Evaluating Nationwide Non-Point Source Pollution of Crop Farming and Related Environmental Risk in China

Yang Duan [1,2], Hongqiang Jiang [1,2], Xiao Huang [3], Wenhui Zhu [4], Jie Zhang [5,*], Bo Wang [6] and Wenjun Wu [1,2,*]

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Section S1. Detailed calculation of the dissolved and adsorbed NPS pollutants components

1. Natural correction factor nf

nf represents the natural correction factor which is in connection with the slope, vegetation coverage and soil texture, and the equations are shown below(Wang *et al.*, 2010):

$$nf = G_{cf} \times V_{cf} \times S_{cf} \tag{S1}$$

$$G_{cf} = \frac{G - G_{\min}}{G_{\max} - G_{\min}} \tag{S2}$$

$$V_{cf} = \frac{V - V_{\min}}{V_{\max} - V_{\min}} \tag{S3}$$

$$S_{cf} = \frac{S - S_{\min}}{S_{\max} - S_{\min}} \tag{S4}$$

Where G_{cf}, V_{cf} and S_{cf} are slope, vegetation and soil texture correction factor. G, V and S stand for slope, vegetation cover and clay content of the grid. G_{max} and G_{min} are the maximum and minimum slope in the study area. V_{max} and V_{min} represent the maximum and minimum vegetation cover in the study area. S_{max} and S_{min} represent the maximum and minimum clay content in the study area.

2. Soil erosion amount A

The soil erosion amount A is calculated based on the universal soil loss equation (USLE) by wischmeier(Wischmeier, 1977) as follows:

$$A = R \times K \times L \times S \times C \times P \tag{S5}$$

where R is the average annual rainfall erosivity factor in MJ mm/(ha h) which are calculated by the storm energy(E) and peak 30-min rainfall intensity(I_{30})(Zhang *et al.*, 2016); K is the soil erodibility factor（t•hm^2•MJ•mm）; the variables L and S are the slope-length factors, respectively is the cover-management factor, and P is the supporting practice factor.

$$R = \sum_{i=1}^{12} \left\{ 1.735 \times 10^{(1.5 \times \lg \frac{P_i^2}{P} - 0.8188)} \right\} \tag{S6}$$

where P is the average annual rainfall(mm);P_i is the monthly average rainfall(mm).K is calculated as the following equation(Sharpley and Williams, 1990):

$$\begin{aligned} K = & \left\{ 0.2 + 0.3 \exp\left[-0.0256 SAN (1 - SIL / 100) \right] \right\} \left(\frac{SIL}{CLA + SIL} \right)^{0.3} \\ & \left(1 - \frac{0.25C}{C + \exp(3.72 - 2.95C)}\right)\left(1 - \frac{0.75SNI}{SNI + \exp(-5.51 + 22.9SNI)}\right) \end{aligned} \tag{S7}$$

$$SNI = (1 - SAN) / 100 \tag{S8}$$

where SAN is the sand content(2-0.05mm), SIL is the silt content (0.05-0.002mm), CLA is the clay content, C is the soil organic carbon content (%). The slope factor L and steepness factor S can be formulated according to the Eq. (S13) and Eq. (S14):

$$L = \left(\frac{\lambda}{22.13} \right)^m \tag{S9}$$

$$m = \begin{cases} 0.2 & \theta < 1^\circ \\ 0.3 & 1^\circ \le \theta < 3^\circ \\ 0.4 & 3^\circ \le \theta < 5^\circ \\ 0.5 & \theta > 5^\circ \end{cases} \tag{S10}$$

$$S = \begin{cases} 10.8 \sin\theta + 0.03 & \theta < 5^\circ \\ 16.8 \sin\theta - 0.05 & 5^\circ \le \theta < 10^\circ \\ 21.91 \sin\theta - 0.96 & 10^\circ \le \theta < 29^\circ \\ 9.5988 & \theta > 29^\circ \end{cases} \tag{S11}$$

where θ is the slope gradient, and the factor C can be calculated as follows (Cai CF, 2000):

$$C = \begin{cases} 1 & c = 0 \\ 0.6508 - 0.3436 \lg c & 0 < c \le 78.3\% \\ 0 & c > 78.3\% \end{cases} \tag{S12}$$

Where c is the average annual vegetation coverage. The value of P for different land-use type are set to different value depending on the land-use type(Mei, 2014).

