

# Multi-scale Modeling of Nutrient Pollution in the Rivers of China

Xi Chen,<sup>\*,†,‡,§</sup> Maryna Stokal,<sup>‡</sup> Michelle T.H. Van Vliet,<sup>‡</sup> John Stuiver,<sup>§</sup> Mengru Wang,<sup>‡</sup> Zhaohai Bai,<sup>†</sup> Lin Ma,<sup>\*,†,§</sup> and Carolien Kroeze<sup>‡</sup>

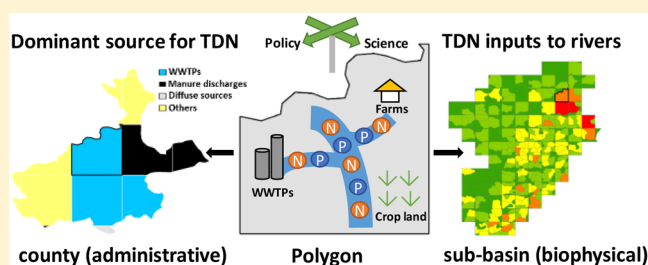
<sup>†</sup>Key Laboratory of Agricultural Water Resources, Hebei Key Laboratory of Soil Ecology, Center for Agricultural Resources Research, Institute of Genetics and Developmental Biology, Chinese Academy of Sciences, 286 Huaizhong Road, Shijiazhuang 050021, China

<sup>‡</sup>Water Systems and Global Change Group, Wageningen University & Research, Droevendaalsesteeg 4, Wageningen 6708 PB, The Netherlands

<sup>§</sup>Laboratory of Geo-information Science and Remote Sensing, Wageningen University and Research, Droevendaalsesteeg 3, Wageningen 6708 PB, The Netherlands

## S Supporting Information

**ABSTRACT:** Chinese surface waters are severely polluted by nutrients. This study addresses three challenges in nutrient modeling for rivers in China: (1) difficulties in transferring modeling results across biophysical and administrative scales, (2) poor representation of the locations of point sources, and (3) limited incorporation of the direct discharge of manure to rivers. The objective of this study is, therefore, to quantify inputs of nitrogen (N) and phosphorus (P) to Chinese rivers from different sources at multiple scales. We developed a novel multi-scale modeling approach including a detailed, state-of-the-art representation of point sources of nutrients in rivers. The model results show that the river pollution and source attributions differ among spatial scales. Point sources accounted for 75% of the total dissolved phosphorus (TDP) inputs to rivers in China in 2012, and diffuse sources accounted for 72% of the total dissolved nitrogen (TDN) inputs. One-third of the sub-basins accounted for more than half of the pollution. Downscaling to the smallest scale (polygons) reveals that 14% and 9% of the area contribute to more than half of the calculated TDN and TDP pollution, respectively. Sources of pollution vary considerably among and within counties. Clearly, multi-scale modeling may help to develop effective policies for water pollution.



## 1. INTRODUCTION

Anthropogenic nutrient inputs of nitrogen (N) and phosphorus (P) to water systems deteriorate water quality globally.<sup>1,2</sup> Elevated levels of human-generated nutrients in watersheds are exported to rivers and then to coastal systems.<sup>3</sup> Global nutrient models such as the Global Nutrient Export from Watersheds 2 (Global NEWS-2)<sup>4</sup> on a basin scale and the Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE-GNM)<sup>5</sup> on a 0.5° grid scale were developed to quantify nutrient pollution in water systems and identify the sources. China, with its rapid economic growth, has been suffering from this problem during the last decades.<sup>6–10</sup> Point sources are important reasons for this pollution.<sup>6,11–14</sup> While most studies only address point sources in terms of the sewage discharges of human waste, the fast increase in the direct discharge of animal manure has become an alarming trend in water pollution in China.<sup>6,10,13</sup> The ongoing transition toward industrialized livestock production has decoupled crop and livestock production, resulting in the direct discharges of manure to recipient water bodies.<sup>15–17</sup> Regional nutrient models such as the Model to Assess River Inputs of Nutrients to Seas (MARINA 1.0)<sup>14</sup> on the sub-basin scale have been

developed for China and address direct manure discharges. Recent studies have indicated that a considerable amount of manure is being discharged to surface waters<sup>12,13</sup> and that the impact on water quality is larger than that of overfertilization in China.<sup>6,14</sup> Despite these model improvements, we still identify several knowledge gaps for nutrient N and P modeling for China in general and in relation to point sources.

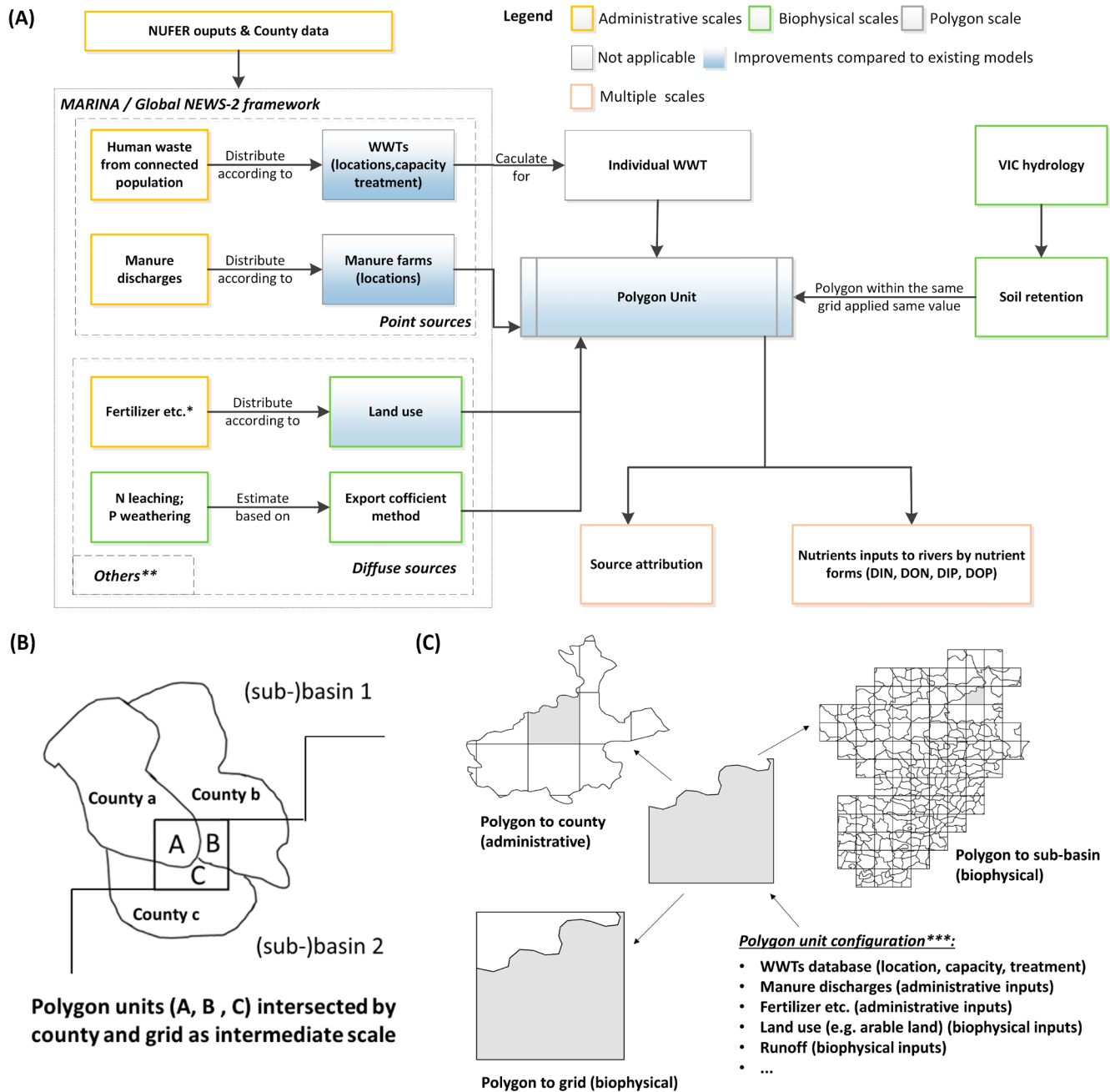
First, the understanding of environmental impacts and the development of associated management strategies of nutrient pollution is still humbled by traditional modeling scales. Existing nutrient models used to quantify the nutrient pollution in rivers and coastal seas are on biophysical scales or nonadministrative scales (e.g., basin scales or grid scales).<sup>4,5,14</sup> Inputs from national/provincial scales are aggregated or disaggregated to biophysical scales such as grids or (sub)basins. However, these original administrative inputs, which contain important drivers (e.g., population and

Received: December 30, 2018

Revised: July 17, 2019

Accepted: July 19, 2019

Published: July 19, 2019



**Figure 1.** (A) Overview of the multi-scale modeling framework; DIN, DON, DIP, and DOP in the box refer to dissolved inorganic (DIN, DIP) and dissolved organic (DON, DOP) nitrogen and phosphorus. (B) Illustrations of polygon-based units as the intermediate modeling scale. (C) Illustrations of the multi-scale framework: from polygon units to different biophysical and administrative scales. \*This box also includes manure application on land, biological N<sub>2</sub> fixation, atmospheric N deposition, crop export, and human waste applied on land from people that are not connected to sewage systems. \*\*Others include human waste from unconnected populations that are directly discharged to water bodies. \*\*\*For a complete overview of inputs and sources see section S1 and Tables S1,S2.

agricultural activities) for nutrient pollution are erased in the models. This hinders the analysis of ecological impacts and the associated source attribution at the administrative (e.g., county) levels and results in a mismatch in the spatial levels needed to develop suitable mitigation policies and management strategies. This poses questions to the traditional way of modeling on biophysical scales, such as what is the appropriate modeling scale for water pollution management? Should we enable our models to run on both biophysical and administrative scales and how? Some intermediate-scale models such as the Hydrological Response Unit in Soil and

Water Assessment Tool (SWAT) model<sup>18</sup> or the Soil type Land use Combination in Hydrological Predictions for the Environment (HYPE) model<sup>19</sup> have been introduced to account for subvariability within the modeling unit. However, the main focus of these models is still on the representation of biophysical characteristics and associated levels. The concept and need for multi-scale modeling are also emerging in other modeling fields that have a strong link between social and ecological systems and nutrient pollution, such as the field of ecological-economic modeling.<sup>20</sup> The recently developed Multi-Scale Integrated Model of Ecosystem Services model

proposes a “locations” concept to enable the model to be configured on multiple scales.<sup>21,22</sup> However, this model focuses on ecosystems services; the concept to bridge modeling results on biophysical scales and policy making on administrative scales is still absent. To our knowledge, a formalized modeling framework for multi-scale nutrient modeling representing both biophysical and administrative levels does not exist.

