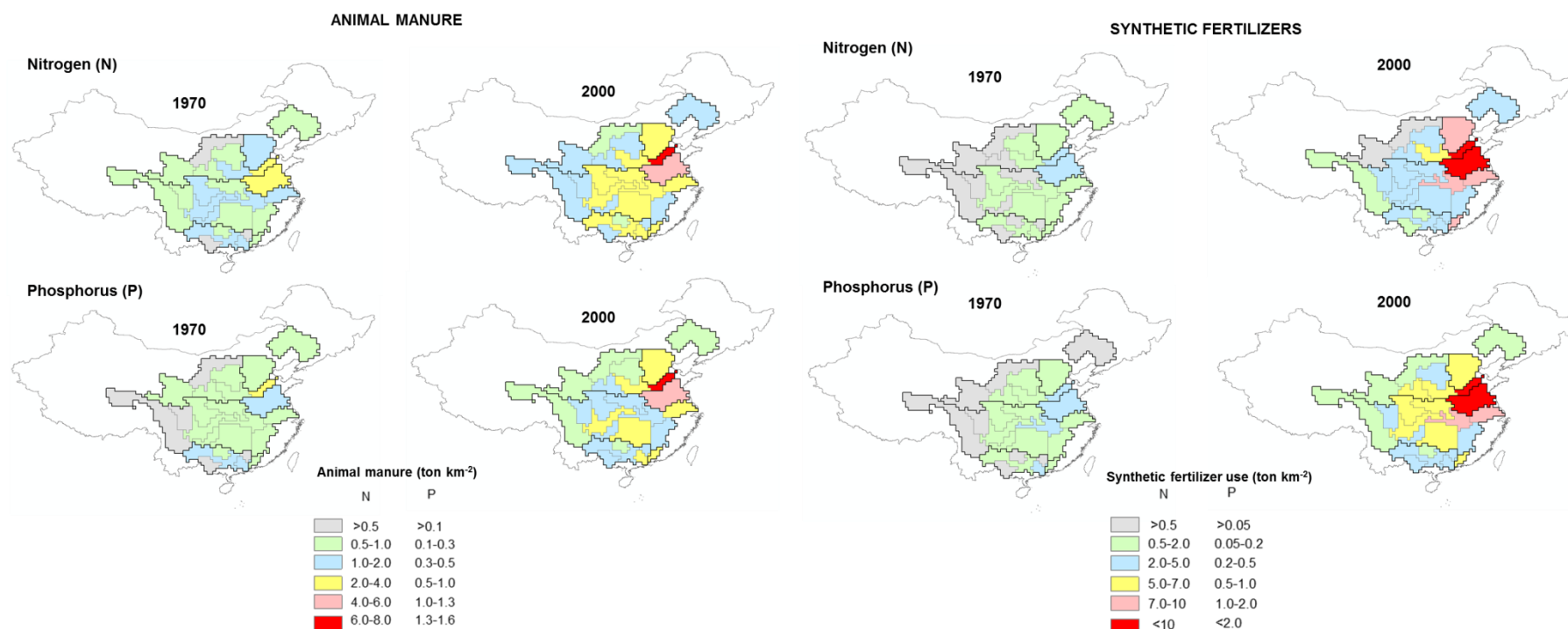
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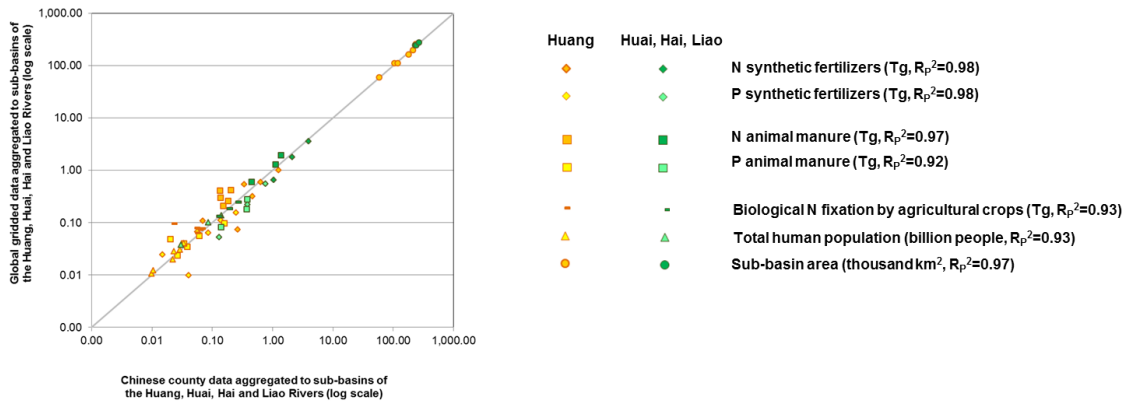
**Figure S7. Inputs and outputs of the sub-basin scale Global NEWS-2 model, combined with information from the NUFER model.** Global NEWS-2 is short for **N**utrient **E**xport from **W**ater**S**heds, and NUFER for **N**utrient flows in **F**ood chains, **E**nvironment and **R**esources use. The original Global NEWS-2 [1] did not consider direct discharges of nutrients from animal manure to rivers, and the model was not applied on a sub-basin scale. We ran the model at the sub-basin scale with direct inputs of animal manure to rivers (fractions of the total animal manure that is directly discharged to rivers; see these values in Figure S2). These direct inputs for each sub-basin were calculated from provincial data (see Figure S2) by the NUFER model [2]. Other required inputs to calculate nutrient export from land to rivers of sub-basins were derived from the gridded database (0.5 latitude by 0.5 longitude). Sewage-related inputs were prepared by Van Drecht, *et al.* [13], land-related inputs by Bouwman, *et al.* [12] and hydrology-related inputs by Fekete, *et al.* [11]. These inputs were also used in the original basin scale Global NEWS-2 model [1]. See Methods for detailed description of calculating nutrient export from land to rivers of sub-basins by source category.



