Supporting Information for:

Hotspots for Nitrogen and Phosphorus Losses from Food Production in China: a County-scale Analysis

Mengru Wang,[†,‡,*] Lin Ma,[†,*] Maryna Strokal,[‡] Wenqi Ma,[§] Xuejun Liu,[⊥] Carolien Kroeze[‡]

† Key Laboratory of Agricultural Water Resources, Center for Agricultural Resources Research, Institute of Genetics and Developmental Biology, Chinese Academy of Sciences, 286 Huaizhong Road, Shijiazhuang 050021, China

‡ Water System and Global Change Analysis Group, Wageningen University & Research, Droevendaalsesteeg 4, 6708 PB Wageningen, The Netherlands

§ College of Resources and Environmental Sciences, Agricultural University of Hebei, Baoding, 071001, China

⊥ College of Resources and Environmental Sciences, China Agricultural University, Beijing 100193, China

*Corresponding authors (Email: mengru.wang@wur.nl, malin1979@sjziam.ac.cn)

S1 Agricultural transition in China

We analyzed N and P losses from food production for 1990, 2000 and 2012. We selected these years to reflect the period during which the transition took place in food production in agriculture in China. Chinese food production has been shifting from traditional production systems to intensive industrial production systems since 1990s.[1-2] This transition includes changes in both crop and animal production systems. Traditional-oriented agriculture was dominant in the 1990s, before the transition in agriculture started (Figure S1A). Traditional animal systems are usually small in size and typically combined with crop production. Crop residues and by-products are used as feed to animals. Animals in a way convert crop residues to organic fertilizers for crops. Therefore, the inputs to this system from synthetic fertilizers, animal feed and other sources are relative low. The productivity of animal products is generally low in these traditional systems because of the inefficient animal species that are raised.[3-5] Industrial, highly intensive systems dominate agricultural production in large parts in China since 2000s. This reflects the ongoing transition in Chinese food production (Figure S1B). Industrial animal production systems are large in size and are separated from crop production. These industrial farms mainly use concentrated feed, the quality of which is higher than crop residues in the traditional systems. Genetically improved species are raised in these farms to increase their productivity. The animal manure is usually collected, and discharged to surface waters or landfills without treatment.[1, 3-6] Consequently, the nutrient inputs to current food production systems as synthetic fertilizers and animal feed are relatively high; there is little nutrient exchange between crop and animal production within the system.

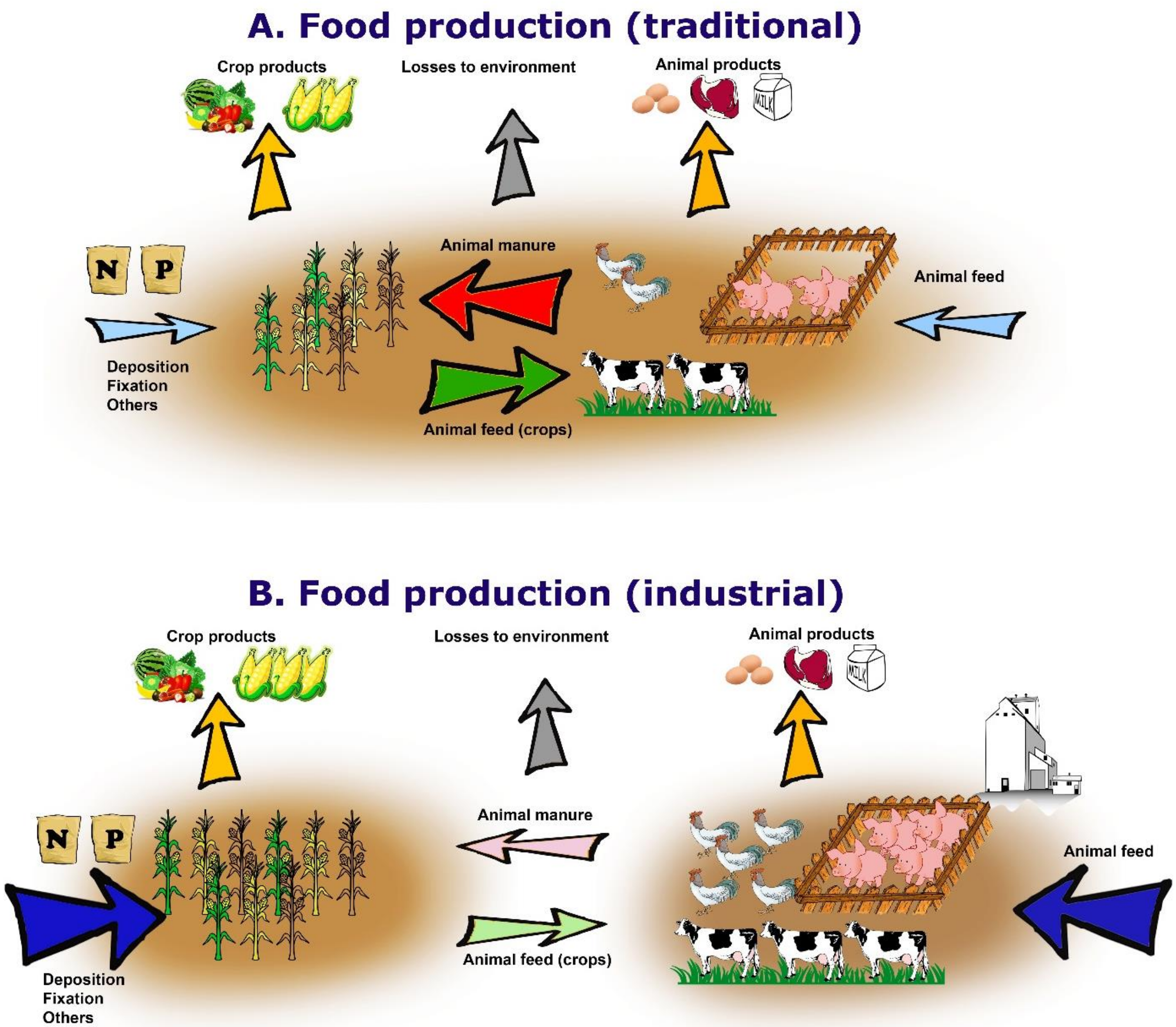

Figure S1 Conceptual structure of (A) traditional-oriented food (crop and animal) production systems in China, and (B) industrial-oriented food (crop and animal) production system. System A represents the food production system before the agricultural transition. System B illustrates food production systems after the transition. Nitrogen and phosphorus inputs to the system are presented by blue arrows. Nitrogen and phosphorus output via main products are presented by yellow arrows. The nutrient exchange within the crop and animal production system are presented by red (animal manure) and green (animal feed made of crops) arrows. Nutrient losses to the environment are presented by grey arrows.

S2 *NUFER* model description

The original *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model was developed by Ma et al.[7] to quantify N and P flows in the food chain of China. The food chain in *NUFER* consists of four compartments: crop production, animal production, food processing and food consumption. The crop and animal production compartments include 18 crop types and 11 animal categories.[7] N and P enter the food chain via synthetic fertilizers, biological N fixation, atmospheric N deposition and imported products. They leave the food chain via exported food products, and N and P losses to the air and waters (ground and surface water). The nutrient cycling among different compartments is included via residues, crop and animal products, manure and human waste.

For each compartment, input-output balances of N and P are calculated using the equations that are available in the supporting information of Ma et al.[7] at https://dl.sciencesocieties.org/publications/jeq/supplements/39/jeq39-4-1279.pdf. In these equations the total N and P outputs from one compartment equals to the total N and P inputs to that compartment. The N and P content in the imports, exports of and exchange within the food chain are calculated on the basis of the input data described in section S2.1 and the input-output balance approach. The N and P use efficiencies in crop production and animal production are defined by the ratio of N and P in the main crop and animal products and the total N and P input of crop and animal production. Details on the calculations in N and P use efficiencies are explained in section S2.5.

The original *NUFER* model calculates nutrient flows at the national level for each year from 1980 to 2010, and for the year 2030. In addition, regional nutrient flows can be calculated for 31 provinces for 2005. In this study we applied *NUFER* for all Chinese counties using county information[8-10] for 1990, 2000 and 2012, which was not done before. We used the crop and animal production compartments in *NUFER* and improved *NUFER* by including dry N deposition that was not included in the original model. The details in applying *NUFER* to the county scale is described in the following sections S2.1 – S2.5.

S2.1 County model inputs

The input of original *NUFER* includes annual inputs data for the 31 provinces and China: (i) human activities in the food chain (e.g., fertilizer and animal manure applications, cultivated areas, animal numbers), (ii) transformation and partitioning coefficients (e.g., the partitioning of animal products into edible and other parts, N and P excretion values per animal category), (iii) N and P content and loss factors (e.g., N and P content in harvested crop and in animal products, and N and P leaching factor in crop production). The model inputs for human activities in the food chain mainly come from the statistical sources[11-13] and survey reports (survey reports from Agricultural University of Hebei, China Agricultural University, and the National Agro-Technical Extension and Service Center during 1999 to 2008). The transformation and partitioning coefficients, N and P content and loss factors are derived from literature sources and from surveys data. The information about these literature and surveys could be found in Ma et al.[7] and Ma et al.[14].