Second, an understanding of the impacts of point sources is hindered by the generally poor representation of spatial locations in the existing nutrient models for water quality.<sup>4,5,14</sup> Point sources, by definition, are emitted in a discernible and concentrated way.<sup>23</sup> The spatial characteristics of the place where point sources are located, such as the geomorphology of a river network, hydrological conditions, and the spatial relations with dams, strongly influence water quality impacts.<sup>4,5,24–26</sup> Global nutrient models, such as the IMAGE-GNM model, represent nutrient processes on a grid scale. In contrast, the global NEWS-2 model quantifies nutrient export by rivers on a basin scale. Regional models such as MARINA 1.0 for China take the sub-basin as the calculation unit. However, these global and regional models do not take into account the spatial locations of point sources. These models quantify the point sources on every modeled unit, which in principle is similar to diffuse sources assuming they are emitted from each unit. The Spatially Referenced Regression on Watershed Attributes (SPARROW) model provides catchment scale estimates of the nutrient loads of human waste emitted from centralized Waste Water Treatment Plants (WWTPs) for the southeastern United States based on locations and monitored effluent data.<sup>26</sup> However, this approach is highly dependent on the quality and adequacy of the monitoring data. An alternative modeling approach is needed, in particular for regions where monitoring data are limited. Furthermore, up-to-date quantitative data of point source inputs of nutrients to Chinese water systems are needed.<sup>6,12,27,28</sup> The MARINA 1.0 model is one of the few models that account for direct manure discharges, and its quantifications are valid until the year 2000.<sup>6,14</sup> Understanding the recent impacts of point source nutrient pollution is necessary for developing effective mitigation management strategies.<sup>29,30</sup>

Therefore, the objective of this study is to quantify inputs of nitrogen (N) and phosphorus (P) to Chinese rivers from different sources at multiple scales. To this end, we (1) develop a multi-scale modeling framework to bridge the biophysical and administrative scales and (2) improve the modeling of point sources of nutrients (human waste emitted from WWTPs and direct manure discharge) by incorporating the locations of the point sources and updating the associated inputs.

## 2. METHODOLOGY

We take three existing models as a starting point: the MARINA 1.0<sup>14</sup> model, the NUTrient flows in Food Chains, Environment and Resources use (NUFER) model<sup>13,31</sup> and the Variable Infiltration Capacity (VIC) hydrological model.<sup>32–34</sup> In the following section, we first introduce our newly developed multi-scale modeling framework. We then describe how we use these three models and the particular improvements made for point sources.

**2.1. Multi-scale Framework and Polygon-Based Model.** Our multi-scale approach takes polygon units as a basis. Polygons are the intermediate scale between the

biophysical ( $0.5^\circ \times 0.5^\circ$  grid) and administrative scales (county) (Figure 1B). Chinese counties are at a lower administrative level than cities, and the  $0.5^\circ$  grid is a common scale for global hydrological models. Therefore, we delineate our polygons from the intersections of counties and grids, keeping the characteristics from both the biophysical and administrative units (e.g., land cover, agricultural activities, and population). Background on the study area (e.g., maps and current nutrient management strategies) is described in section S1.1.

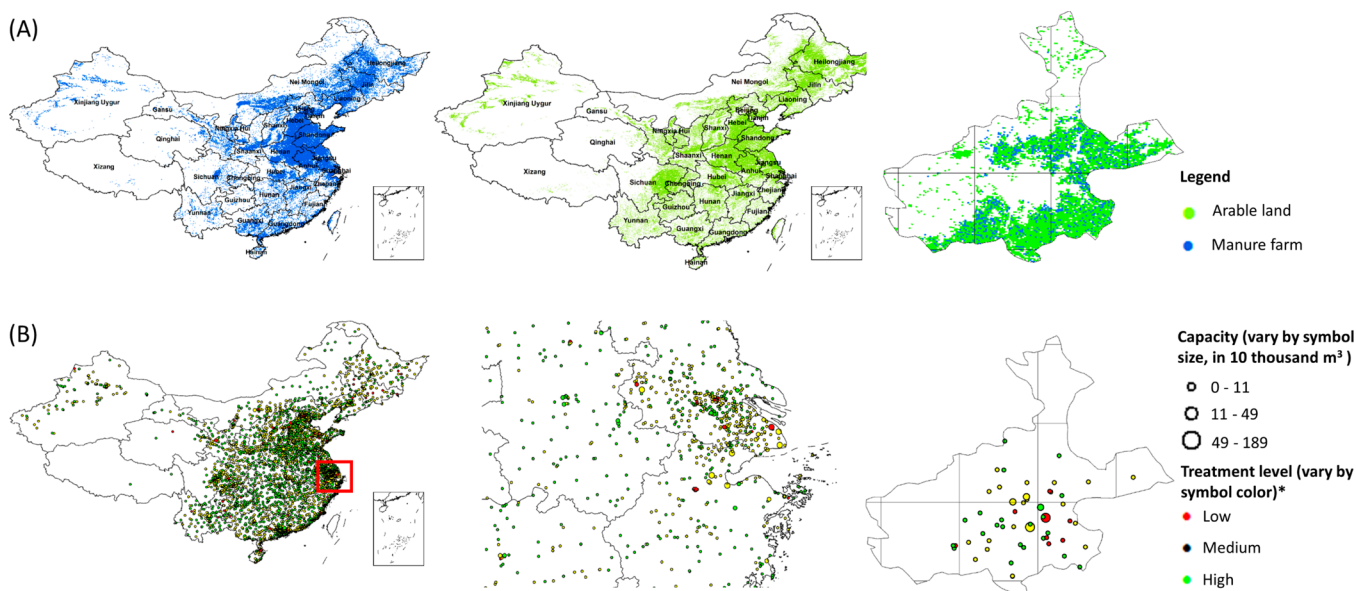
The multi-scale modeling framework can be used to both (1) model different processes (Figure 1A) and (2) produce the outputs (e.g., the inputs of N and P to rivers) on multiple scales (Figure 1C). For the first aspect, nutrient flows are quantified on different scales as appropriate (Figure 1A). For instance, the quantity of human waste emitted from centralized WWTPs is calculated on the individual WWTP level and stored in the associated polygons according to the exact locations of the WWTPs. Manure discharges and diffuse sources are distributed from the counties to the polygons as a function of manure farm locations and land use and are thus quantified on the polygon scale. Nutrient retention in soils for diffuse sources is calculated as a function of total runoff, which is on the grid scale.

Model outputs can be produced on multiple scales, including the polygon, grid, county, sub-basin, and basin levels. Polygons serve as a storage carrier for information and finally as a bridge for upscaling and downscaling (Figure 1C). Polygons are characterized by a univocal spatial relation that links them to the corresponding grid, county, and sub-basin. The quantifications on different scales are stored in the polygon units according to the spatial locations. For example, we store the individual WWTP data to polygons according to the exact locations of the WWTPs. This approach ensures that calculations can be traced back to the smallest units and aggregated (e.g., for total nutrient inputs) or recalculated (e.g., for the source attribution) for the different scales. More details are listed in section S1.2.

**2.2. Model Description.** The multi-scale model is based on three models: the MARINA 1.0<sup>14</sup> model, the NUFER<sup>13,31</sup> model, and the VIC hydrological model.<sup>32–34</sup> The MARINA 1.0 model quantifies the annual river export of nutrients by source from Chinese sub-basins with an improved modeling approach for animal and human wastes. The NUFER model quantifies nutrient flows in the food chain of China at the county, provincial, and national scales. The model was developed based on statistical data, field surveys, and literature. The VIC model is a macro-scale hydrological model that has been widely applied for river basins worldwide.