Section S2. Detailed calculation the inputs in the N/P balance model

1. Mineral fertilizer

The amount of nutrient from the chemical fertilizer is sum of the N fertilizer (P fertilizer) and the nutrient in the compound fertilizer. This can be expressed as:

$$Ftlz_N = (Pftlz + Pftlz_{syn} \times fcnt_n) \times ratio_{N-ag} \tag{S13}$$

$$Ftlz_P = (Pftlz \times 0.4366 + Pftlz_{syn} \times fcnt_p) \times ratio_{P-ag} \tag{S14}$$

Where $Ftlz_N$ and $Ftlz_P$ is the input of N and P via the mineral fertilizer, $Pftlz$ is the consumption of the N fertilizer and P fertilizer, 0.4366 is the conversion factor from P_2O_5 to P, $Pftlz_{syn}$ is the consumption of the compound fertilizer, $fcnt$ is the N and P content in the compound fertilizer with a different value in six regions of China (Li and Jin, 2011)(Table S1), $ratio_{N-ag}$ and $ratio_{P-ag}$ are the proportion of the mineral fertilizer consumption used for crop with a value of 94% in N and 92.6% in P(Liu, 2005).

2. Excrements of animal

Professional and large-scale livestock and poultry farming has gradually become the main production method of livestock and poultry products. The resulting large amount of excrements returning to field is the primary source of nutrients in farmland. A method considering the breeding stage, raised period for different livestock is used to calculate the nutrient input in manure with a livestock category of pig, cattle, large domestic animal (horse, mule and donkey), sheep and goat, rabbit, poultry and so on. And each of these categories is divided into the some stages according to their different excretion coefficients. This can be expressed as:

$$N_{im} = \sum_{i=1}^{n} \frac{Q_i \times T_i \times q_i \times r}{365 \times 1000} \tag{S15}$$

$$P_{im} = \sum_{i=1}^{n} \frac{Q_i \times T_i \times q_i \times r}{365 \times 1000} \tag{S16}$$

Where N_{im} and P_{im} is nutrient input of farmland from the excrements, Q_i is the feeding amount, T_i is the raising period, q_i is the excretion coefficient, r is the ratio of excrement returning to the field with the value of 47%、57%、42%、53% and 51% for pig, beef, horse, sheep, chicken manure, respectively. These coefficients listed in Table S2 are based on the previous studies(Wang *et al.*, 2014)

3. Straw manure input

Crop straw, as a biomass by-product of agricultural production, has huge potential utilization value. The recycled straw can increase soil nutrient content, promote soil carbon sequestration and improve soil structure. The computing method for straw manure input can be expressed as：

$$N_{is} = \sum_{i=1}^{n} g_i \times p_i \times a_{ni} \times r_i \times 0.85 \quad \text{(S17)}$$

$$P_{is} = \sum_{i=1}^{n} g_i \times p_i \times a_{pi} \times r_i \times 0.85 \quad \text{(S18)}$$

Where g_i is the crop yield, p_i is the ratio of grain to straw, a_{ni} and a_{pi} are the concentration of N and P in straw, r_i is the ratio of straw returning to the field, 0.85 is the conversion factor after removing moisture from straw. The corresponding parameters are listed in the Table S3 and S4(ZHANG *et al.*, 2017).

4. Cake fertilizer input

The cake fertilizer input is estimated by the rate of cake, rate of crop to oil, N/P in cake and the ratio of cake returning to the field based on the data from “Nation Agro-Tech Extension and Service Center” (Table S5).

$$N_{ic} = \sum_{i=1}^{n} g_i \times o_i \times c_i \times a_{ni} \times r \quad \text{(S19)}$$

$$P_{ic} = \sum_{i=1}^{n} g_i \times o_i \times c_i \times a_{pi} \times r \quad \text{(S20)}$$

Where g_i is the crop yield, o_i is the rate of crop to oil, c_i is the rate of cake, a_{ni} and a_{pi} are the N/P in cake, r is the ratio of cake returning to the field.