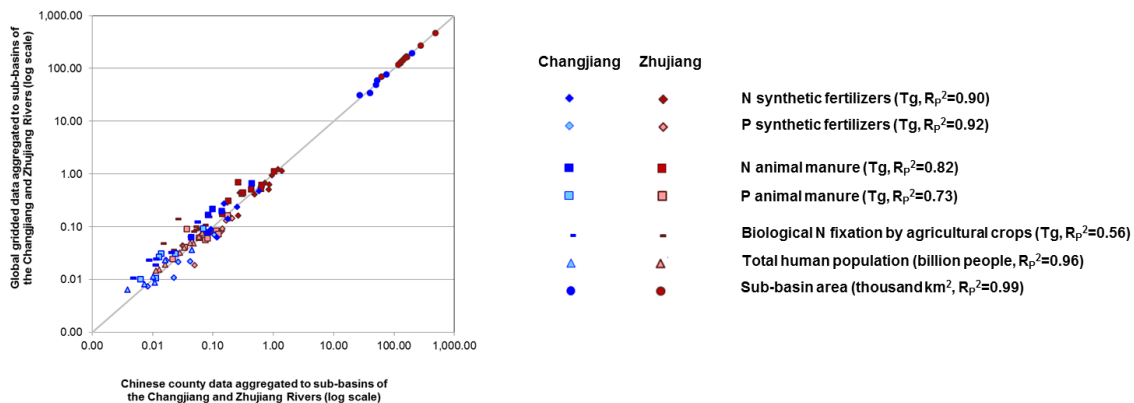
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2 **Figure S8. Nitrogen (N) and phosphorus (P) in animal manure and in synthetic fertilizers (ton km<sup>-2</sup>) by sub-basin in 1970 and 2000.** Animal manure and synthetic  
3 fertilizers were taken from Global NEWS-2 [1, 12] (see Methods and Figure S7). Animal manure in Global NEWS-2 was already corrected for N losses during storage and  
4 housing. Animal manure was prepared by Bouwman, et al. [12] for Global NEWS-2 (see Methods and Figure S7). Maps show that N and P in animal manure increased  
5 between 1970 and 2000. This is a result of increasing numbers of animals in China to meet the demand for meat (see Figures S3 and S5 as an example). Nutrient inputs to  
6 land from synthetic fertilizers are also higher in 2000 than in 1970. Higher values are observed for areas that are located closer to the coastal waters (e.g., Huai sub-basin; see  
7 Figure S1 for locations of the sub-basins). Those areas are largely dominated by cropland [74] and synthetic fertilizer applications are thus larger than in the other areas.