These models provide a strong modeling basis (MARINA 1.0) and inputs (NUFER, VIC) for this study (Figure 1A). Inputs of N and P to Chinese rivers by polygon are quantified following the modeling approaches of the MARINA 1.0 model, while the modeling of point sources is improved. The Chinese county database for 2012 (2338 counties) and the NUFER model county outputs<sup>13,35</sup> are used as inputs (e.g., fertilizer applied) to our model. The VIC model provides total runoff on a  $0.5^\circ \times 0.5^\circ$  grid.<sup>33,34</sup>

The newly developed model quantifies the dissolved N and P inputs to rivers by source for the year 2012 on both biophysical scales (e.g., sub-basin and grid) and administrative scales (e.g., county). The main equation to quantify the inputs



**Figure 2.** (A) Locations of arable land and manure farms from land-use data ( $1 \times 1$  km) and an example of the polygon units with locations in the urban areas of Beijing. (B) The locations of WWTPs categorized by the daily average capacity (in 10 thousand  $m^3$ ) and the treatment efficiencies (%) for nitrogen (see Figure S4 for phosphorus). The detailed examples illustrate part of the coastal areas (indicated by the red box) and the urban areas of Beijing. Treatment levels include low (N removal rate  $\leq 35\%$ ), medium ( $35\%–55\%$ ) and high ( $\geq 55\%$ ) removal, following Van Drecht et al.<sup>45</sup>

of N and P to rivers by nutrient forms follows the MARINA 1.0 model:<sup>14</sup>

$$\begin{aligned} RS_{total,F} &= RS_{dif,F} + RS_{pnt,F} + RS_{others,F} \\ &= (FE_{ws,F} \times WS_{dif,F} + RS_{dif,EC,F}) + RS_{pnt,F} \\ &\quad + RS_{others,F} \end{aligned} \quad (1)$$

where  $RS_{total,F}$  is the total inputs of nutrient form (F) from land to rivers by a polygon ( $kg\ year^{-1}$ ). Nutrient forms include dissolved inorganic (DIN, DIP) and dissolved organic (DON, DOP) N and P. River sources (RS) of inputs include point sources ( $RS_{pnt,F}$ ), diffuse sources ( $RS_{dif,F}$ ), and other sources ( $RS_{others,F}$ ) ( $kg\ year^{-1}$ ). Diffuse sources ( $RS_{dif,F}$ ) include explicit land sources ( $WS_{dif,F}$ ), which are corrected for soil retention ( $FE_{ws,F}$ ), and parametrized export processes ( $RS_{dif,EC,F}$ ). ( $RS_{others,F}$ ) is the direct discharges of nutrient form (F) to rivers from the human waste of the population that is not connected to WWTPs by polygon ( $kg\ year^{-1}$ ).

Many aspects are improved, especially the quantification of point sources. Below, we describe the model by each of the source categories, with an emphasis on the improvements compared to the existing models. Extended model descriptions are in section S1.

Point sources of the total input of nutrient form (F) to rivers ( $RS_{pnt,F}$ ,  $kg\ year^{-1}$ ) include manure direct discharges ( $RS_{pnt-ma,F}$ ) and human waste emitted from WWTPs ( $RS_{pnt-con,F}$  (eq 2):

$$RS_{pnt,F} = RS_{pnt-ma,F} + RS_{pnt-con,F} \quad (2)$$

For the manure direct discharges ( $RS_{pnt-ma,F}$ ,  $kg\ year^{-1}$ ), we improve the MARINA 1.0 approach in two main ways. (1) We update the inputs of direct manure discharges from the provincial to the county level to year 2012. For this, we use the county information from the NUFER model on the total manure excretion, which is quantified based on county

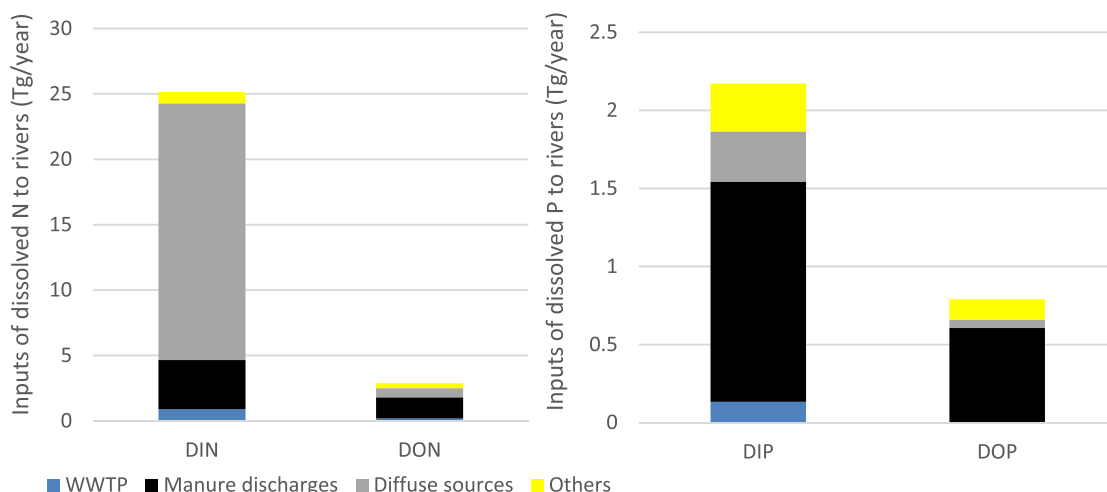
databases of Chinese statistics, and fractions of direct manure discharge, which are derived from on-site surveys.<sup>13,35</sup> (2) We include the locations of manure farms that were used for the detailed assignment of manure discharges from counties to polygons. The rural residential area from the land-use data ( $1 \times 1$  km grid)<sup>36</sup> is used as a proxy for the locations of farms (Figure 2A). This was done with the following equations:

$$RS_{pnt-ma,F}(\text{county}) = T(E)ma_{dis} \times EF_{pnt-ma,F} \quad (3)$$

$$RS_{pnt-ma,F}(\text{polygon}) = RS_{pnt-ma,F}(\text{county}) \times \text{Frac}_{farms\_county} \quad (4)$$

where  $T(M)ma_{dis}$  is the nutrient element E (N or P) of animal manure that is directly discharged to rivers ( $kg\ year^{-1}$ ); and  $EF_{pnt-ma,F}$  is the manure fraction of element E (N or P) entering rivers as form F (0–1). This fraction is taken from the MARINA 1.0 model and is derived based on the literature.<sup>14,37–40</sup>  $\text{Frac}_{farm\_country}$  is the fraction of animal farms in the county that are located in the polygon (section S1.3).

The modeling of human waste from centralized WWTPs ( $RS_{pnt-con,F}$ ,  $kg\ year^{-1}$ ) is based on our unique WWTP database, which includes more than 4204 WWTPs across China (section S3). The database includes the longitude and latitude, average daily treatment capacity, treatment technologies, and associated treatment efficiencies for individual WWTPs (Figure 2B). The lists of WWTPs are obtained from the national list of operating WWTPs for the year 2014<sup>41</sup> and the National Intensive Monitoring and Control Enterprise List for WWTPs for 2016.<sup>42</sup> We manually search for the address, locate the latitude and longitude of the individual WWTPs, and derive the treatment efficiencies for 46 technologies applied in those WWTPs based on a literature review and expert knowledge (sections S1.3 and S2). We also updated the associated model parameters such as the urban and rural populations from the county database and treatment rates at the city and county



**Figure 3.** Nitrogen (N) and phosphorus (P) inputs to rivers in China (Tg/year) by form: dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), dissolved inorganic phosphorus (DIP), dissolved organic phosphorus (DOP), and by source. 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<sub>2</sub> fixation by agricultural crops and by natural vegetation, atmospheric N deposition, leaching of organic matter, and weathering of P-containing minerals from agricultural and non-agricultural soils, and human waste from populations that are unconnected to sewage systems that stay on the land. Others refer to the human waste from urban and rural populations that are unconnected to sewage systems but discharged directly to rivers.

levels from the China Urban-Rural Construction Statistical Yearbook for 2012 and 2014.<sup>43,44</sup>

The nutrient inputs from individual WWTPs can be quantified following the method of Van Drecht et al.<sup>45</sup> as follows (details can be found in section S1.3):

$$RS_{\text{pnt\_con},F} = \{(1 - hw_{\text{rem},E}) \times \text{PopCon}_{\text{WWTP}} \times E(E)_{\text{pnt}} \times \text{Frac}_{\text{treat}}\} \times FE_{\text{pnt},F} + (1 - \text{Frac}_{\text{treat}}) \times \text{PopCon}_{\text{WWTP}} \times E(E)_{\text{pnt}} \times FE_{\text{pnt},F} \quad (5)$$

where  $hw_{\text{rem},E}$  is the removal of nutrient element E (N or P) during treatment in sewage systems (0–1);  $E(E)_{\text{pnt}}$  is the inputs of nutrient N or P to watersheds (land) resulting from human excrement (for N and P) and P detergents (kg person<sup>-1</sup> year<sup>-1</sup>);  $FE_{\text{pnt},F}$  is the fraction of sewage effluents exported to rivers as nutrient form (F) (0–1);  $FE_{\text{pnt},F}$  is directly proportional to the N removal rate for DIN ( $FE_{\text{pnt},\text{DIN}}$ ), while for other nutrient forms it is the calibrated constant, as with the Global NEWS-2 model;  $\text{PopCon}_{\text{WWTP}}$  is the population number connected to the individual WWTP; and  $\text{Frac}_{\text{treat}}$  is the percentage of the total wastewater transported to the WWTPs that is treated (0–1). The outputs from individual WWTPs (section S3) are located to polygons according to the locations of the WWTPs (Figure 2B) to calculate the dissolved N and P inputs to rivers by polygon according to eq 1.