5. Atmospheric deposition

Atmospheric deposition includes wet and dry deposition. Wet deposition mainly depends on the emission of ammoniacal N and fertilizing amount. With the rapid development of stock husbandry, fertilizer usage and fossil fuel burning, the amount of atmospheric N has increased rapidly in recent years. The result shows the N deposition input along with the rainfall is 8-30 $kg.ha^{-1}.yr^{-1}$ in northern China while the value is 16.5-34.95 kg•ha-1•yr-1 because of the plentiful precipitation(Zhang Min, 1992). The wet deposition flux of N for every province is based on published paper(Wang *et al.*, 2014). The dry deposition for N is mainly from the re-deposition of volatile ammonia and other N compounds (such as NO_2). The amount of dry deposition for N is calculated as follows:

$$Dpzt_d = (\mathrm{Ftlz}_n + \mathrm{N}_{im}) \times p_v \times coff_{dpzt} \times p_f \times 1.25 \quad \text{(S21)}$$

Where $Dpzt_d$ is the amount of dry deposition for N, $Ftlz_n$ and N_{im} are the N of fertilizer and manure, p_v is the loss rate of ammonia volatilization with a value of 24.5% for fertilizer and 30% for manure, $coff_{dpzt}$ is the deposition rate of ammonia with a default value of 15%, P_f is the proportion of cultivated land to total land area.

6. Seed

The method for seed input is expressed as follows:

$$N_{is} = \sum_{i=1}^{2} \sum_{j=1}^{9} area_{ij} \times sd_{ij} \times sow_{ij} \times 0.001 \quad \text{(S22)}$$

where N_{is} is the nutrient input(tons), $area_{ij}$ is the sowing area(hm^2), sd_{ij} is the seeding rate(kg•hm^{-2}), sow_{ij} is the nutrient content in seeds (%) and the specific values are showed in Table S6(Peng *et al.*, 2004).

7. Biological N fixation

Biological N fixation contains symbiotic and non-symbiotic N fixation. We assume 2/3 of the N absorbed by leguminous plants is from the symbiotic N fixation. The value of N uptake per crop production is 72 kg•t^{-1} and 68 kg•t^{-1} for soybean and peanut(R.K, 1998). The non-symbiotic N fixation rate in paddy and upland fields is set as 30 kg•hm^{-2}•yr^{-1} and 15 kg•hm^{-2}•yr^{-1}(Liu, 2005).

$$Bnf = Bnf_{sym} + Bnf_{non} \quad \text{(S23)}$$

$$Bnf_{sym} = \sum_{i=1}^{2} soybean_i \times scnt_i \times 0.6 \times 0.001 \quad \text{(S24)}$$

$$Bnf_{non} = \sum (area_p \times nf_p + area_d \times nf_d) \times 0.001 \quad \text{(S25)}$$

Where Bnf is the total biological N fixation, Bnf_{sym} is the symbiotic N fixation and Bnf_{non} is the non-symbiotic N fixation. $soybean_i$ is the yield of leguminous plants (i=1 soybean, i=2 peanut), $scnt_i$ is the value of N uptake per crop production. $area_p$ and $area_d$ are the area of paddy and upland fields, respectively, nf_p and nf_d are non-symbiotic N fixation rate in paddy and upland fields, respectively.

8. Irrigation input

The nutrient from irrigation depends on the content in the water and the irrigation amount. The value in northern China is smaller than in southern China, and in this paper the value in northern China is set as 4.7 kg •hm^{-2}•yr^{-1} and 6.0 kg •hm^{-2}•yr^{-1} in southern China.

Section S3. Detailed calculation the outputs in the N/P balance model

1.Crop uptake

The crop uptake is the main output item of farmland nutrient balance. In this research we choose 13 kinds of stable crops in China with a certain uptake coefficient via reviewing the related literatures in Table S7(Li and Jin, 2011).