### The northern rivers and their sub-basins



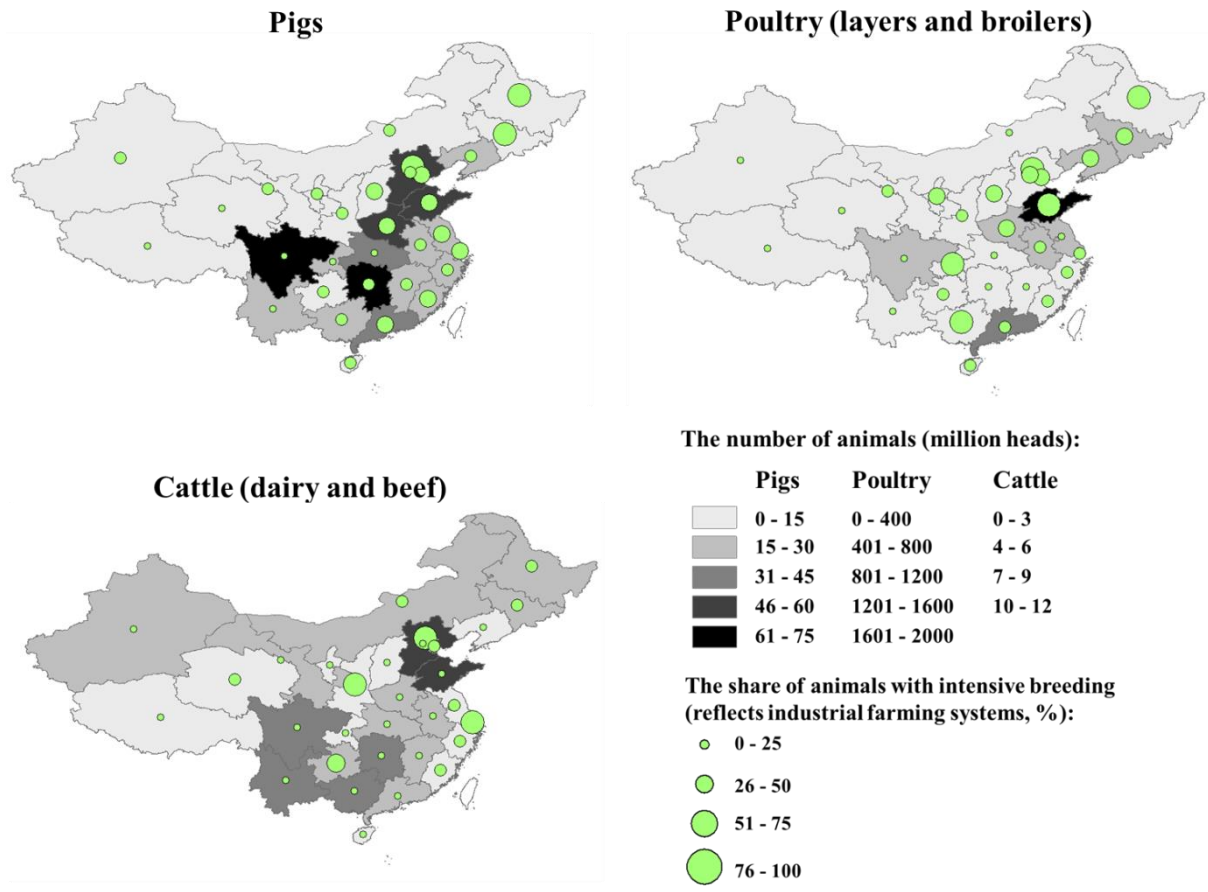
### The central and southern rivers and their sub-basins