Diffuse sources ( $RS_{\text{dif},F}$ , kg year<sup>-1</sup>) include explicit land sources ( $WS_{\text{dif},F}$ , kg year<sup>-1</sup>), which are corrected for soil retention ( $FE_{\text{ws},F}$ , 0–1) before entering rivers, and parametrized export processes ( $RS_{\text{dif},EC,F}$ , kg year<sup>-1</sup>):

$$RS_{\text{dif},F} = (FE_{\text{ws},F} \times WS_{\text{dif},F} + RS_{\text{dif},EC,F}) \quad (6)$$

where  $WS_{\text{dif},F}$  is the total nutrient inputs to land that are corrected for crop export by harvesting and grazing (kg year<sup>-1</sup>);  $FE_{\text{ws},F}$  is the fraction of nutrient form (F) that is exported from land to rivers from diffuse sources (0–1); and

$RS_{\text{dif},EC,F}$  includes the weathering of P-containing minerals (for DIP) and leaching of organic matter (for DON and DOP), and its values are calculated by the export-coefficient approach of the MARINA 1.0 model for polygons.

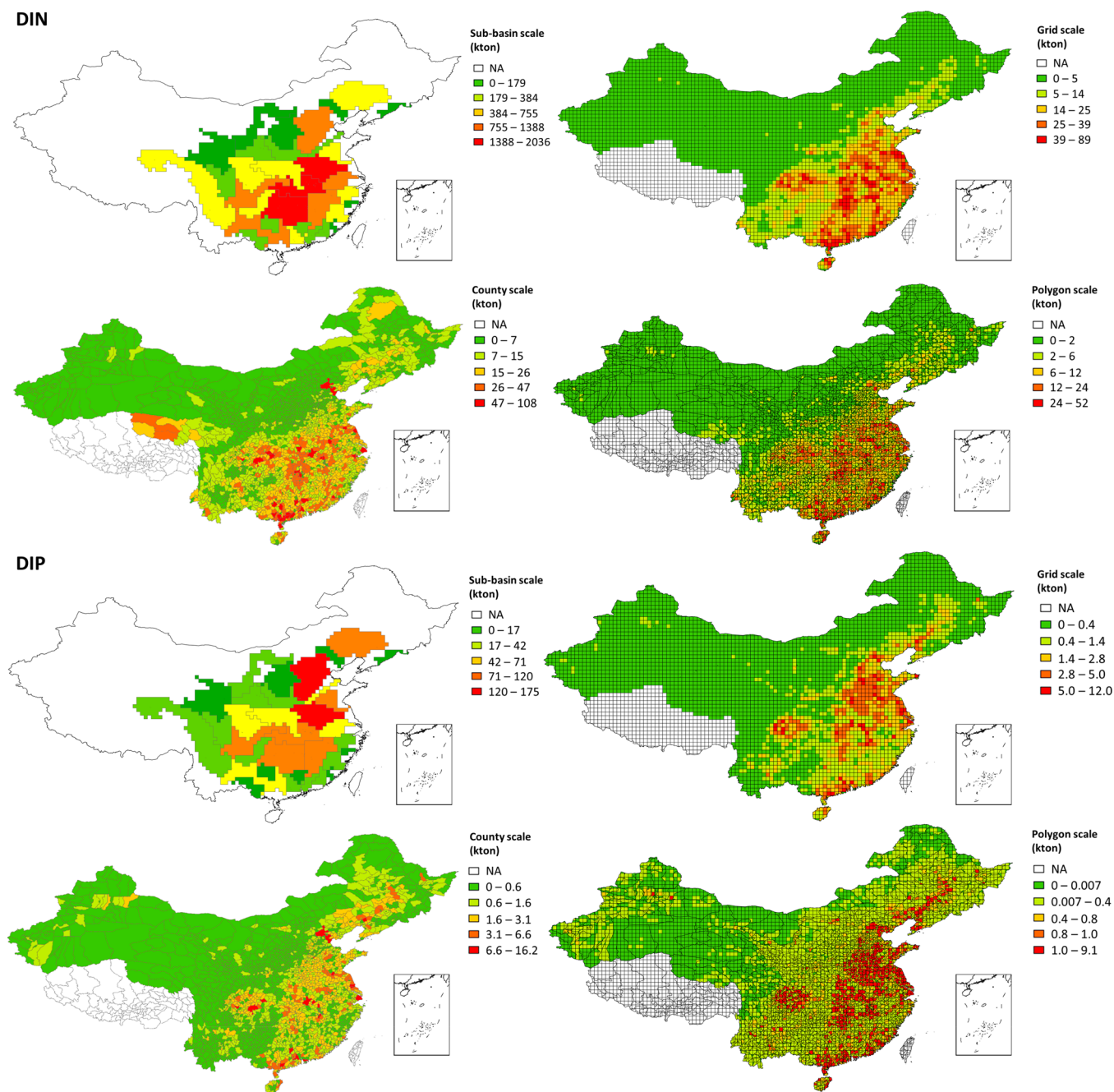
The improvements are mainly made to the explicit land sources (e.g., fertilizers applied, Figure 1A). We derive the model inputs from the Chinese county database for year 2012, and we use the NUFER model county outputs and other local sources (Table S2). Compared to the Global NEWS-2 and MARINA 1.0 models, our inputs are provided on a more detailed and spatially explicit level (county vs national).<sup>4,46</sup> Most of the parameters are distributed to polygons by incorporating the land-use map information (Figure 2A), and for a few model inputs, we applied an area-weighted method (section S1.4).  $FE_{\text{ws},F}$  is calculated as a function of the total runoff for each polygon. The calibrated coefficients in the function are from the MARINA 1.0 model. The total runoff is the average annual total natural runoff from 1970 to 2000 on a 0.5° × 0.5° grid from the VIC model.<sup>33,34</sup> The runoff of the grid in which the polygon is located is applied to the quantifications.

Other sources ( $RS_{\text{other},F}$ ) that are accounted for in our model are human waste from unconnected populations that is directly discharged to rivers according to the MARINA 1.0 model<sup>14</sup> (section S1.5).

### 3. RESULTS

We present the results on biophysical (sub-basin and grid), administrative (county), and polygon scales. First, we analyze the N and P inputs to rivers and their source attributions on selected scales from coarse to detailed, i.e., from the national to the sub-basin scales and then to the most detailed polygon scale. We do not discuss the grid results in detail. We take the Hai basin as an example to demonstrate how a multi-scale modeling approach can bridge the biophysical (sub-basin) and administrative scales (county).

**3.1. National, Sub-Basin, and Polygon Analysis.** *National Analysis.* Point sources contribute considerably to

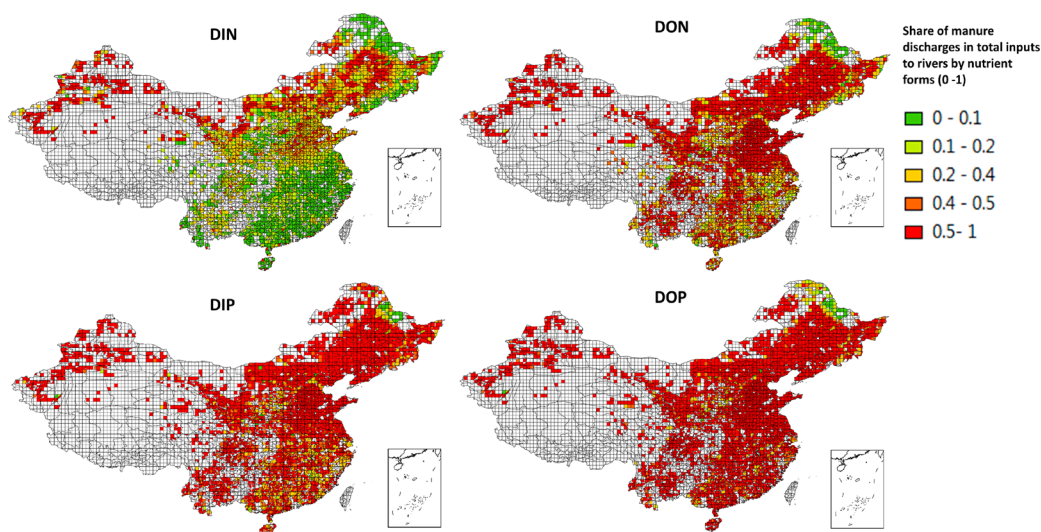


**Figure 4.** Nitrogen and phosphorus inputs to rivers ( $\text{kton year}^{-1}$ ) in the year 2012 on multiple scales (sub-basin, grid, county, and polygon). The results are shown for dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP). For dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP), see Figure S15. NA indicates regions for which detailed input data are not available.