2.Gaseous nitrogen

There is a loss of gaseous nitrogen during the transformation of inorganic nitrogen including ammonia volatilization and denitrification. The factors affecting the rate of denitrification and volatilization mainly cover fertilization frequency, fertilizer type, crop type, and farming operation(Chen *et al.*, 2008). The amount of ammonia volatilization and denitrification outputs can be calculated by the following equations.

$$Gas_{NH_3} = Ftlz \times \frac{(fvnt_p \times area_p + fvnt_d \times area_d)}{fvnt_p + fvnt_d} + N_{im} \times fvnt_a \qquad \text{(S26)}$$

$$Gas_{NOx} = Ftlz \times \frac{(fdnt_p \times area_p + fdnt_d \times area_d)}{fdnt_p + fdnt_d} + N_{im} \times fdnt_a \qquad \text{(15)}$$

Where the Gas_{NH_3} and Gas_{NOx} are the ammonia volatilization and denitrification outputs. $Ftlz$ is the input of N via the mineral fertilizer. $fvnt_p$ and $fdnt_p$ are the volatilization and denitrification loss rate for paddy with the value of 11% and 20%, respectively. $fvnt_d$ and $fdnt_d$ are the volatilization and denitrification loss rate for upland with the value of 10% and 14%, respectively. $area_p$ and $area_d$ are the area of the paddy and upland. N_{im} is the N of farmland from the excrements. $fvnt_a$ and $fdnt_a$ are the volatilization and denitrification loss rate for upland with the manure of 13%.

3. Runoff and Leaching

The nutrient output from runoff and leaching are calculated by using empirical coefficient method and the runoff and leaching coefficient are from the output in the first national pollution census (Table S8). For lack of the correlation parameter for the phosphorus runoff and leaching, 20% of the phosphorus fertilizer input is considered as the amount of runoff and leaching based on previous study(Li and Jin, 2011).

Section S4. Details data source and data-pre-processing

The study area contained the entire land area of mainland China, including 31 provinces, autonomous regions and municipalities, while Hong Kong, Macao and Taiwan and the South China Sea Islands were out of study area due to limitation regarding data available from these regions. Input data in this study complied as two categories: statistical data and spatial data. The statistical data, such as consumption of chemical fertilizers, crops sown area, crop yield, amount of livestock and poultry, rural population, the effective irrigated area, was obtained from the China Agricultural Yearbook, China Rural Statistical Yearbook, as well as provincial and municipal statistical yearbooks in 2015. More than 600,000 data items from 341 municipal administrative units were entered into database in the calculation. The spatial data, such as the land cover data, soil attribute data, vegetation cover data, were obtained from various public sources. Details about the input data are found in Table S1.

In this study, data pre-progressing contained the missing data's filling-up and interpolation of spatial data in GIS. Because of the complicated index system with more than 60 fields, much data was not counted in county-level. The scarce data were generated by checking out other relevant yearbooks or statistical data, or using the data in adjacent years. Considering the small proportion of general planting area in municipal districts, it was necessary to combine them, and a total of 2,151 statistical units were saved after the combination. Then the GIS software was used to superimpose the county-level data results with the boundary of the administrative region for spatial processing.