We ran the *NUFER* model all counties (more than 3,000) in China (Figures 1 and 2). County-scale model input datasets were developed, including (i) human activities in the food chain, (ii) transformation and partitioning coefficients, (iii) N and P content and loss factors. Model inputs for human activities in crop and animal production are from the county statistical reports[8-10]. Other model inputs related to human activities are mainly from statistical sources[11-12] and survey reports (survey reports from Agricultural University of Hebei, China Agricultural University, and the National Agro-Technical Extension and Service Center during 1999 to 2011). The transformation and partitioning coefficients, N and P content factors are from Ma et al.[7] available at https://dl.sciencesocieties.org/publications/jeq/supplements/39/jeq39-4-1279.pdf. We updated in *NUFER* the N and P loss factors for 1990, 2000 and 2012 based on the approach that is described in the Supporting Information of Ma et al.[14] available at http://www.sciencedirect.com/science/article/pii/S0048969712003890?via%3Dihub#s0090. This includes the N and P loss factors for leaching, surface runoff and erosion in crop production, and N and P losses to waters from direct discharge of animal manure in animal production, and for emissions of ammonia (NH_3) and nitrous oxide (N_2O) from crop and animal production systems to the air. These factors for China and their ranges for counties are summarized in Tables S8 – S9 as the losses by sources to the total N and P input to crop production or total N and P in animal manure in animal production.

S2.2 Animal numbers on the county scale

The inputs of animal numbers in *NUFER* are for pig, dairy cow, beef cattle, other cattle, poultry, sheep and goat, horse, mule and donkey, and rabbit. However, animal numbers are only available for big animals (sum of pig, cattle, horse, mule and donkey), pig and poultry in the county dataset in 2012 (See Table S1). Animal numbers for other animals are derived based on the available animal numbers in the county dataset in 2012 and the ratio of big animals, pig and poultry to other animals on the provincial level in 2012.[15] Frist, the animal numbers by categories that need to be derived on the county scale in 2012 for *NUFER* was identified (Table S2). Second, the identified required animal numbers were derived based on Equations 1-12 in Box S1.

Table S1 Available animal numbers and animal products in the county dataset for 2012.

Animal numbers and products	Unit	Variable in Box S1
Big animals (in stock)	1000 head	$N_{big\ animals,\ in,\ c}$
Pig (in stock and slaughtered)	1000 head	n.a. [ii]
Egg	kton	$T_{egg,\ poultry,\ c}$
Milk	kton	$T_{milk,\ dairy\ cow,\ c}$
Total meat [i]	kton	$T_{total\ meat,\ all\ animals,\ c}$
Pork	kton	$T_{pork,\ pig,\ c}$

[i] Total meat includes meat from all animals including pig, cattle, sheep and goat, poultry and rabbit
[ii] Not available

Table S2 Required animal numbers for 11 animal categories in the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. in 2012 on the county scale.

Animal category	Number type	Need to derive	Variable in Box S1
Pig	slaughtered	no	n.a.
Dairy cow	in stock	yes	$N_{dairy\ cow,\ in,\ c}$
Beefcattle [i]	slaughtered	yes	$N_{beefcattle\ and\ othercattle,\ sl,\ c}$
Othercattle [i]	in stock	no	n.a.
Poultry	slaughtered	yes	$N_{poultry,\ sl,\ c}$
Sheep and goat	slaughtered	yes	$N_{sheep\ and\ goat,\ sl,\ c}$
Horse	in stock	yes	$N_{horse,\ in,\ c}$
Mule and donkey	in stock	yes	$N_{mule\ and\ donkey,\ in,\ c}$
Rabbit [ii]	slaughtered	no	n.a. [iii]

[i] Other cattle was combined with beefcattle in calculation the other cattle numbers are relatively small according to MOA [15]
[ii] Numbers of rabbits were ignored because rabbit numbers are relatively small according to MOA [15]
[iii] Not available

Box S1 Equations to animal numbers on the county scale in 2012.

Deriving animal numbers on the county scale in 2012

For mule and donkey, horse in stock

$N_{\text{mule and donkey, in, c}} = N_{\text{big animals, in, c}} * (N_{\text{mule and donkey, in, p}} / N_{\text{big animals, in, p}})^{1}$ (1)

$N_{\text{horse, in, c}} = N_{\text{big animals, in, c}} * (N_{\text{horse, in, p}} / N_{\text{big animals, in, p}})^{1}$ (2)

For dairy cow in stock

$A_{\text{milk, dairy cow, p}} = T_{\text{milk, dairy cow, p}} / N_{\text{dairy cow, in, p}}{}^{1,1}$ (3)

$N_{\text{dairy cow, in, c}} = T_{\text{milk, dairy cow, c}} / A_{\text{milk, dairy cow, p}}{}^{1,2}$ (4)

For slaughtered beef cattle and other cattle

$N_{\text{beefcattle and othercattle, in, c}} = N_{\text{big animals, in, c}} * (N_{\text{beefcattle and othercattle, in, p}} / N_{\text{big animals, in, p}})$ (5)

$R_{\text{beefcattle and othercattle, sl/in, p}} = N_{\text{beefcattle and othercattle, sl, p}} / N_{\text{beefcattle and othercattle, in, p}}{}^{1,1}$ (6)

$N_{\text{beefcattle and othercattle, sl, c}} = N_{\text{beefcattle and othercattle, in, c}} * R_{\text{beefcattle and othercattle, sl/in, p}}{}^{1,3}$ (7)

For slaughtered poultry

$A_{\text{egg, poultry, p}} = T_{\text{egg, poultry, p}} / N_{\text{poultry, sl, p}}{}^{1,2}$ (8)

$N_{\text{poultry, sl, c}} = T_{\text{egg, poultry, c}} / A_{\text{egg, poultry, p}}{}^{1,2}$ (9)

For sheep and goat

$T_{\text{sheep and goat meat, sheep and goat, c}} = T_{\text{total meat, all animals, c}} * (T_{\text{sheep and goat meat, sheep and goat, p}} / T_{\text{total meat, all animals, p}})^{2}$ (10)

$A_{\text{sheep and goat meat, sheep and goat, p}} = T_{\text{sheep and goat meat, sheep and goat, p}} / N_{\text{sheep and goat, sl, p}}{}^{1,2}$ (11)

$N_{\text{sheep and goat, sl, c}} = T_{\text{sheep and goat meat, sheep and goat, c}} / A_{\text{sheep and goat meat, sheep and goat, p}}{}^{1,2}$ (12)

[1] $\mathbf{N}_{\textbf{animal type, in/sl, c/p}}$ refers to the animal number (N) (by animal category) that is in stock (in) or slaughtered (sl) on the county (c) or provincial (p) level in 2012.
[2] $\mathbf{A}_{\textbf{product, animal type, c/p}}$ refers to the average production (A) of animal product (product) (e.g., milk, pork) per animal (by animal category, e.g., dairy cow, pig) on the county (c) or provincial (p) level in 2012; and $\mathbf{T}_{\textbf{product, animal type, c/p}}$ refers the total production (T) of animal product (product) from animals (by animal category) on the county(c) or provincial(p) level in 2012.
[3] $\mathbf{R}_{\textbf{animal type, sl/in, c/p}}$ represents the ratio (R) of animals (by animal category) that are slaughtered (sl) to that are in stock (in) on the county(c) or provincial(p) level in 2012.

Units of all $N_{\text{animal type, in/sl, c/p}}$ variables are 1000 head year^{-1}, of $A_{\text{product, animal type, c/p}}$ and $T_{\text{product, animal type, c/p}}$ variables are kton year^{-1}.

S2.3 Livestock unit conversion

Livestock unit (LU, also referred to as cow equivalents) is used as unit for comparing total animal numbers of all categories among hotspots and non-hotspots in this study. Exchange ratios for livestock unit are derived based on feed requirements or metabolic weight, and are used to convert animal numbers to LU. The exchange ratios we used are shown in Table S3. The animal numbers of different animal types were converted to livestock units of dairy cows using these exchange ratios. For poultry an average (0.01) of exchange ratios for broilers and layer hens is used. The converted results of animal livestock units are used in Figures 3 and 4.

Table S3 Exchange ratios for livestock unit in China that are derived based on Eurostat [16].

Animal Type	Exchange ratio
Dairy cow	1
Beefcattle	1
Othercattle	1
Pig	0.3
Poultry (broiler)	0.007
Poultry (layer)	0.014
Sheep and goat	0.1
Horse	1
Mule and donkey	1
Rabbit	0.2

S2.4 Atmospheric N deposition

Atmospheric N deposition is considered an nutrient input to land in *NUFER*. However, the original *NUFER* model only included wet N deposition. Dry deposition in China has been reported to increase over the past decades.[17-19] The annual dry deposition was comparable to wet deposition from 2010 to 2014.[19] Therefore, ignoring dry N deposition in *NUFER* may lead to an underestimation of N inputs to crop production in China.

In this study, we included both wet N deposition and dry N deposition as N inputs to crop production in *NUFER*. National averages for wet N deposition and dry N deposition for 1990, 2000, and 2012 were derived based on monitoring and modelling data from Xu et al.[19] and Liu et al.[20]. These national averages were used for all counties in *NUFER* (see Box S2, $Ic_{wet\ deposition}$).