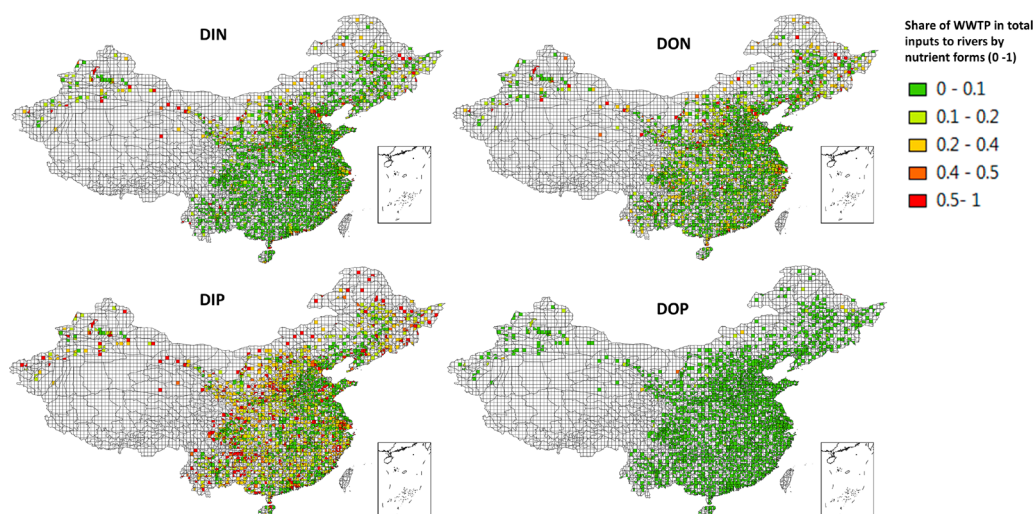
the total dissolved phosphorus (TDP) of river pollution, while diffuse sources dominate the total dissolved nitrogen (TDN) (Figure 3, Figure S7). We quantify approximately 28 Tg of the TDN inputs to Chinese rivers, of which approximately 90% is in the form of DIN. Diffuse sources account for 78% of the total DIN inputs, while the point sources account for 18%. However, for DON, the share of point sources is up to 63%. The contribution of diffuse sources to DON is much lower than to DIN because soil retention tends to be substantially higher for DON. As a result, the percentage of N in soils that is not retained and thus enters rivers is higher for DIN than for DON ( $FE_{ws,DIN}$  and  $FE_{ws,DON}$  are 31% and 0.4%, respectively, at the national scale). The TDP inputs to rivers are 3 Tg

according to our estimation. The share of DIP and DOP point sources are 73% and 79%, respectively. For all nutrient forms, manure discharge accounts for up to 80% of the total point source pollution. The much larger share of manure discharges in TDP inputs compared to TDN inputs is due to the lower percentages of DIP and DOP entering rivers from diffuse sources ( $FE_{ws,DIP}$  and  $FE_{ws,DOP}$  are 6% and 0.4%, respectively, at the national scale).

**Sub-Basin Analysis.** The N and P inputs to rivers and the source attributions differ among nutrient forms and sub-basins (Figure 4 and Figure S11). N and P inputs to rivers are concentrated in the Hai basin and Dongting and in the middle downstream of the Changjiang. In these four sub-basins, the



**Figure 5.** Share of manure discharges in the total inputs of dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), dissolved inorganic phosphorus (DIP), and dissolved organic phosphorus (DOP) to rivers in China at the polygon scale (fraction, 0–1).



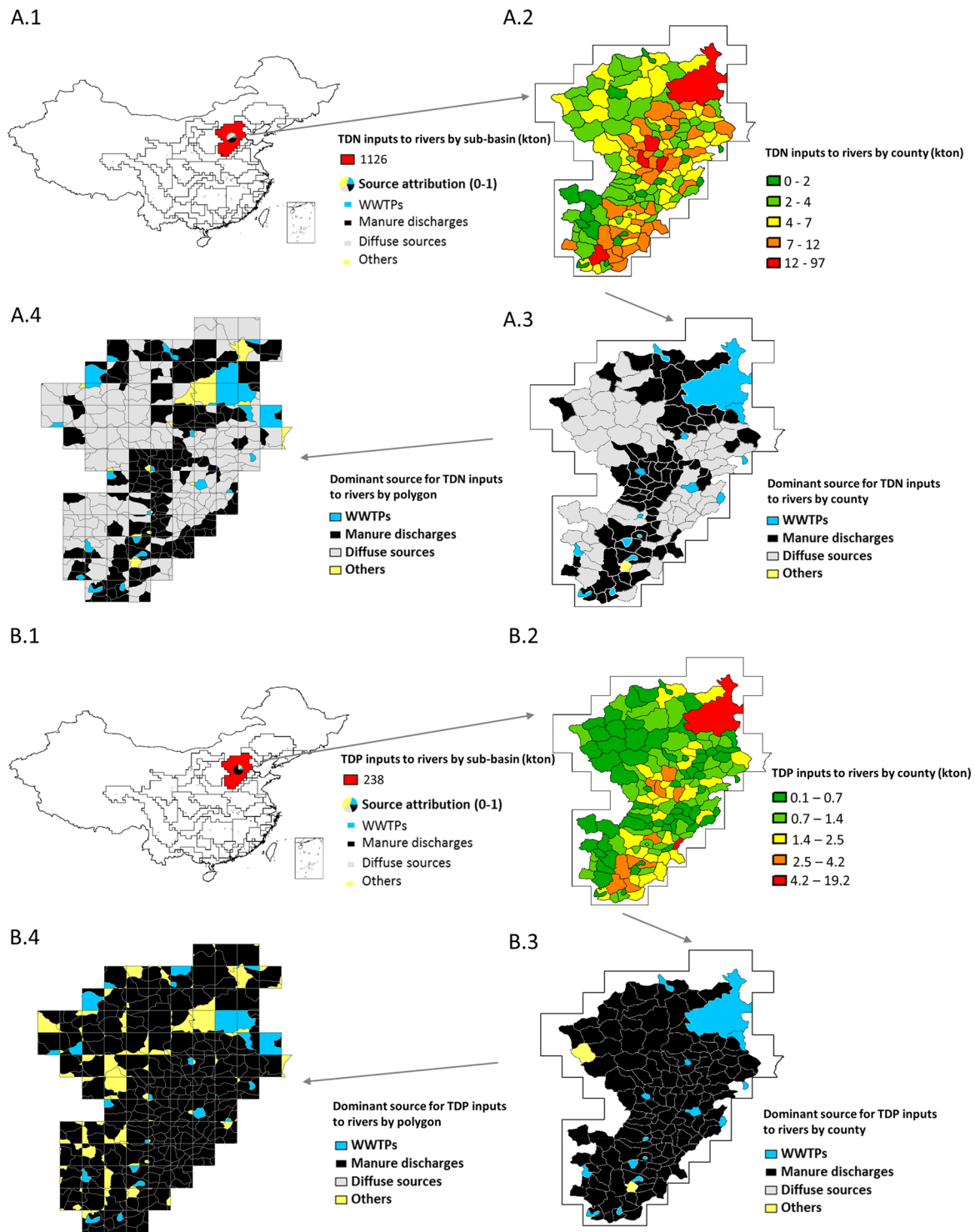
**Figure 6.** Share of human waste from wastewater treatment plants (WWTPs) in total inputs of dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON), dissolved inorganic phosphorus (DIP), and dissolved organic phosphorus (DOP) to rivers in China at the polygon scale (fraction, 0–1).

inputs are on average 1.5 (TDN) and 2 (TDP) times higher than in other sub-basins. In most sub-basins, the main sources of DIN in rivers are diffuse sources such as fertilizer and manure application on land. In Tongdaogual and Liao, however, the direct discharge of manure is a dominant source. This can be partly explained by the fact that precipitation and runoff are relatively low in these sub-basins, resulting in lower diffuse inputs. For DON, DIP, and DOP, point sources and, in particular, point sources of manure are the dominant source in almost all sub-basins. Human waste emitted from WWTPs is an equally important source as manure discharges in some sub-basins where urbanization rates are high, such as Dongjiang and Qujiang.

**Polygon Analysis.** N and P inputs to rivers are relatively high in southern and eastern China where agriculture and industrialized farming are important and urbanization rates are high (Figure 4, Figure S8). The coastal area of Liaoning, the middle part of Jilin, the entire Shandong, the middle of Hebei, Beijing, Tianjin, Chongqing, the northeast of Henan, and

coastal areas of Guangxi and Guangdong contribute more to N and P inputs to rivers than other areas (Figure 4, Figure S10). These areas are hotspots of pollution because of the combined effects of intensive human activities, high runoff, and multiple point source locations. The dominant sources differ among nutrient forms and polygons (Figures 5 and 6). In most parts of China, point sources are more important sources of nutrients in rivers compared to diffuse sources. This is especially true for point source inputs of animal manure (Figure 5), except for DIN. Spatial variations in nutrient inputs to rivers for DON, DIP, and DOP are generally in line with spatial distributions of livestock farms (Figure S8). For DIN, agricultural diffuse sources such as fertilizer application are the most important source (Figure S12). WWTPs are a dominant source in urban areas, including major cities such as Shanghai, Guangzhou, and Beijing (more analyses in Figures S13 and S14).

**Analysis Across Scales.** National analyses indicate that point sources are the dominant sources of TDP in rivers, while



**Figure 7.** Hai basin as an example of multi-scale analysis: (A.1, B.1) Total dissolved nitrogen and phosphorus (TDN, TDP) inputs to rivers and their source attributions at the sub-basin scale (in kton); (A.2, B.2) TDN and TDP inputs to rivers by county (in kton); (A.3, B.3) Dominant sources of TDN and TDP inputs to rivers by county; and (A.4, B.4) Dominant sources of TDN and TDP inputs to rivers by polygon. WWTPs refer to human waste emitted from wastewater treatment plants. Manure discharges refer to the direct discharge of manure to water bodies. Diffuse sources include synthetic fertilizer use, manure applied on land, biological N<sub>2</sub> fixation by agricultural crops and by natural vegetation, atmospheric N deposition, leaching of organic matter, weathering of P-containing minerals from agricultural and non-agricultural soils, and human waste applied on land from populations that are not connected to sewage systems. Others refer to the human waste discharges from urban and rural populations that are not connected to sewage systems.

diffuse sources dominate TDN river pollution. The results on the sub-basin scale reveal more spatial variability of nutrient pollution and their sources. We calculate that 35% and 31% of the sub-basin area contributes to more than 50% of the total

sub-basin TDN and TDP pollution. Zooming further into the polygon scale, we find that an even smaller area accounts for most of the total nutrient pollution. We narrow down the area by factors of 2.5 (14% vs 35%) and 3 (9% vs 31%) for TDN



and TDP, respectively. We also find that point sources alone can contribute to most of the DON, DIP, and DOP pollution. For DON, we determine that 23% of the total area contributes to more than 50% of the total pollution by point sources; the percentage is 15% and 11% for DIP and DOP, respectively (Figure S9). These differences between the sub-basin and polygon scales are mainly because agricultural activities (fertilizer applied), animal production (manure farms), and highly urbanized cities (WWTPs) are concentrated in small areas. Sub-basins are typically coarser than these areas. Thus, sub-basins are not detailed enough to zoom into the locations of pollutants. Zooming into more detailed spatial levels (polygon) allows us to better locate polluting areas and identify associated sources. This illustrates one of the strengths of our multi-scale approach.