Supplementary Figures

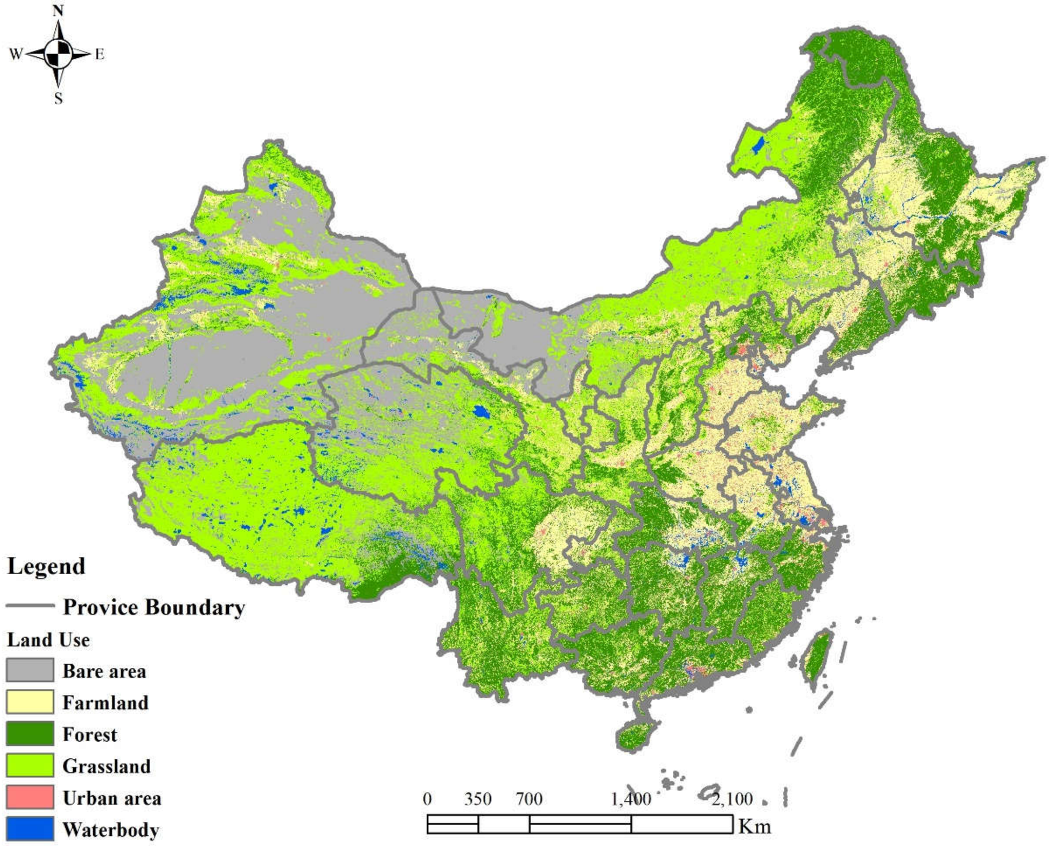


Figure S1. The distribution of land use in China in 2015(100m resolution)