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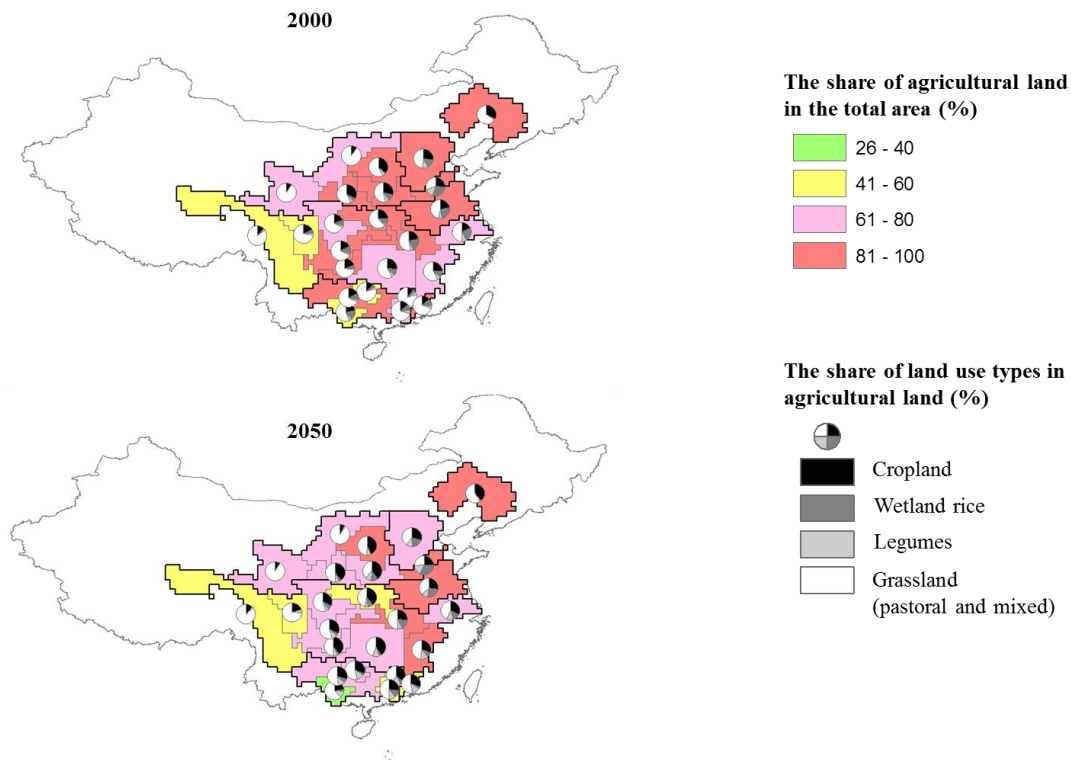
2 **Figure S9. Gridded data compared to county data that were aggregated to sub-basins of the northern**  
 3 **(Huang, Huai, Hai, Liao), central (Changjiang) and southern (Zhujiang) rivers for the year 2000.**  
 4 Comparison is for the following model inputs: N and P synthetic fertilizers (Tg), N and P animal manure  
 5 excretion (Tg), Biological N<sub>2</sub> fixation by agricultural crops (Tg), total human population (billion people)  
 6 and sub-basin areas (thousand km<sup>2</sup>). R<sub>p</sub><sup>2</sup> in brackets is the Pearson's coefficient of determination (the square of the  
 7 correlation coefficient). The drainage areas of the Huang, Changjiang and Zhujiang rivers are divided into six,  
 8 ten and six sub-basins, respectively. The drainage areas of the Huai, Hai and Liao rivers are individual sub-  
 9 basins (see Figure S1). The gridded data were aggregated to sub-basins and used in this study for modeling total  
 10 dissolved N and P inputs to rivers of these sub-basins (see Figure S7 for sources). County data [68] provided  
 11 information on the number of animals, human population, synthetic fertilizer use, and sown areas per county. N  
 12 and P animal excretion per county was calculated using the number of animals and N and P excretion rates per  
 13 animal category (e.g., pigs, cows, poultry) from Ma, et al. [2]. Biological N fixation by agricultural crops per  
 14 county was calculated using sown areas and N fixation ratios per crop category (e.g., beans, peanut) from Ma, et  
 15 al. [2]. These calculations were performed by Wang M. (pers. comm.) using the NUFER model [2]. Next, this  
 16 county information was then aggregated to sub-basins using ArcGIS functions. The areas of counties were  
 17 calculated in ArcGIS and aggregated to sub-basins.

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**Figure S10. The number of animals (million heads) and the share of animals with the intensive breeding (reflects industrial farming systems, %) by province in 2005.** Information is from the NUFER model [2]. In NUFER animal production (see details in [2]) includes two intensities of animal breeding, reflecting traditional and industrial farming systems. The high intensity types of animal breeding are defined for pig systems (> 50 capita), dairy cattle (> 5 capita), beef cattle (> 50 capita), layer poultry (>500 capita), and meat poultry (> 2000 capita).



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**Figure S12. Agricultural land in sub-basins in 2000 and 2050 (%).** Legume crops include pulses and soybean. Cropland includes other crops such as cereals and maize. Information is from the gridded data of the Global *NEWS-2* model [1, 12]. The total areas of the basins and the share of sub-basins in the basin area are given in Figure S1. The year 2050 is based on Global Orchestration (GO) of the Millennium Ecosystem Assessment scenarios and represents the globalized scenario with reactive environmental management [14, 15].

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