**3.2. Illustrating the Potential of Multi-scale Modeling.** Our multi-scale modeling approach can quantify nutrient inputs to rivers and their source attributions on both biophysical (e.g., sub-basin) and administrative scales (e.g., county). We select the Hai basin as an example to demonstrate how our approach can potentially contribute to local water quality analyses and more tailor-made water management strategies. The Hai basin is a highly polluted basin with relatively high N and P inputs to rivers (Figure S11). Most parts of Beijing, Tianjin, and Hebei provinces, where agricultural activities are intensive and urbanization rates are high, are located in the basin (Figure S8). Thus, we zoom in and trace the sources and their associated impacts for the administrative units (Figure 7). The results indicate that there is high spatial variability in both nutrient inputs and source attributions within the basin.

**Nutrient Inputs.** We identify the hotspot counties, which only cover 29% and 24% of the total basin area but contribute to more than 50% of the TDN and TDP pollution. These hotspot counties are 54 and 41 out of 149 total counties for TDN and TDP, respectively. Most of these (39 out of 56) are hotspots for both N and P. The urban areas of Beijing (in the north of the basin), Guantao (in the southern margin of the basin), and Dingzhou (in the middle of the basin) are top 3 contributors to the total TDN and TDP pollution. At the polygon scale, we can narrow down the areas within these counties by more than 50% (for TDN and TDP, 41% and 36% of the area, respectively, contributes to more than 50% of the total hotspot pollution). The different impacts of administrative units on nutrient pollution result from the combined effects of biophysical factors (e.g., runoff and land-use), administrative drivers (population and agricultural activities), and point source locations.

**Source Attribution.** We also show that there is high spatial variability in dominant sources across scales, among and even within the counties. For the Hai basin as a whole, the dominant sources for TDN and TDP are both manure discharges. However, zooming into the county scale, the dominant sources differ. For TDN, we calculate that in 48% of the counties, manure discharges are the dominant source, while in 42% of the counties, diffuse sources dominate, with 9% from WWTPs and 1% from “others”, which includes human waste discharges from unconnected populations. For TDP, the variation is smaller. Manure discharges dominate in most counties. However, in 10% of the counties, WWTPs dominate. Zooming into the polygon scale, we find that the dominant sources even vary within the counties. For example, in the urban area of Beijing, which contributes more nutrient

inputs to rivers than the other counties in Hai, the dominant sources at the county scale for both TDN and TDP are WWTPs. However, in the 14 polygons within the county, the dominant sources differ (Figure 7). For TDN, WWTPs dominate in up to 44% of the total area, followed by “others” in up to 36% of the area, manure discharges in up to 19% of the area, and diffuse sources in only 1% of the area. For TDP, the dominant sources include all source categories except diffuse sources, and “others” are the dominant source for most polygon areas (37%), followed by manure discharges (34%) and WWTPs (29%). The results confirm that it is relevant and useful to quantify the impacts on water quality and sources on administrative units.

## 4. DISCUSSION

**4.1. Model Evaluation.** We evaluate our model by discussing<sup>47</sup> (1) model uncertainties, (2) model inputs, (3) model approach, (4) sensitivity analysis, and (5) model outputs and trends compared to other models and studies (this discussion is extended in section S4).

First, all models have their uncertainties. For instance, our model includes some calibrated coefficients from the Global NEWS-2 model.<sup>4</sup> Applying these to scales other than the basin scale introduces uncertainty. This holds for the equation for soil retention ( $FE_{ws,F}$ ) and particularly for DIN, which is dominated by diffuse sources. Nevertheless, our retention for DIN on the basin scale such as Changjiang (49%) captures the increasing trend of  $FE_{ws,F}$  for DIN, which increased from 0.11 to 0.61 from 1970 to 2003.<sup>48</sup> This agrees with studies that suggest that the capacity in the watershed to retain N can be diminished due to increasing N inputs from human activities.<sup>49–51</sup> For DIP, Harrison et al.<sup>52</sup> also applied  $FE_{ws,F}$  at the grid (0.5°) level with satisfactory model performance. Another source of uncertainty is the 30 years average (1970–2000) that we use for total runoff, ignoring annual variability. In addition, our manure discharges are based on field surveys and expert knowledge.<sup>13,53</sup> We use rural residential areas as a proxy for the locations of farms and downscale from counties to polygons using an area-weighted method. This introduces uncertainties because these are not real locations, and the area-weighted distribution does not represent real animal numbers. We recognize and quantify this uncertainty by sensitivity analyses (Table S6). Our approach is similar to that of Zhao et al.,<sup>54</sup> who used rural residential locations as a proxy for farms and evenly allocated  $NH_3$  emissions from manure to these locations.

Second, we consider our model inputs to be appropriate for a number of reasons. They are from widely accepted sources and widely used models such as the NUFER and VIC models (section S4.1). County data are from statistics yearbooks that are considered to be reliable sources.<sup>13,35</sup> Data on wastewater treatment plants are from government documents, exact locations, the literature and expert knowledge (section S3). Third, our model approach for diffuse sources compares well to other models (section S4.2; Table S5) and studies (section S4.4), and we improve the method for point sources. There are fewer studies on the direct discharges of manure to rivers than on WWTPs. Further, we have more up-to-date data for WWTPs, and we quantify data at the individual WWTP level. For diffuse sources, we adopt the commonly used method in validated models. The nutrient budget on land from human activities is first quantified and corrected for the nutrient retention by soil. The soil retention is quantified by different

approaches using calibrated (our model) or uncalibrated (IMAGE-GNM) parameters. Our modeled soil retention parameters are in acceptable ranges (section S4.4). Fourth, our sensitivity analysis includes important model input parameters (section S4.3) and indicates that the model is fairly robust, with an elementary effect<sup>46,47</sup> that is smaller than 1, and in most cases, substantially smaller (Table S6).

Finally, we compare our results with those of other models (Figure S6) and studies (Table S8). The models cover the main basins (Figure S1), scales (sub-basin, grid), and model approaches (Table S5). Our estimates for 2012 are generally higher than the results for 2000 from the MARINA 1.0<sup>6,14</sup> and IMAGE-GNM<sup>5,55</sup> model. This is in line with actual increases over time (Table S7). Our calculated manure N discharge for 2012 (3.9 Tg) is 14% lower than that of the MARINA 1.0 model (4.5 Tg), and our 2012 P estimate is 57% higher. This difference results from the combined effect of (1) a reduction in the direct discharge of manure between 2000 and 2012 and (2) differences in N and P ratios that are used to estimate manure excretion: we used the NUFER model for this,<sup>13</sup> while the MARINA 1.0 model used IMAGE data.<sup>46</sup> Our source attributions are generally in line with those of the MARINA 1.0 model, but they differ from those of the IMAGE-GNM model. An important reason for this is that the IMAGE-GNM model does not account for the direct discharge of manure to rivers in China. Liu et al.<sup>56</sup> quantified 729 kton of TP inputs to rivers with the IMAGE-GNM model for Changjiang for the year 2010, while we quantified that manure discharges are up to 718 kton despite other sources (Table S8). For N, both models agree that diffuse sources dominate N inputs to rivers. Because the share of particular N is small<sup>57,58</sup> and the modeled soil retentions are comparable, the differences mainly result from N nutrient budgets (our 19.4 vs 14.2 Tg). This is mainly due to different input sources (we use the NUFER model and county statistics). We also compare with other published studies for major basins. The similarities and differences are summarized in Table S8 and discussed in section S4.4. All the above aspects build trust in our model.

**4.2. Findings and Outlook.** We developed a model to quantify N and P inputs to Chinese rivers by source on both biophysical (grid and sub-basin) and administrative (county) scales for the year 2012. The results show that the calculated river pollution and source attributions differ among spatial scales. On the national scale, point sources have the largest share (75%) in TDP inputs to rivers, while diffuse sources dominate (72%) TDN inputs. The results on the sub-basin scale show differences in nutrient pollution and the dominant sources among sub-basins. Our polygon-scale analyses illustrate best how large the spatial variability is: a relatively small area is responsible for most of the river pollution. We find that 14% (vs 35% for the sub-basin) and 9% (vs 32% for the sub-basin) of the total area contribute to more than 50% of the TDN and TDP pollution. We also find that point sources alone concentrated in a relatively small area contribute to more than half of the DON (23% area), DIP (15% area), and DOP (11% area) pollution. Our results confirm that it is relevant and useful for water quality models to quantify pollution levels on administrative scales for more tailor-made and effective problem-solving.