The trends in bulk N deposition from 1980 to 2010 reported by Liu et al.[20] were used to derive wet N deposition rates of China in 1990 and 2000. Bulk deposition is a simple measure of N input from precipitation, including wet N deposition and a small fraction of the dry deposition. In this study, we used bulk deposition rates to present wet deposition rate. The linear regression function of bulk deposition rate from 1980 to 2010 (Box S2) in Liu et al.[20] was used to calculate the bulk N deposition rates of China in 1990 and 2000. The wet deposition rates of China in 1990 and 2000 were calculated to be equal to bulk deposition rates and are shown in Table S5. The wet deposition rate of China in 2012 was derived based on the average wet deposition of monitoring sites in China from 2006 to 2010 by Xu et al.[19] and is shown in Table S5.

Dry N deposition was calculated as wet N deposition multiplied by the ratio of dry N deposition to wet N deposition. The ratio of dry N deposition to wet N deposition of China was reported to be around 1:1 by Xu et al.[19] from 2006 to 2014 (See Table S4). Assuming that the ratio of dry N deposition to wet N deposition does not change much over time, this ratio was 1:1 for both 1990 and 2000. For 1990 and 2000, the dry N deposition rates were calculated as wet N deposition multiply the ratio of dry N deposition to wet N deposition (Table S5). The dry deposition rate of China in 2012 was derived based on the average dry deposition of monitoring sites in China from 2006 to 2010 by Xu et al.[19] and is shown in Table S5.

Box S2 Calculation method of bulk N deposition in 1990 and 2000 in China. The linear regression function is from Liu et al.[20].

Linear regression equation of bulk N deposition in China:

$y = 0.411x - 804.353$

x is the year(time), y is the bulk N deposition

Bulk N deposition in 1990 and 2000 in China:

$$\text{bulk N deposition}_{1990} = 0.411 * 1990 - 804.353 \quad (1)$$

$$\text{bulk N deposition}_{2000} = 0.411 * 2000 - 804.353 \quad (2)$$

Table S4 Ratio of dry N deposition to wet N deposition. The average wet and dry N deposition from 2006 to 2014 is from Xu et al.[19].

Region	Average dry N deposition from 2006 to 2014	Average wet N deposition from 2006 to 2014	Ratio of dry N deposition to wet N deposition
	kg N ha^{-1} year^{-1}	kg N ha^{-1} year^{-1}	-
China	18.7	18.2	1

Table S5 Wet N deposition (kg N ha^{-1} year^{-1}) and dry N deposition (kg N ha^{-1} year^{-1}) over agricultural land in 1990, 2000 and 2012 in China.

	1990[i]	2000[i]	2012[ii]
	kg N ha^{-1} year^{-1}	kg N ha^{-1} year^{-1}	kg N ha^{-1} year^{-1}
Wet N deposition	13.5	17.6	18.2
Dry N deposition	13.5	17.6	18.7

[i] The wet and dry N deposition in 1990 and 2000 were calculated according to the method described in the text and Equations in Box S2.
[ii] The wet and dry N deposition in 2012 were from Xu et al.[19].

S2.5 Calculating N and P losses, N and P use efficiencies

We calculated N and P losses from food production using the *NUFER* model for 1990, 2000 and 2012 for all counties in China. Losses include leaching, surface runoff and erosion in crop production, and N and P losses from direct discharge of animal manure in animal production to waters, and from emissions of ammonia (NH_3) and nitrous oxide (N_2O) from crop and animal production systems to the air. Losses of NH_3 and N_2O emissions in crop production were calculated as N-source specific fractions of N inputs via fertilizers (synthetic fertilizer and animal manure). N losses by surface runoff, leaching and erosion were quantified as soil and climate specific fractions of the N surplus, while P losses by surface runoff, leaching and erosion were quantified as the climate, soil type and geomorphology specific fractions of the total P input and P in soil. Losses of NH_3, N_2O emissions and direct discharge of animal manure were calculated as specific fractions to the total N or P in animal manure. The specific fractions are part of the *NUFER* model inputs (N and P loss factors) and the approach to derive these factors is as we described in the section S2.1. It is assumed in *NUFER* that animal manure is treated only if farmers recycle the manure on land. This means that N and P in animal manure that is removed from treatment will be used as fertilizer for crops. The N and P stayed in animal manure after treatment will be discharged to waters. The manure treatment was therefore considered implicitly in this study. The rates for direct discharge of manure differ among animal types as indicated in Table S2 in the Supporting Information of Ma et al.[14] available at https://www.sciencedirect.com/science/article/pii/S0048969712003890?via%3Dihub#s0090 and differ among years according to Ma et al.[14] and Hou et al.[21]. Food wastes from food production, processing and consumption that are emitted to non-arable land may result in indirect N and P losses to the environment. These indirect N and P losses from non-arable land were not considered in this study.

N and P use efficiencies of food production were quantified using the *NUFER* model for 1990, 2000 and 2012 in this study. The N and P use efficiencies were calculated as outputs of N and P via main products divided by the total inputs of N and P to this system (see Equation in Box S3). The results of N and P use efficiencies are shown in Figures 3 and 4.

Box S3 Equation to quantify nitrogen use efficiency (NUE) and phosphorus use efficiency (PUE) of food production by the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. The equation is based on Wang et al.[22].

$$NUE \text{ or } PUE = (O_{main\ product} / I) \times 100\% \quad (1)$$

Where,

I	Input of N or P to food production including crop and animal production
Ic	Input of N or P to crop production
Ia	Input of N or P to animal production
O	Output of N or P from food production including crop and animal production
Oc	Output of N or P from crop production
Oa	Output of N or P from animal production
$O_{main\ product}$	$Oc_{crop\ product} - Ia_{crop} + Oa_{main\ product}$
$Oc_{main\ product}$	$Oc_{crop\ product}$
$Oa_{main\ product}$	$Oa_{meat} + Oa_{egg} + Oa_{milk} + Oa_{fish} + Oa_{by\text{-}products\ to\ food}$
I	$Ic_{fertilizer} + Ic_{human\ waste} + Ic_{wet\ deposition} + Ic_{dry\ deposition} + Ic_{seed} + Ic_{fixation} + Ic_{irrigation} + Ic_{straw\ residues} + Ic_{crop\ by\text{-}products} + Ia_{crop\ by\text{-}products} + Ia_{animal\ by\text{-}products} + Ia_{kitchen\ residues} + Ia_{other\ feed}$
$Ic_{wet\ deposition}$	Input of N via atmospheric wet nitrogen deposition
$Ic_{dry\ deposition}$	Input of N via atmospheric dry nitrogen deposition
$Ic_{fertilizer}$	Input of N or P via synthetic fertilizer including single and compound fertilizer
Ic_{manure}	Input of N or P via animal manure application on land
$Ic_{human\ waste}$	Input of N or P via human waste application on land
Ic_{seed}	Input of N or P via seed
$Ic_{fixation}$	Input of N via biological nitrogen deposition
$Ic_{irrigation}$	Input of N or P via irrigation
$Ic_{straw\ residues}$	Input of N or P via straw residues
$Ic_{crop\ by\text{-}products}$	Input of N or P via crop by-products from food processing
$Ia_{crop\ products}$	Input of N or P via grain and roots
Ia_{straw}	Input of N or P via straw
Ia_{grass}	Input of N or P via grass from natural grass land
$Ia_{crop\ by\text{-}products}$	Input of N or P via crop by-products from food processing
$Ia_{animal\ by\text{-}products}$	Input of N or P via animal by-products from food processing
$Ia_{kitchen\ residues}$	Input of N or P via kitchen residues
$Ia_{other\ feed}$	Input of N or P via rough grazing and forests, and other sources

Units of all I, Ic, Ia, O, Oc and Oa variables are kton N(P) year^{-1}.

S2.6 Model evaluation

NUFER has been widely use in Chinse studies to analyze N and P flows, as well as the associated N and P use efficiencies in food production and consumption.[5-6, 22-24] This model uses data from Chinese statistics as model input. There might be uncertainties in the statistics, but these statistical yearbooks are known to be the most reliable data source in China. The model coefficients (e.g., nutrient content in crops, nutrient loss factors) were derived from other peer reviewed papers and interviews of farmers in China.[7] In this study, we improved the *NUFER* model and applied it to the counties in China. Our results are comparable with other studies on N and P losses in China[25-28] (Table 1) as we described in the discussion section in the main text. Therefore, despite the uncertainties related to the statistics, model coefficients, and assumptions that are used in model calculations, we evaluate that our model produce reasonable results for N and P losses in China.

S3 Other supplementary Tables and Figures

Figures

Figure S1 Conceptual structure of (A) traditional-oriented food (crop and animal) production systems in China, and (B) industrial-oriented food (crop and animal) production system. System A represents the food production system before the agricultural transition. System B illustrates food production systems after the transition. Nitrogen and phosphorus inputs to the system are presented by blue arrows. Nitrogen and phosphorus output via main products are presented by yellow arrows. The nutrient exchange within the crop and animal production system are presented by red (animal manure) and green (animal feed made of crops) arrows. Nutrient losses to the environment are presented by grey arrows.