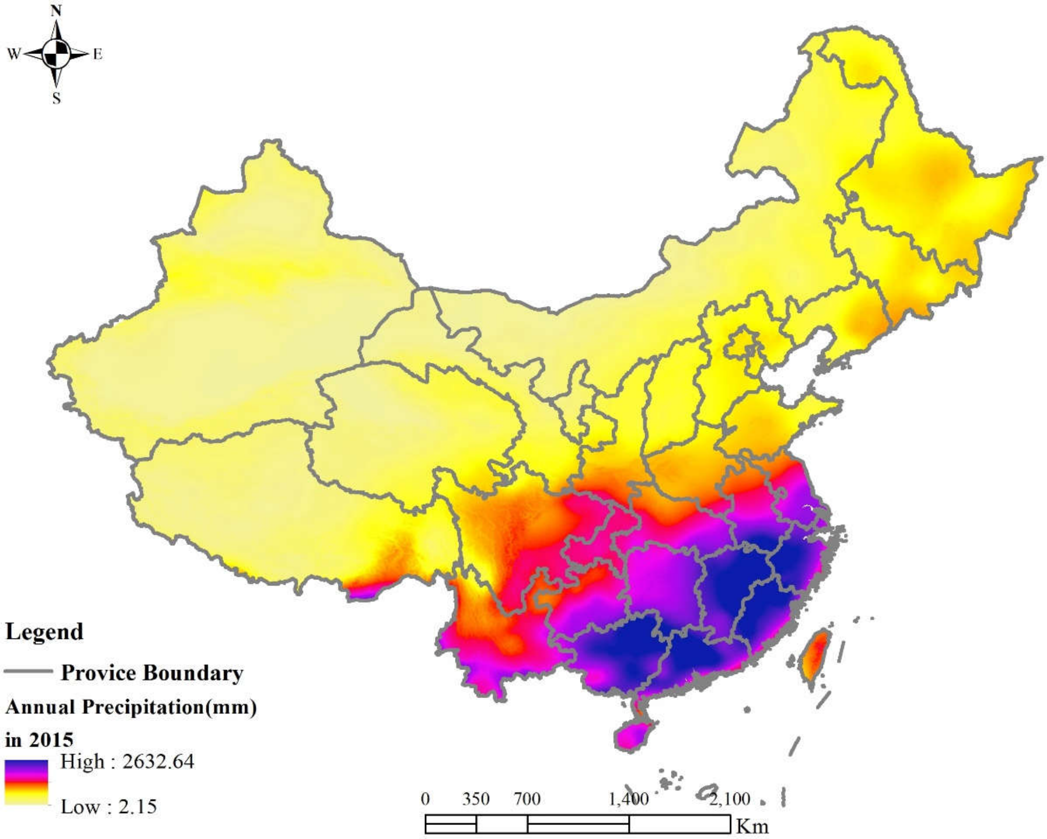
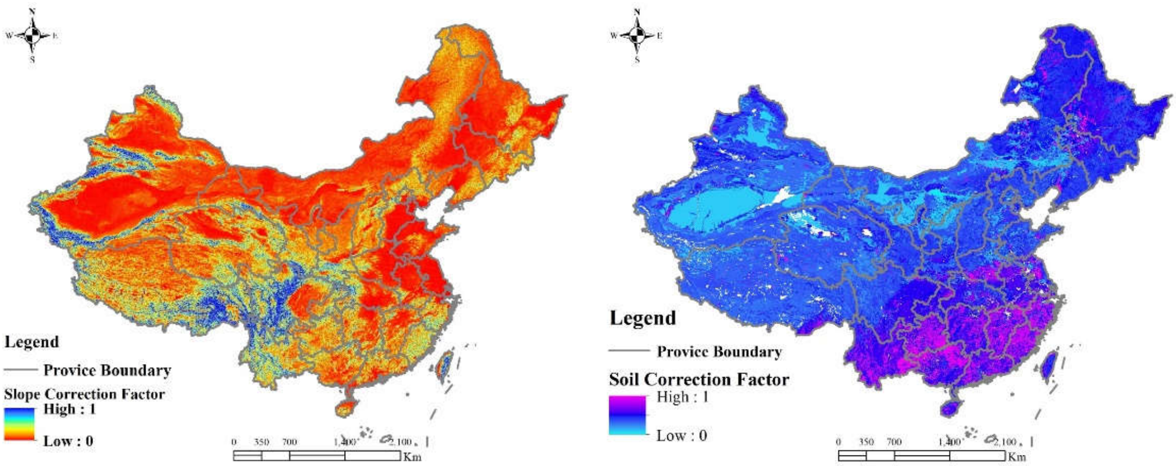
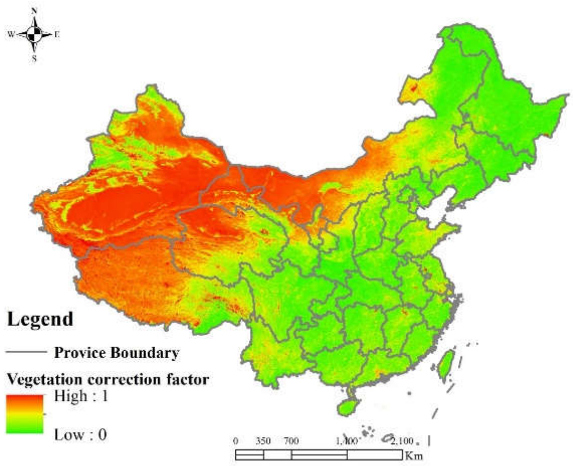


Figure S2. The Spatiotemporal distribution of annual precipitation in 2015

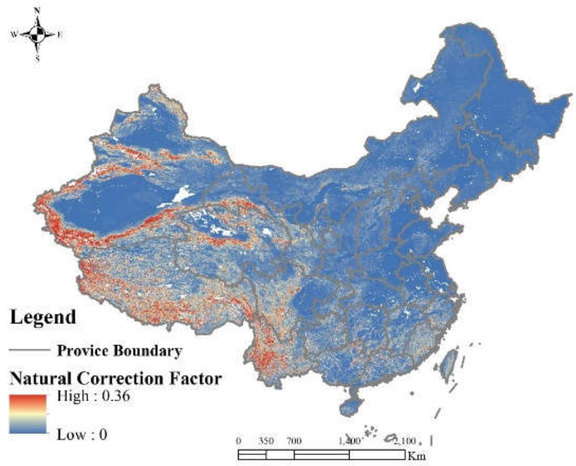


(a)