We consider this study to be a new effort for the spatially explicit modeling of nutrient inputs to rivers on multiple scales. Our estimates for point source inputs (sewage and manure discharges) are more complete, updated, and detailed than

earlier estimates for China. Moreover, we present a novel formalized framework for multi-scale modeling of nutrients in rivers. This modeling approach to link biophysical and administrative scales is an important step toward an improved understanding of environmental impacts. To our knowledge, this is the first attempt to quantify the impacts on nutrient river pollution and source attribution for individual administrative units (2238 counties) for all of China. Moreover, the multi-scale approach helps to better formulate water pollution management strategies at the local level. Chinese governments have issued a series of policies to regulate the pollution from industrial livestock production.<sup>29,30,59,60</sup> Proposed measures include zoning for livestock production, manure treatment and utilization, integrated crop and livestock systems, biogas production and treatment, and end-of-pipe treatment technologies.<sup>29,59</sup> However, these policy recommendations are at the level of the seven regions in China,<sup>23,54</sup> which are large regions. The recommendations thus ignore subregion differences. Studies have illustrated that the effectiveness of measures can be influenced by local characteristics such as crop land capacity, farming scale, and runoff.<sup>61–63</sup> Additionally, our results confirm that pollution levels and dominant sources differ among counties. This implies that management options should be assessed and recommended on a site-specific basis rather than on a region-aggregated level. We believe that our multi-scale model outputs can support local decision making for various stakeholders. For instance, basin managers and associated county mayors could develop county-specific reduction targets and policy plans, in line with the relative shares of counties in the water pollution of larger basins. In addition, county mayors could choose effective solutions targeting dominant sources.

This multi-scale modeling framework has the potential for wider applicability in other regions that experience similar environmental problems. The principles of modeling on the intermediate units between the biophysical and administrative scales and of quantifying processes on different scales as appropriate could be applied to other world regions. The main challenge would be the availability of input data. For instance, if one has only national inputs, the advantage of highlighting the spatial variability would be constrained. Additionally, determining the appropriate scales for different processes takes effort and has to take into account both the available approaches and the scale of the available data. Moreover, our database of point sources is useful for studies on other pollutants in wastewater<sup>64</sup> or other processes such as nutrient pollution of groundwater.<sup>65</sup> We aim to further develop the model to understand the nutrient impacts on full water systems, including groundwater, coastal water and lakes, at multiple scales.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b07352.

Model description (S1), treatment efficiencies of wastewater treatment technologies (S2), centralized wastewater treatment plants database (S3), model evaluation (S4), and results (S5) (PDF)

WWTPs database (XLSX)

## AUTHOR INFORMATION

### Corresponding Authors

\*E mail: [xi.chen@wur.nl](mailto:xi.chen@wur.nl).

\*E mail: [malin1979@sjziam.ac.cn](mailto:malin1979@sjziam.ac.cn).

### ORCID

Xi Chen: 0000-0003-2799-1724

Lin Ma: 0000-0003-1761-0158

### Notes

The authors declare no competing financial interest.

## ACKNOWLEDGMENTS

We gratefully acknowledge the sponsors of this research: National Key R&D Program of China (2016YFE0103100), Wageningen Institute for Environment and Climate Research (WIMEK) of Wageningen University & Research, National Key Research and Development Program of China (2016YFD0800106), the Hundred Talent Program of the Chinese Academy of Science, and the Distinguished Young Scientists Project of Natural Science Foundation of Hebei (D2017503023).

## REFERENCES

- Glibert, P. M. Eutrophication, harmful algae and biodiversity—Challenging paradigms in a world of complex nutrient changes. *Mar. Pollut. Bull.* **2017**, *124* (2), 591–606.
- Kroeze, C.; Hofstra, N.; Ivens, W.; Löhr, A.; Stokral, M.; van Wijnen, J. The links between global carbon, water and nutrient cycles in an urbanizing world—the case of coastal eutrophication. *Curr. Opin. Env. Sust.* **2013**, *5* (6), 566–572.
- Galloway, J. N.; Townsend, A. R.; Erisman, J. W.; Bekunda, M.; Cai, Z.; Freney, J. R.; Martinelli, L. A.; Seitzinger, S. P.; Sutton, M. A. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. *Science* **2008**, *320* (5878), 889–892.
- Mayorga, E.; Seitzinger, S. P.; Harrison, J. A.; Dumont, E.; Beusen, A. H. W.; Bouwman, A. F.; Fekete, B. M.; Kroeze, C.; Van Drecht, G. Global nutrient export from Water Sheds 2 (NEWS 2): model development and implementation. *Environ. Model. Software* **2010**, *25* (7), 837–853.
- Beusen, A.; Van Beek, L.; Bouwman, A.; Mogollón, J.; Middelburg, J. Coupling global models for hydrology and nutrient loading to simulate nitrogen and phosphorus retention in surface water—description of IMAGE-GNM and analysis of performance. *Geosci. Model Dev.* **2015**, *8* (12), 4045.
- Stokral, M.; Ma, L.; Bai, Z.; Luan, S.; Kroeze, C.; Oenema, O.; Velthof, G.; Zhang, F. Alarming nutrient pollution of Chinese rivers as a result of agricultural transitions. *Environ. Res. Lett.* **2016**, *11* (2), 024014.
- Qu, H. J.; Kroeze, C. Past and future trends in nutrients export by rivers to the coastal waters of China. *Sci. Total Environ.* **2010**, *408* (9), 2075–2086.
- Liu, D.; Keesing, J. K.; He, P.; Wang, Z.; Shi, Y.; Wang, Y. The world's largest macroalgal bloom in the Yellow Sea, China: formation and implications. *Estuarine, Coastal Shelf Sci.* **2013**, *129*, 2–10.
- The bulletin of ecology and environment of China*; Ministry of Ecology and Environment of the People's Republic of China, 2017.
- The first national pollution census bulletin*; Ministry of Environmental Protection of the People's Republic of China; National Bureau of Statistics of the People's Republic of China; Ministry of Agriculture of the People's Republic of China, 2010.
- The first national pollution census bulletin*; Ministry of Environmental Protection of the People's Republic of China; National Bureau of Statistics of the People's Republic of China; Ministry of Agriculture of the People's Republic of China, 2010.
- Ma, L.; Velthof, G. L.; Wang, F. H.; Qin, W.; Zhang, W. F.; Liu, Z.; Zhang, Y.; Wei, J.; Lesschen, J. P.; Ma, W. Q.; et al. Nitrogen and phosphorus use efficiencies and losses in the food chain in China at

regional scales in 1980 and 2005. *Sci. Total Environ.* **2012**, *434*, 51–61.

(13) Wang, M.; Ma, L.; Stokral, M.; Ma, W.; Liu, X.; Kroeze, C. Hotspots for Nitrogen and Phosphorus Losses from Food Production in China: A County-Scale Analysis. *Environ. Sci. Technol.* **2018**, *52* (10), 5782–5791.

(14) Stokral, M.; Kroeze, C.; Wang, M.; Bai, Z.; Ma, L. The MARINA model (Model to Assess River Inputs of Nutrients to seAs): model description and results for China. *Sci. Total Environ.* **2016**, *562*, 869–888.

(15) Wang, F.; Dou, Z.; Ma, L.; Ma, W.; Sims, J. T.; Zhang, F. Nitrogen mass flow in China's animal production system and environmental implications. *J. Environ. Qual.* **2010**, *39* (5), 1537–1544.

(16) Wang, F.; Sims, J. T.; Ma, L.; Ma, W.; Dou, Z.; Zhang, F. The phosphorus footprint of China's food chain: implications for food security, natural resource management, and environmental quality. *J. Environ. Qual.* **2011**, *40* (4), 1081–1089.

(17) Bai, Z.; Ma, L.; Ma, W.; Qin, W.; Velthof, G. L.; Oenema, O.; Zhang, F. Changes in phosphorus use and losses in the food chain of China during 1950–2010 and forecasts for 2030. *Nutr Cycl Agroecosyst* **2016**, *104*, 361–372.

(18) Arnold, J.; Kinyri, J.; Srinivasan, R.; Williams, J.; Haney, E.; Neitsch, S. *SWAT 2012 input/output documentation*; Texas Water Resources Institute, 2013.

(19) Lindström, G.; Pers, C.; Rosberg, J.; Strömqvist, J.; Arheimer, B. Development and testing of the HYPE (Hydrological Predictions for the Environment) water quality model for different spatial scales. *Nord. Hydrol.* **2010**, *41* (3–4), 295–319.

(20) Schlueter, M.; McAllister, R.; Arlinghaus, R.; Bunnefeld, N.; Eisenack, K.; Hoelker, F.; Milner Gulland, E.; Müller, B.; Nicholson, E.; Quaas, M.; et al. New horizons for managing the environment: A review of coupled social-ecological systems modeling. *Nat. Resour. Model.* **2012**, *25* (1), 219–272.

(21) Boumans, R.; Roman, J.; Altman, I.; Kaufman, L. The Multiscale Integrated Model of Ecosystem Services (MIMES): Simulating the interactions of coupled human and natural systems. *Ecosyst. Serv.* **2015**, *12*, 30–41.

(22) Andrade, D. C.; Romeiro, A. R.; Boumans, R.; Sobrinho Paiva, R.; Tôsto, S. G. In *New methodological perspectives on the valuation of ecosystem services: toward a dynamic-integrated valuation approach, Embrapa Monitoramento por Satélite-Artigo em anais de congresso (ALICE), 2010*. In: *International society for ecological economics conference, 1, Oldenburg. Advancing sustainability in a time of crisis*; ISEE, Bremen, 2010.

(23) EPA, Clean Water Act. In *Section 502*; United States Environmental Protection Agency: 1972; Vol. 14.

(24) Wollheim, W. M.; Peterson, B. J.; Thomas, S. M.; Hopkinson, C.; Vörösmarty, C. Dynamics of N removal over annual time periods in a suburban river network. *J. Geophys. Res.* **2008**, *113* (G3), 1 DOI: [10.1029/2007JG000660](https://doi.org/10.1029/2007JG000660).