Figure S2 Nitrogen (N) losses (kg N km^{-2} $year^{-1}$) to the air and waters from leaching, runoff and erosion, direct discharge of manure, ammonia (NH_3) and nitrous oxide (N_2O) emissions, and the total N losses (kg N km^{-2} $year^{-1}$) from food production in 2000. The N losses were quantified using the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. The intervals for the four groups in this figure were defined based on quantiles (25%, 50%, 75%) of N losses of all counties in 2012: group I (0-25%), group II (25-50%), group III (50-75%), group IV (75-100%). Counties in group IV were qualified as hotspots. The names of the Agro-Ecological Zones are available in Figure S4.

Figure S3 Phosphorus (P) losses (kg P km^{-2} $year^{-1}$) to waters from leaching, runoff and erosion, direct discharge of manure, and the total P losses (kg P km^{-2} $year^{-1}$) from food production in 2000. The P losses were quantified using the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. The intervals for the four groups in this figure were defined based on quantiles (25%, 50%, 75%) of P losses of all counties in 2012: group I (0-25%), group II (25-50%), group III (50-75%), group IV (75-100%). Counties in group IV were qualified as hotspots. The names of the Agro-Ecological Zones are available in Figure S4.

Figure S4 Boxplots for nitrogen (N): synthetic fertilizer (ton km^{-2} $year^{-1}$), animal number in livestock unit (lu km^{-2} $year^{-1}$, see Supporting Information for converting animal numbers in livestock unit), share of sown area of vegetable and fruit to the total sown area (%), N use efficiency (NUE) of food production, urban population (% of the total population), rural labor (capita km^{-2} $year^{-1}$) among the four groups of total N losses (see Figure 1) in 1990 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

Figure S5 Boxplots of for phosphorus (P): synthetic fertilizer (ton km^{-2} $year^{-1}$), animal number in livestock unit (lu km^{-2} $year^{-1}$, see Supporting Information for converting animal numbers in livestock unit), share of sown area of vegetable and fruit to the total sown area (%), P use efficiency (PUE) of food production, urban population (% of the total population), rural labor (capita km^{-2} $year^{-1}$) among the four groups of total P losses (see Figure 1) in 1990 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

Figure S6 Boxplots for nitrogen (N): intensive pig production (> 50 head pig per farm) (%), intensive dairy cow production (> 5 dairy cow per farm) (%), intensive beefcattle production (> 50 head beefcattle per farm) (%), intensive poultry (layers) production (> 500 layers per farm) (%), and intensive poultry (broilers) production (> 2000 broilers per farm) (%) among the four groups of N losses from direct discharge of manure to waters (see Figure 1) in 2012 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

Figure S7 Boxplots for phosphorus (P): intensive pig production (> 50 head pig per farm) (%), intensive dairy cow production (> 5 dairy cow per farm) (%), intensive beefcattle production (> 50 head beefcattle per farm) (%), intensive poultry (layers) production (>

500 layers per farm) (%), and intensive poultry (broilers) production (> 2000 broilers per farm) (%) among the four groups of P losses from direct discharge of manure to waters (see Figure 1) in 2012 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

Figure S8 Nine Agro-Ecological Zones (AEZs) in China. The AEZs are: (I) Northeast China, (II) Inner Mongolia and Great Wall Vicinity, (III) North China Plain, (IV) Loess Plateau, (V) Middle and Lower Yangtze River, (VI) Southwest China, (VII) South China, (VIII) Gansu and Xinjiang, and (IX) Tibetan Plateau. Source: Sun and Shen [29].

Figure S9 The nitrogen (N) fertilizer use, phosphorus (P) fertilizer use, crop yields, animal number, arable land for the nine Agro-Ecological Zones (AEZs) in 1990 and 2012. The nine AEZs include: (I) Northeast China, (II) Inner Mongolia and Great Wall Vicinity, (III) Huang Huai Hai, (IV) Loess Plateau, (V) Middle and Lower Yangtze River, (VI) Southwest China, (VII) South China, (VIII) Gansu and Xinjiang, and (IX) Tibetan Plateau. The location of the AEZs are available in Figure S8.

Tables

Table S1 Available animal numbers and animal products in the county dataset for 2012.

Table S2 Required animal numbers for 11 animal categories in the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. in 2012 on the county scale.

Table S3 Exchange ratios for livestock unit in China that are derived based on Eurostat[16].

Table S4 Ratio of dry N deposition to wet N deposition. The average wet and dry N deposition from 2006 to 2014 is from Xu et al.[19].

Table S5 Wet N deposition (kg N ha^{-1} $year^{-1}$) and dry N deposition (kg N ha^{-1} $year^{-1}$) over agricultural land in 1990, 2000 and 2012 in China.

Table S6 Losses in hotspots (% of the total losses in China) for N from leaching, runoff and erosion, direct discharge of animal manure, ammonia (NH_3) and nitrous oxide (N_2O)

emission, and for P from leaching, runoff and erosion, and direct discharge of animal manure, and for total N and total P losses from food production, and the associated hotspot area (% of the total area, and the hotspot area in km^2). The location of the hotspots are shown in Figures 1 and 2.

Table S7 Mean synthetic fertilizer application, animal numbers, share of sown area of vegetable and fruit to the total sown area, N and P use efficiencies (NUE and PUE) of crop and animal production, urban population, total output value of agriculture and forestry, and farmer's income of all counties in group I, II and III, and of all counties in group IV for total nitrogen (TN) and total phosphorus (TP)losses in 2012. The groups of the counties are shown in Figures 1 and 2.

Table S8 Nitrogen (N) and Phosphorus (P) loss factors for leaching, surface runoff, erosion to waters, ammonia (NH_3) and nitrous oxide (N_2O) emissions to the air from crop production of China for 1990, 2000 and 2012. The range (minimum and maximum) of N and P loss factors for Chinese counties are shown in the brackets in the table. These factors are derived based on the study of Ma et al.[14].

Table S9 Nitrogen (N) and Phosphorus (P) loss factors for direct discharge of manure to waters, ammonia (NH_3) and nitrous oxide (N_2O) emissions to the air from animal manure of China for 1990, 2000 and 2012. The range (minimum and maximum) of N and P loss factors for Chinese counties are shown in the brackets in the table. These factors are derived based on the study of Ma et al.[14].

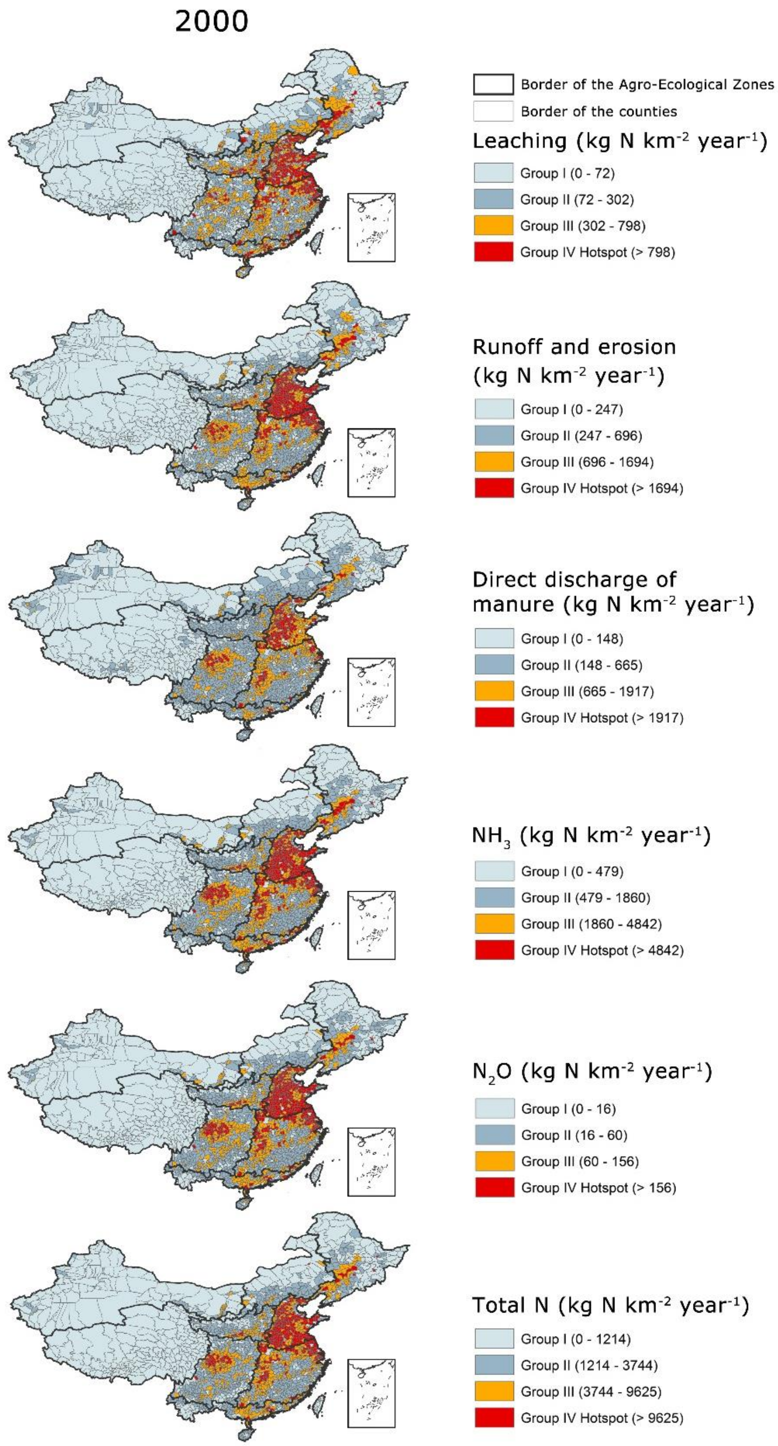

Figure S2 Nitrogen (N) losses (kg N km^{-2} year^{-1}) to the air and waters from leaching, runoff and erosion, direct discharge of manure, ammonia (NH_3) and nitrous oxide (N_2O) emissions, and the total N losses (kg N km^{-2} year^{-1}) from food production in 2000. The N losses were quantified using the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. The intervals for the four groups in this figure were defined based on quantiles (25%, 50%, 75%) of N losses of all counties in 2012: group I (0-25%), group II (25-50%), group III (50-75%), group IV (75-100%). Counties in group IV were qualified as hotspots. The names of the Agro-Ecological Zones are available in Figure S4.