(25) Harrison, J. A.; Maranger, R. J.; Alexander, R. B.; Giblin, A. E.; Jacinthe, P.-A.; Mayorga, E.; Seitzinger, S. P.; Sobota, D. J.; Wollheim, W. M. The regional and global significance of nitrogen removal in lakes and reservoirs. *Biogeochemistry* **2009**, *93* (1–2), 143–157.

(26) McMahon, G.; Tervelt, L.; Donehoo, W. *Methods for estimating annual wastewater nutrient loads in the southeastern United States*; 2331–1258; Geological Survey, 2007.

(27) Liu, X.; Li, S. Temporal and spatial distribution of nutrient resource from livestock and poultry feces and its returning to cropland (in Chinese). *Transactions of the Chinese Society of Agricultural Engineering* **2018**, *34* (4), 1–14.

(28) Liu, X.; Xu, J.; Wang, F.; Zhang, F.; Ma, W. The resource and distribution of nitrogen nutrient in animal excretion in China (in Chinese). *Journal of Agricultural University Hebei* **2005**, *28* (5), 27–32.

(29) *Action plan for manure recycling and utilization (2017–2020)*. In Ministry of Agriculture and Rural Affairs of the People's Republic of China, 2017.

- (30) *Polution Prevention for Industrial Livestock Production*; No. 643; State Council of the People's Republic of China, 2014.
- (31) Ma, L.; Ma, W.; Velthof, G.; Wang, F.; Qin, W.; Zhang, F.; Oenema, O. Modeling nutrient flows in the food chain of China. *J. Environ. Qual.* **2010**, *39* (4), 1279–1289.
- (32) Liang, X.; Lettenmaier, D. P.; Wood, E. F.; Burges, S. J. A simple hydrologically based model of land surface water and energy fluxes for general circulation models. *J. Geophys. Res.* **1994**, *99* (D7), 14415–14428.
- (33) van Vliet, M. T.; Sheffield, J.; Wiberg, D.; Wood, E. F. Impacts of recent drought and warm years on water resources and electricity supply worldwide. *Environ. Res. Lett.* **2016**, *11* (12), 124021.
- (34) Van Vliet, M. T.; Wiberg, D.; Leduc, S.; Riahi, K. Power-generation system vulnerability and adaptation to changes in climate and water resources. *Nat. Clim. Change* **2016**, *6* (4), 375.
- (35) Data Center for Resources and Environmental Sciences Chinese Academy of Sciences In *Data Center for Resources and Environmental Sciences*; Chinese Academy of Sciences, 2014.
- (36) Data Center for Resources and Environmental Sciences Chinese Academy of Sciences In *Data Center for Resources and Environmental Sciences*; Chinese Academy of Sciences, 2012.
- (37) Jensen, L. S. Animal manure fertiliser value, crop utilisation and soil quality impacts. *Animal Manure Recycling: Treatment and Management* **2013**, 295–328.
- (38) Huang, S.; Ma, Y.; Bao, D.; Guo, D.; Zhang, S. Manures behave similar to superphosphate in phosphorus accumulation in long-term field soils. *Int. J. Plant. Prod.* **2011**, *5* (2), 135–146.
- (39) Li, G.; Li, H.; Leffelaar, P. A.; Shen, J.; Zhang, F. Characterization of phosphorus in animal manures collected from three (dairy, swine, and broiler) farms in China. *Plos One* **2014**, *9*, No. e102698.
- (40) Schoumans, O. *Phosphorus leaching from soils: process description, risk assessment and mitigation*; WUR Wageningen: UR, 2015.
- (41) *National list of operating centralized wastewater treatment plants*; Ministry of Ecology and Environment of the People's Republic of China, 2014.
- (42) *National Intensive Monitoring and Control Enterprise List No.116*; Ministry of Ecology and Environment of the People's Republic of China, 2016.
- (43) *China Urban-Rural Construction Statistical Yearbook 2014*; Ministry of Housing and Urban-Rural Development of the People's Republic of China, 2014.
- (44) MOHURD *China Urban-Rural Construction Statistical Yearbook 2012*; Ministry of Housing and Urban-Rural Development of the People's Republic of China.: 2012.
- (45) Van Drecht, G.; Bouwman, A. F.; Harrison, J.; Knoop, J. M. Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050. *Global. Biogeochem. Cy.* **2009**, *23* (4), n/a.
- (46) Bouwman, A. F.; Beusen, A. H. W.; Billen, G. Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050. *Global. Biogeochem. Cy.* **2009**, *23* (4), n/a.
- (47) Stokal, M. *River export of nutrients to the coastal waters of China: the MARINA model to assess sources, effects and solutions*. Wageningen University, 2016.
- (48) Wang, J.; Li, X.; Yan, W.; Wang, F.; Ma, P. Watershed nitrogen export model related to changing nitrogen balance and hydrology in the Changjiang River basin. *Nutr. Cycling Agroecosyst.* **2014**, *98* (1), 87–95.
- (49) Yan, W.; Mayorga, E.; Li, X.; Seitzinger, S. P.; Bouwman, A. Increasing anthropogenic nitrogen inputs and riverine DIN exports from the Changjiang River basin under changing human pressures. *Global. Biogeochem. Cy.* **2010**, *24* (4), n/a.
- (50) Aber, J. D. Nitrogen cycling and nitrogen saturation in temperate forest ecosystems. *Trends Ecol. Evol.* **1992**, *7* (7), 220–224.
- (51) Kahl, J. S.; Norton, S. A.; Fernandez, I. J.; Nadelhoffer, K. J.; Driscoll, C. T.; Aber, J. D. Experimental inducement of nitrogen saturation at the watershed scale. *Environ. Sci. Technol.* **1993**, *27* (3), 565–568.
- (52) Harrison, J. A.; Bouwman, A.; Mayorga, E.; Seitzinger, S., Magnitudes and sources of dissolved inorganic phosphorus inputs to surface fresh waters and the coastal zone: A new global model. *Global. Biogeochem. Cy.* **2010**, *24*, (1), n/a
- (53) Bai, Z.; Ma, L.; Jin, S.; Ma, W.; Velthof, G. L.; Oenema, O.; Liu, L.; Chadwick, D.; Zhang, F. Nitrogen, phosphorus, and potassium flows through the manure management chain in China. *Environ. Sci. Technol.* **2016**, *50* (24), 13409–13418.
- (54) Zhao, Z.; Bai, Z.; Winiwarter, W.; Kiesewetter, G.; Heyes, C.; Ma, L. Mitigating ammonia emission from agriculture reduces PM<sub>2.5</sub> pollution in the Hai River Basin in China. *Sci. Total Environ.* **2017**, *609*, 1152–1160.
- (55) Beusen, A.; Bouwman, A.; Van Beek, L.; Mogollón, J.; Middelburg, J. Global riverine N and P transport to ocean increased during the twentieth century despite increased retention along the aquatic continuum. *Biogeosci. Disc.* **2015**, *12* (23), 20123.
- (56) Liu, X.; Beusen, A. H.; Van Beek, L. P.; Mogollón, J. M.; Ran, X.; Bouwman, A. F. Exploring spatiotemporal changes of the Yangtze River (Changjiang) nitrogen and phosphorus sources, retention and export to the East China Sea and Yellow Sea. *Water Res.* **2018**, *142*, 246.
- (57) Liu, S. M.; Zhang, J.; Chen, H.; Wu, Y.; Xiong, H.; Zhang, Z. Nutrients in the Changjiang and its tributaries. *Biogeochemistry* **2003**, *62* (1), 1–18.
- (58) Shen, Z.-L.; Liu, Q. Nutrients in the changjiang river. *Environ. Monit. Assess.* **2009**, *153* (1–4), 27–44.
- (59) *The planning of pollution prevention for livestock production for '12th five year'*; Ministry of Environment Protection P.R. China; Ministry of Agriculture P.R. China, 2012.
- (60) Jin, S.; Han, D.; Wu, N. Evaluation on prevention policies for livestock and poultry pollution in China (in Chinese). *Issue in Agricultural Economy* **2018**, *3*, 119–126.
- (61) Wang, Z.; Qian, X.; Shen, G. Analysis and Application of Pollution Reduction Techniques for Scale Livestock and Poultry Farms in Shanghai (in Chinese). *Journal of Agro-Environment Science* **2014**, *33* (10), 2030–2035.
- (62) Li, J. Innovative research on disposal and recycling modes of waste from livestock and poultry breeding (in Chinese). *Quality and Safety of Agro-products* **2018**, No. 1, 3–7.
- (63) Xuan, M.; Xu, Z.; Wu, G.; Ou, W.; Li, J.; He, W. Analysis of utilization of fecal resources in large-scale livestock and poultry breeding in China (in Chinese). *Journal of Agricultural Resources and Environment* **2018**, *35* (2), 126–132.
- (64) Kroeze, C.; Gabbert, S.; Hofstra, N.; Koelmans, A. A.; Li, A.; Löhr, A.; Ludwig, F.; Stokal, M.; Verburg, C.; Vermeulen, L.; et al. Global modeling of surface water quality: a multi-pollutant approach. *Curr. Opin. Env. Sust.* **2016**, *23*, 35–45.
- (65) Bai, Z.; Lu, J.; Zhao, H.; Velthof, G. L.; Oenema, O.; Chadwick, D.; Williams, J. R.; Jin, S.; Liu, H.; Wang, M.; et al. Designing Vulnerable Zones of Nitrogen and Phosphorus Transfers To Control Water Pollution in China. *Environ. Sci. Technol.* **2018**, *52*, 8987–8988.