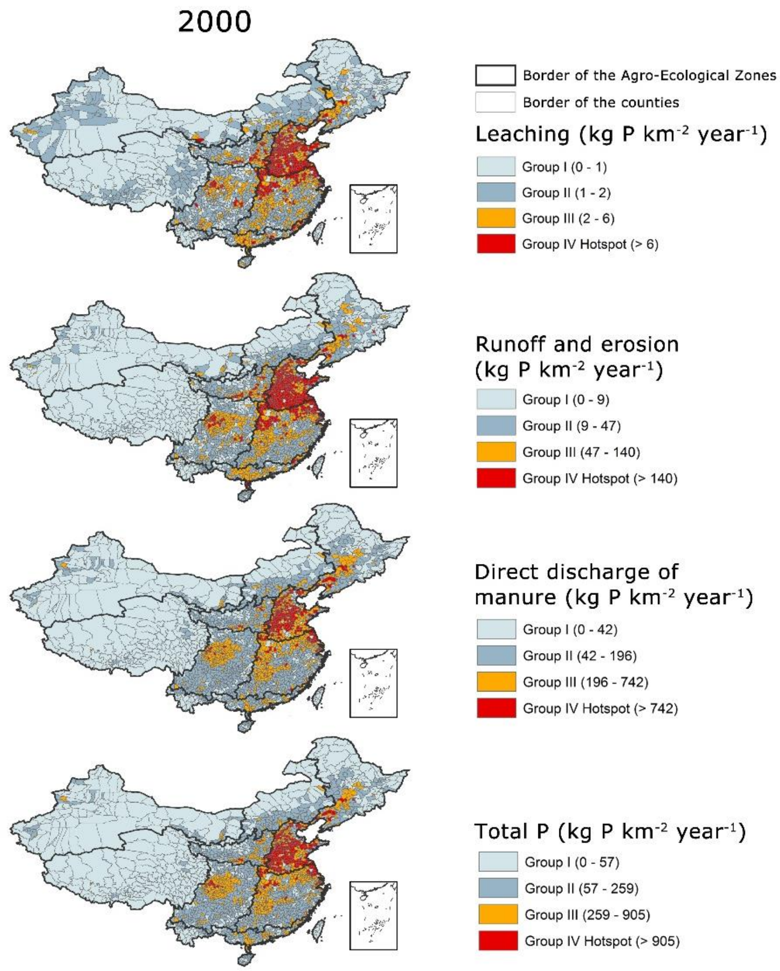

Figure S3 Phosphorus (P) losses (kg P km^{-2} $year^{-1}$) to waters from leaching, runoff and erosion, direct discharge of manure, and the total P losses (kg P km^{-2} $year^{-1}$) from food production in 2000. The P losses were quantified using the *NUFER* (NUtrient flows in Food chains, Environment and Resources use) model. The intervals for the four groups in this figure were defined based on quantiles (25%, 50%, 75%) of P losses of all counties in 2012: group I (0-25%), group II (25-50%), group III (50-75%), group IV (75-100%). Counties in group IV were qualified as hotspots. The names of the Agro-Ecological Zones are available in Figure S4.

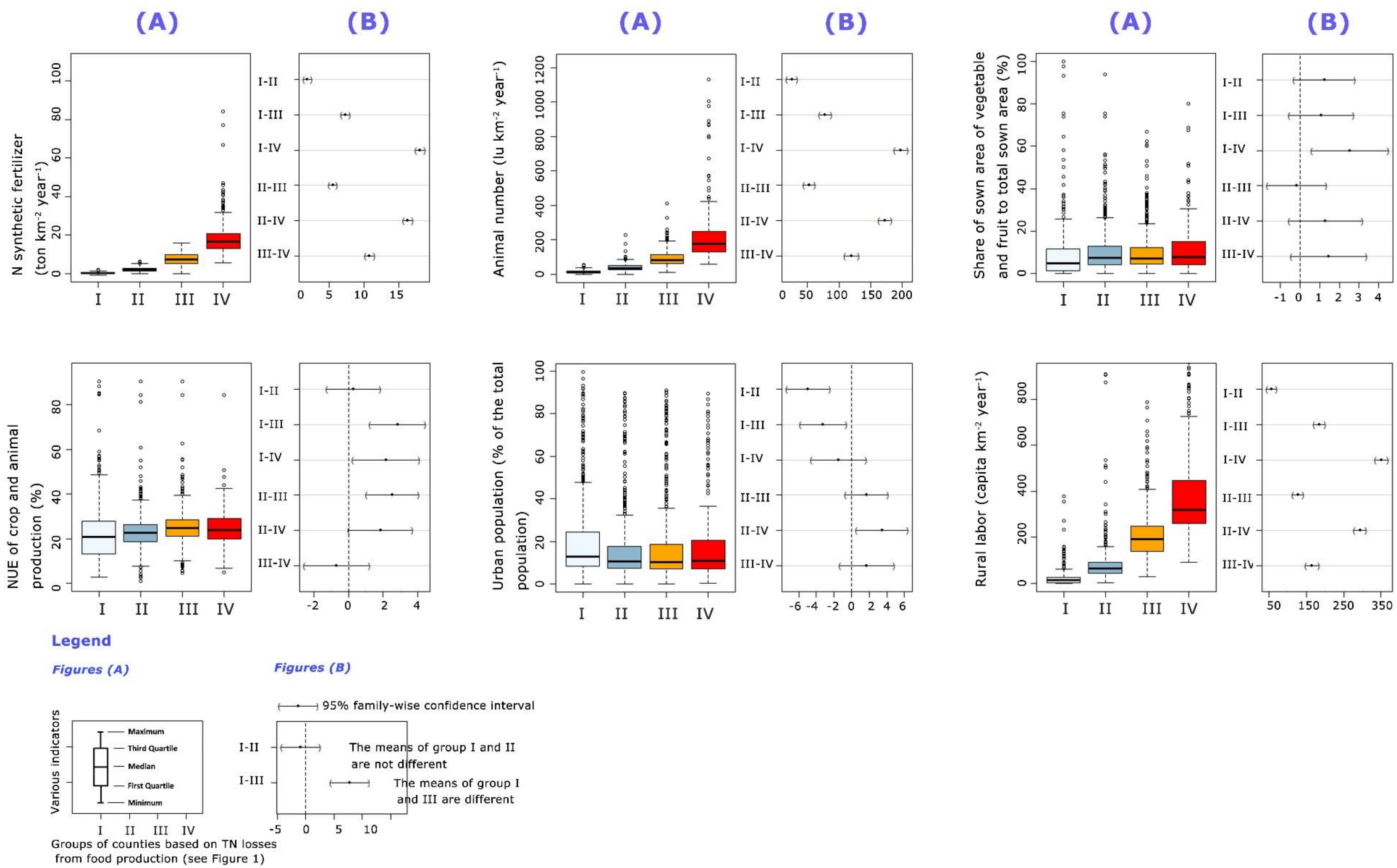

Figure S4 Boxplots for nitrogen (N): synthetic fertilizer (ton km^{-2} year^{-1}), animal number in livestock unit (lu km^{-2} year^{-1}, see Supporting Information for converting animal numbers in livestock unit), share of sown area of vegetable and fruit to the total sown area (%), N use efficiency (NUE) of food production, urban population (% of the total population), rural labor (capita km^{-2} year^{-1}) among the four groups of total N losses (see Figure 1) in 1990 (A), and the pair-wise comparisons from Tukey's Honest

Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

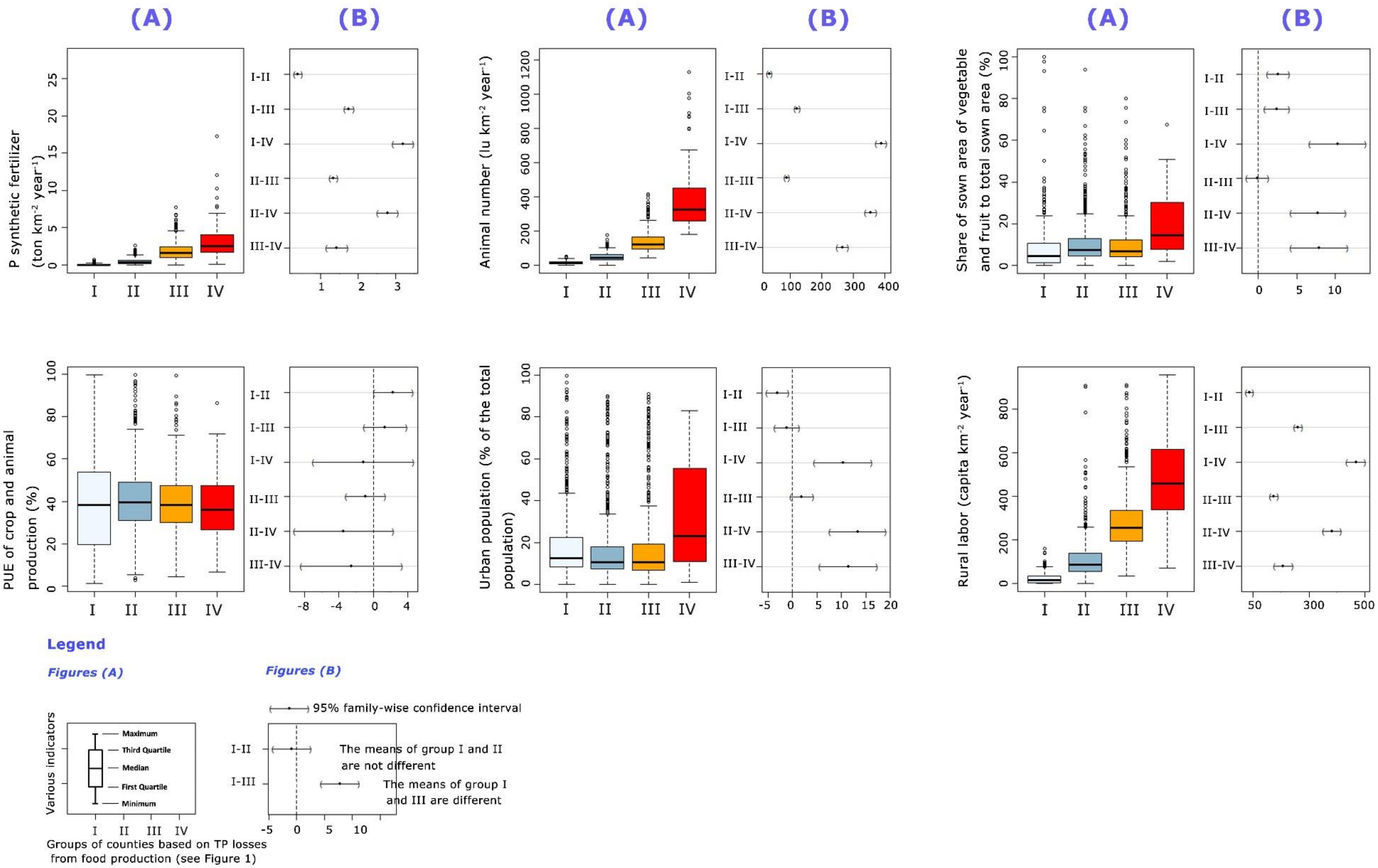

Figure S5 Boxplots of for phosphorus (P): synthetic fertilizer (ton km^{-2} year^{-1}), animal number in livestock unit (lu km^{-2} year^{-1}, see Supporting Information for converting animal numbers in livestock unit), share of sown area of vegetable and fruit to the total sown area (%), P use efficiency (PUE) of food production, urban population (% of the total population), rural labor (capita km^{-2} year^{-1}) among the four groups of total P losses (see Figure 1) in 1990 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

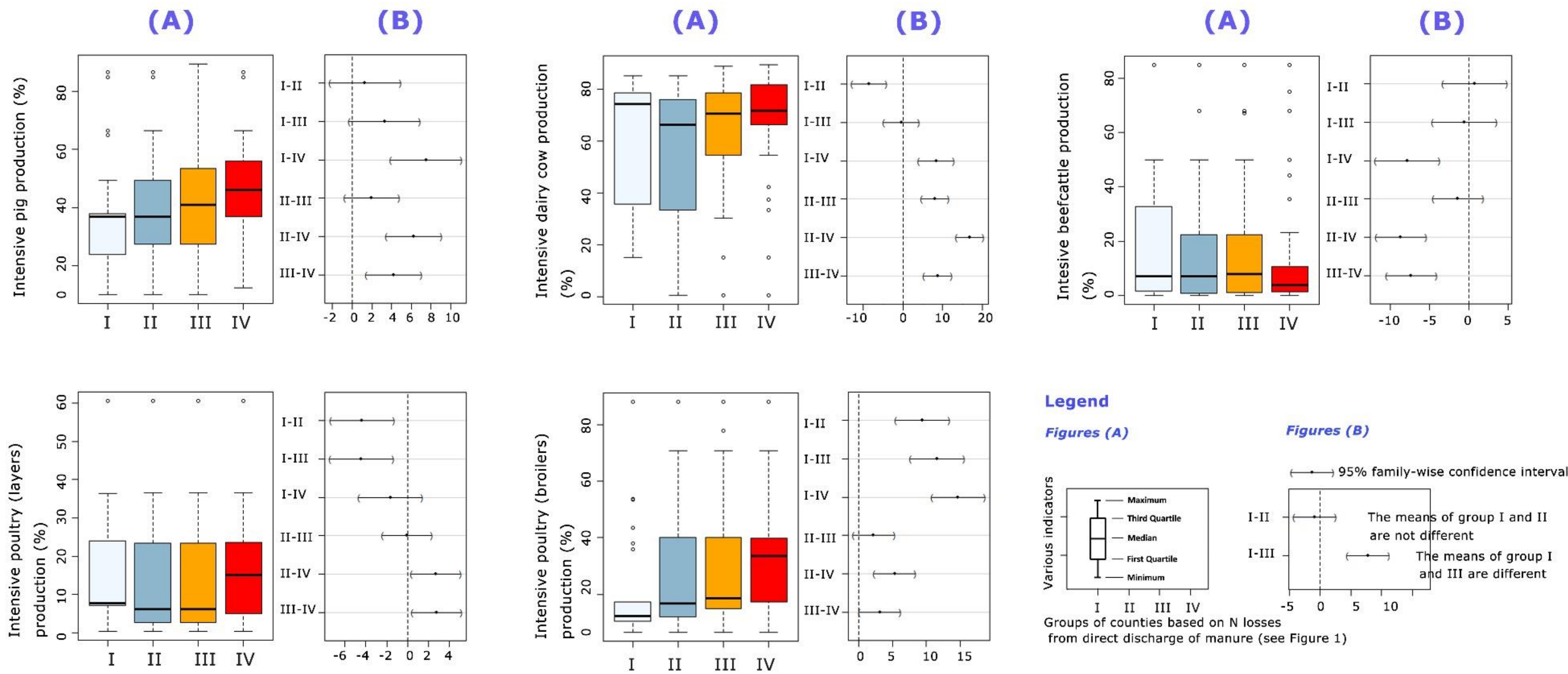

Figure S6 Boxplots for nitrogen (N): intensive pig production (> 50 head pig per farm) (%), intensive dairy cow production (> 5 dairy cow per farm) (%), intensive beefcattle production (> 50 head beefcattle per farm) (%), intensive poultry (layers) production (> 500 layers per farm) (%), and intensive poultry (broilers) production (> 2000 broilers per farm) (%) among the four groups of N losses from direct discharge of manure to waters (see Figure 1) in 2012 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

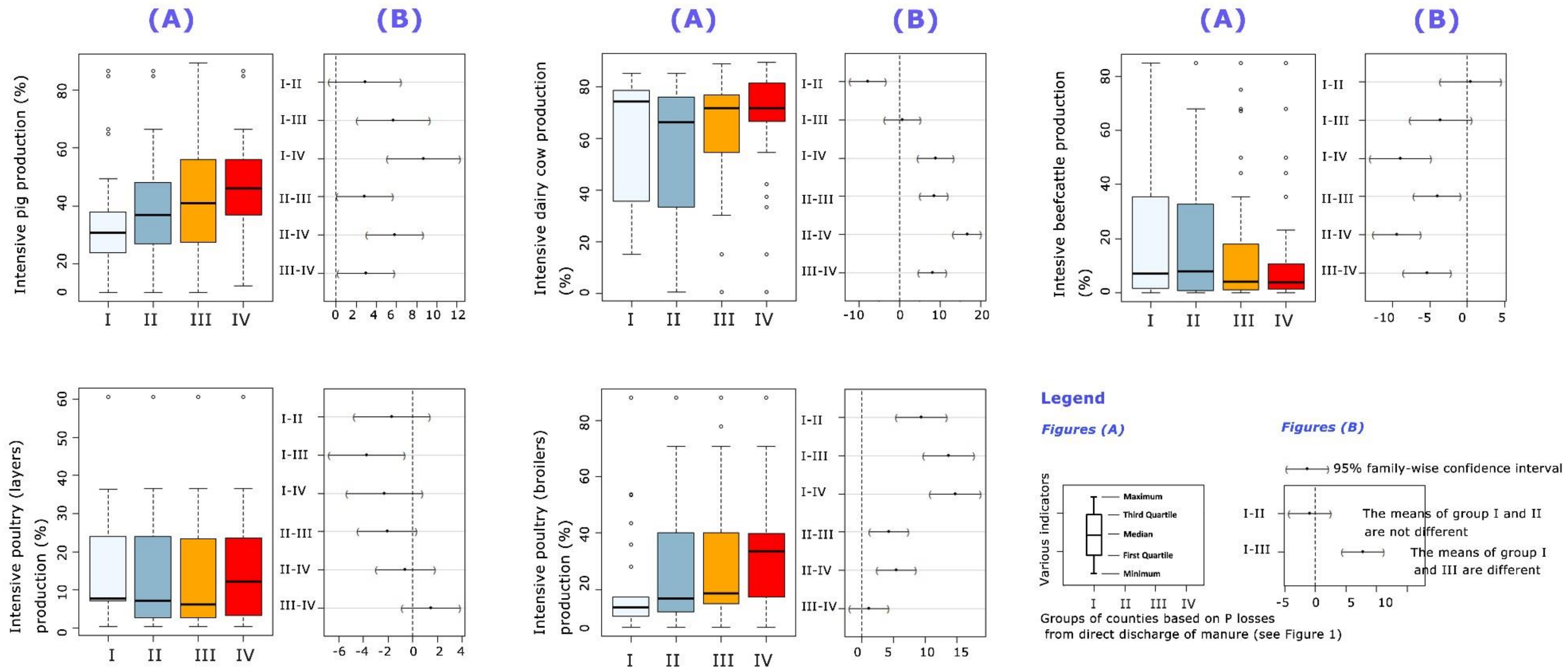

Figure S7 Boxplots for phosphorus (P): intensive pig production (> 50 head pig per farm) (%), intensive dairy cow production (> 5 dairy cow per farm) (%), intensive beefcattle production (> 50 head beefcattle per farm) (%), intensive poultry (layers) production (> 500 layers per farm) (%), and intensive poultry (broilers) production (> 2000 broilers per farm) (%) among the four groups of P losses from direct discharge of manure to waters (see Figure 1) in 2012 (A), and the pair-wise comparisons from Tukey's Honest Significant Difference (Tukey's HSD) among the four groups (B). In figures (B) any 95% confidence intervals that do not contain 0 provide evidence of a difference in the groups.

Figure S8 Nine Agro-Ecological Zones (AEZs) in China. The AEZs are: (I) Northeast China, (II) Inner Mongolia and Great Wall Vicinity, (III) North China Plain, (IV) Loess Plateau, (V) Middle and Lower Yangtze River, (VI) Southwest China, (VII) South China, (VIII) Gansu and Xinjiang, and (IX) Tibetan Plateau. Source: Sun and Shen [29].

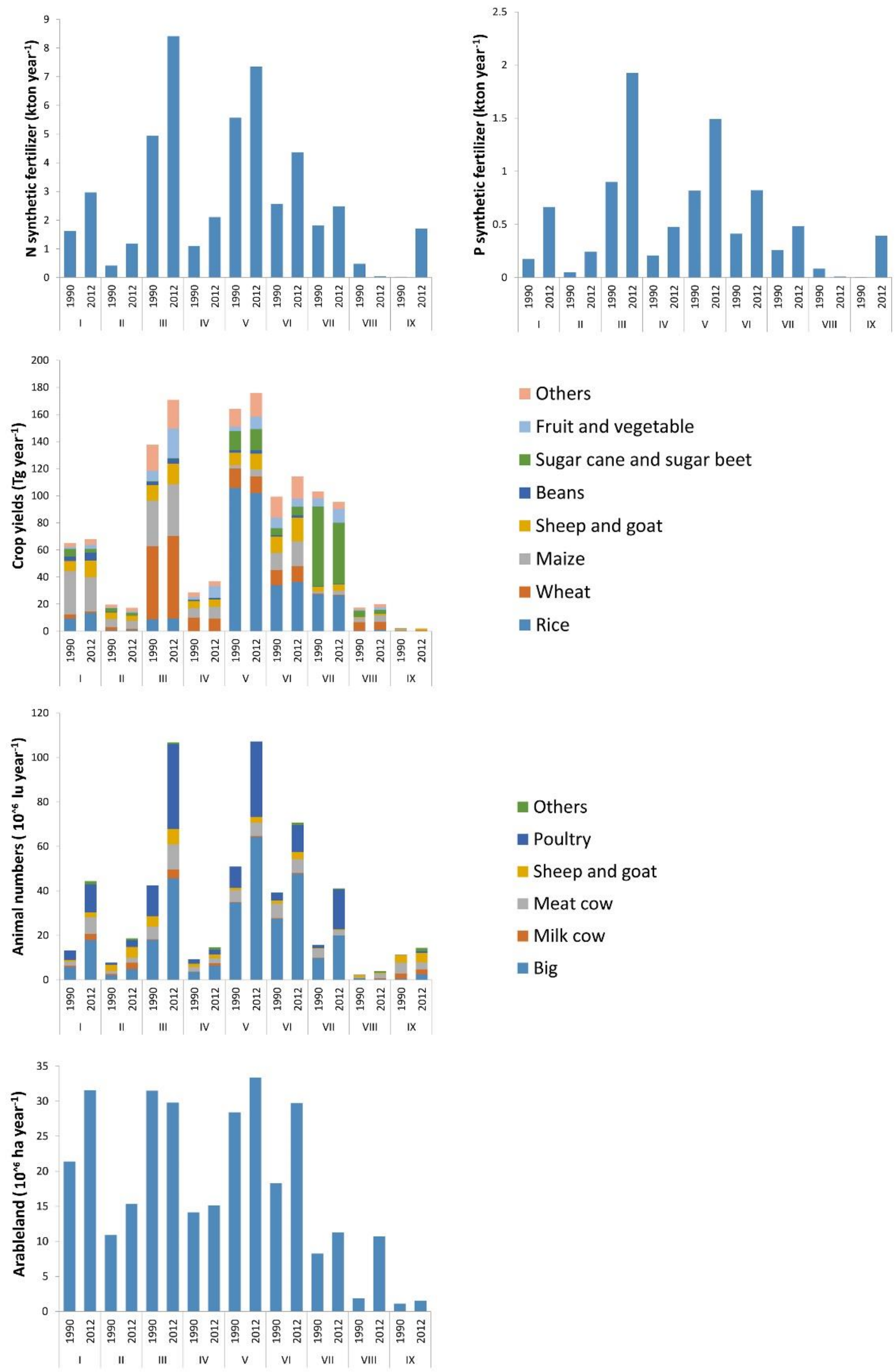

Figure S9 The nitrogen (N) fertilizer use, phosphorus (P) fertilizer use, crop yields, animal number, arable land for the nine Agro-Ecological Zones (AEZs) in 1990 and 2012. The nine AEZs include: (I) Northeast China, (II) Inner Mongolia and Great Wall Vicinity, (III) Huang Huai Hai, (IV) Loess Plateau, (V) Middle and Lower Yangtze River, (VI) Southwest China,

(VII) South China, (VIII) Gansu and Xinjiang, and (IX) Tibetan Plateau. The location of the AEZs are available in Figure S8.

Table S6 Losses in hotspots (% of the total losses in China) for N from leaching, runoff and erosion, direct discharge of animal manure, ammonia (NH_3) and nitrous oxide (N_2O) emission, and for P from leaching, runoff and erosion, and direct discharge of animal manure, and for total N and total P losses from food production, and the associated hotspot area (% of the total area, and the hotspot area in km^2). The location of the hotspots are shown in Figures 1 and 2.

N or P losses	N or P losses in hotspots (% of the total N or P losses in China)			Hotspot area (% of the total area in China) (hotspot area in km^2)		
	1990	2000	2012	1990	2000	2012
N leaching	48%	55%	61%	6% (598,865)	7% (623,711)	9% (884,317)
N runoff and erosion	41%	47%	51%	6% (528,383)	6% (600,875)	9% (811,092)
N direct discharge of manure	6%	33%	57%	0.2% (17,365)	3% (322,739)	9% (883,934)
NH_3	35%	50%	52%	4% (418,325)	7% (652,632)	9% (821,457)
N_2O	35%	50%	52%	4% (410,459)	7% (637,675)	9% (822,338)
Total N	28%	45%	52%	3% (307,850)	6% (557,242)	9% (828,205)
P leaching	42%	67%	69%	3% (280,965)	6% (610,097)	10% (901,719)
P runoff and erosion	39%	55%	58%	4% (366,086)	6% (577,734)	9% (835,228)
P direct discharge of manure	9%	43%	64%	0.3% (26,954)	3% (322,892)	9% (878,681)
Total P	10%	45%	62%	0.4% (35,355)	4% (371,248)	9% (861,781)

Table S7 Mean synthetic fertilizer application, animal numbers, share of sown area of vegetable and fruit to the total sown area, N and P use efficiencies (NUE and PUE) of crop and animal production, urban population, total output value of agriculture and forestry, and farmer's income of all counties in group I, II and III, and of all counties in group IV for total nitrogen (TN) and total phosphorus (TP)losses in 2012. The groups of the counties are shown in Figures 1 and 2.

		TN losses		TP losses	
Mean of	Unit	Group I, II, III	Group IV	Group I, II,III	Group IV
N Synthetic fertilizer	ton km^{-2} $year^{-1}$	4	22	n.a.	n.a.
P Synthetic fertilizer	ton km^{-2} $year^{-1}$	n.a.	n.a.	1	4
Animal numbers	lu km^{-2} $year^{-1}$	59	272	54	283
Share of sown area of vegetable and fruit to the total sown area	%	23	21	22	22
NUE of crop and animal production	%	23	25	n.a.	n.a.
PUE of crop and animal production	%	n.a.	n.a.	30	25
Urban population	% of the total population	21	18	21	18
Rural labor	capita km^{-2} $year^{-1}$	94	294	95	290
Total output value of agriculture and forestry	million yuan km^{-2} $year^{-1}$	1	5	1	5
Farmer's income	1000 yuan $capita^{-1}$ $year^{-1}$	7	9	7	9

n.a. Not available

Table S8 Nitrogen (N) and Phosphorus (P) loss factors for leaching, surface runoff, erosion to waters, ammonia (NH_3) and nitrous oxide (N_2O) emissions to the air from crop production of China for 1990, 2000 and 2012. The range (minimum and maximum) of N and P loss factors for Chinese counties are shown in the brackets in the table. These factors are derived based on the study of Ma et al.[14].

Crop production	N loss factors from total N input to crop production %	P loss factors from total P input to crop production %
	National N loss factor (minimum - maximum)	National N loss factor (minimum - maximum)

	1990	2000	2012	1990	2000	2012
Leaching	5 (0 - 14)	5 (0 - 14)	5 (0 - 14)	0.2 (0 - 0.4)	0.2 (0 - 0.4)	0.2 (0 - 0.4)
Surface runoff	10 (10 - 10)	10 (10 - 10)	10 (10 - 10)	1.5 (1.5 - 1.5)	1.5 (1.5 - 1.5)	1.5 (1.5 - 1.5)
Erosion	0.3 (0.3 - 0.3)	0.3 (0.3 - 0.3)	0.3 (0.3 - 0.3)	3 (0 - 3)	3 (0 - 3)	3 (0 - 3)
NH_3	18 (0.4 - 25)	18 (0.4 - 25)	18 (0.4 - 25)	n.a.	n.a.	n.a.
N_2O	0.7 (0.05 - 1)	0.7 (0.05 - 1)	0.7 (0.05 - 1)	n.a.	n.a.	n.a.

n.a. Not available

Table S9 Nitrogen (N) and Phosphorus (P) loss factors for direct discharge of manure to waters, ammonia (NH_3) and nitrous oxide (N_2O) emissions to the air from animal manure of China for 1990, 2000 and 2012. The range (minimum and maximum) of N and P loss factors for Chinese counties are shown in the brackets in the table. These factors are derived based on the study of Ma et al.[14].

Animal production	N loss factors from total N in animal manure % National N loss factor (minimum - maximum)			P loss factors from total P in animal manure % National N loss factor (minimum - maximum)		
	1990	2000	2012	1990	2000	2012
Direct discharge of manure	15 (6 - 18)	25 (14 - 36)	35 (22 - 56)	24 (8 - 25)	39 (19 - 52)	54 (30 - 76)
NH_3	29 (23 - 41)	29 (22 - 32)	27 (19 - 37)	n.a.	n.a.	n.a.
N_2O	0.5 (0.5 - 0.5)	0.5 (0.5 - 0.5)	0.5 (0.5 - 0.5)	n.a.	n.a.	n.a.

n.a. Not available

References

1. Chadwick, D.; Wei, J.; Yan'an, T.; Guanghui, Y.; Qirong, S.; Qing, C., Improving manure nutrient management towards sustainable agricultural intensification in China. *Agriculture, Ecosystems & Environment* **2015,** *209*, 34-46.
2. Strokal, 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. *Environmental Research Letters* **2016,** *11* (2), 024014.
3. Wang, L., *Modern Chinese pig production*. Jindun Press: Beijing, 2007.
4. Schneider, M., Feeding China's pigs: implications for the environment, China's smallholder farmers and food security. **2011**.
5. Bai, Z. H.; Ma, L.; Oenema, O.; Chen, Q.; Zhang, F. S., Nitrogen and Phosphorus Use Efficiencies in Dairy Production in China. *J. Environ. Qual.* **2013,** *42* (4), 990-1001.
6. Bai, Z.; Ma, L.; Qin, W.; Chen, Q.; Oenema, O.; Zhang, F., Changes in Pig Production in China and Their Effects on Nitrogen and Phosphorus Use and Losses. *Environmental science & technology* **2014,** *48* (21), 12742-12749.
7. Ma, L.; Ma, W.; Velthof, G.; Wang, F.; Qin, W.; Zhang, F.; Oenema, O., Modeling nutrient flows in the food chain of China. *Journal of environmental quality* **2010,** *39* (4), 1279-1289.
8. RESDC http://www.resdc.cn (accessed 02-15).
9. RESDC http://www.resdc.cn (accessed 01-02).
10. RESDC http://www.resdc.cn (accessed 01-02).
11. MOA, The Chinese agricultural statistical report. (In Chinese). Ministry of Agriculture. China Agricultural Press: Beijing, China, 2011.
12. NBSC, China statistical yearbook. (In Chinese). National Bureau of Statistics of China. China Statistic Press: Beijing, China, 2011.
13. MOA, The Chinese agricultural statistical data in 2005. Ministry of Agriculture. China Agricultural Public House: Beijing, 2006.
14. Ma, L.; Velthof, G.; Wang, F.; Qin, W.; Zhang, W.; Liu, Z.; Zhang, Y.; Wei, J.; Lesschen, J.; Ma, W., Nitrogen and phosphorus use efficiencies and losses in the food chain in China at regional scales in 1980 and 2005. *Science of the Total Environment* **2012,** *434*, 51-61.
15. MOA, China livestock yearbook. (In Chinese). Ministry of Agriculture. China Agricultural Press: Beijing, China, 2013.
16. Eurostat. Agricultural Census 2010 - Main Results 2013. http://ec.europa.eu/eurostat/statistics-explained/index.php/Agricultural_census_2010_-_main_results (accessed 19-09-2015).
17. Chen, X. Y.; Mulder, J., Atmospheric deposition of nitrogen at five subtropical forested sites in South China. *Science of the Total Environment* **2007,** *378* (3), 317-330.
18. Lü, C.; Tian, H., Spatial and temporal patterns of nitrogen deposition in China: Synthesis of observational data. *Journal of Geophysical Research: Atmospheres (1984–2012)* **2007,** *112* (D22).
19. Xu, W.; Luo, X.; Pan, Y.; Zhang, L.; Tang, A.; Shen, J.; Zhang, Y.; Li, K.; Wu, Q.; Yang, D., Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. *Atmospheric Chemistry and Physics* **2015,** *15* (21), 12345-12360.
20. Liu, X.; Zhang, Y.; Han, W.; Tang, A.; Shen, J.; Cui, Z.; Vitousek, P.; Erisman, J. W.; Goulding, K.; Christie, P., Enhanced nitrogen deposition over China. *Nature* **2013,** *494* (7438), 459-462.

21. Hou, Y.; Ma, L.; Gao, Z.; Wang, F.; Sims, J.; Ma, W.; Zhang, F., The driving forces for nitrogen and phosphorus flows in the food chain of China, 1980 to 2010. *Journal of environmental quality* **2013,** *42* (4), 962-971.
22. Wang, M.; Ma, L.; Strokal, M.; Chu, Y.; Kroeze, C., Exploring nutrient management options to increase nitrogen and phosphorus use efficiencies in food production of China. *Agricultural Systems* **2018,** *163*, 58-72.
23. 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. *Nutrient cycling in agroecosystems* **2016,** *104* (3), 361-372.
24. Wang, M.; Kroeze, C.; Strokal, M.; Ma, L., Reactive nitrogen losses from China's food system for the shared socioeconomic pathways (SSPs). *Science of The Total Environment* **2017,** *605–606*, 884-893.
25. Liu, X.; Sheng, H.; Jiang, S.; Yuan, Z.; Zhang, C.; Elser, J. J., Intensification of phosphorus cycling in China since the 1600s. *Proceedings of the National Academy of Sciences* **2016,** *113* (10), 2609-2614.
26. Huang, J.; Huang, Z.; Jia, X.; Hu, R.; Xiang, C., Long-term reduction of nitrogen fertilizer use through knowledge training in rice production in China. *Agricultural Systems* **2015,** *135*, 105-111.
27. Zhang, X.; Wu, Y.; Liu, X.; Reis, S.; Jin, J.; Dragosits, U.; Van Damme, M.; Clarisse, L.; Whitburn, S.; Coheur, P.-F., Ammonia emissions may be substantially underestimated in China. *Environmental Science & Technology* **2017**.
28. Gu, B. J.; Ge, Y.; Ren, Y.; Xu, B.; Luo, W. D.; Jiang, H.; Gu, B. H.; Chang, J., Atmospheric Reactive Nitrogen in China: Sources, Recent Trends, and Damage Costs. *Environmental Science & Technology* **2012,** *46* (17), 9420-9427.
29. Sun, H.; Shen, Y., *Agricultural natural resources and regional development in China*. Jiangsu science and technology press: 1994.