(b)

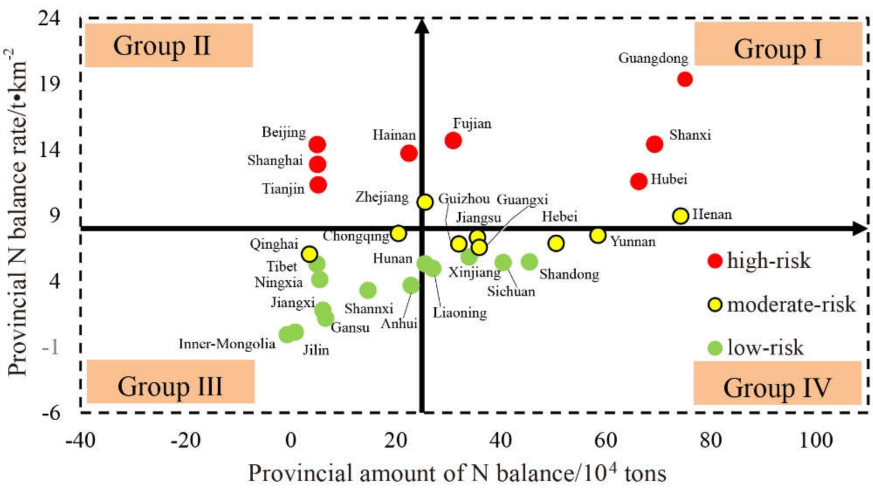


(c)

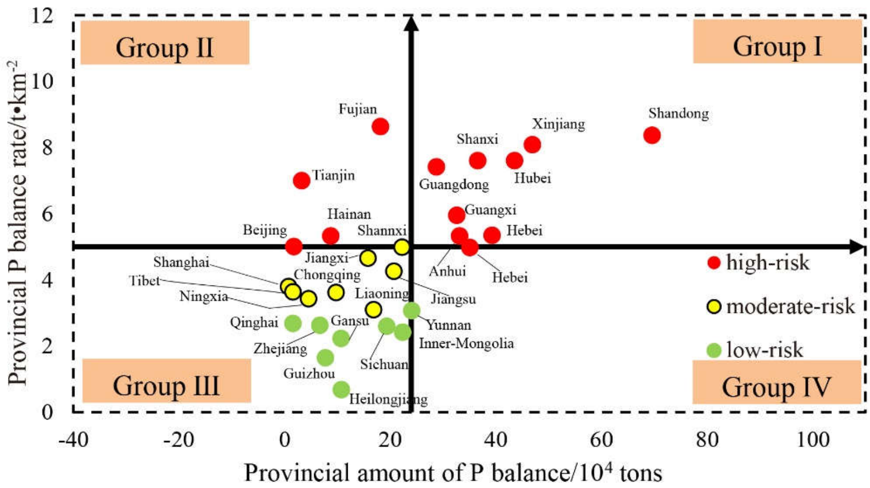


(d)

Figure S3. The distribution of slope correction factor, soil correction factor, vegetation correction factor and natural correction factor in China. (a) slope correction factor; (b) soil correction factor; (c) vegetation correction factor; (d) natural correction factor;



(a)



(b)

Figure S4. Classification of 31 provinces on the basis of amount of nutrient balance and nutrient balance rate.

Group Ⅰ located in the upper right-hand quadrant shows the provinces which have large nutrient surplus such as Guangdong, Hubei. Group II located in the upper left-hand quadrant shows provinces with a small nutrient balance amount but have a large balance rate because of its less arable land. Group III located in the lower left-hand quadrant shows that these provinces have a small nutrient balance either in total amount or in balance rate. Group IV, located in the lower right-hand quadrant shows the provinces have a large nutrient surplus and a small balance rate because of the large amount of the arable land.

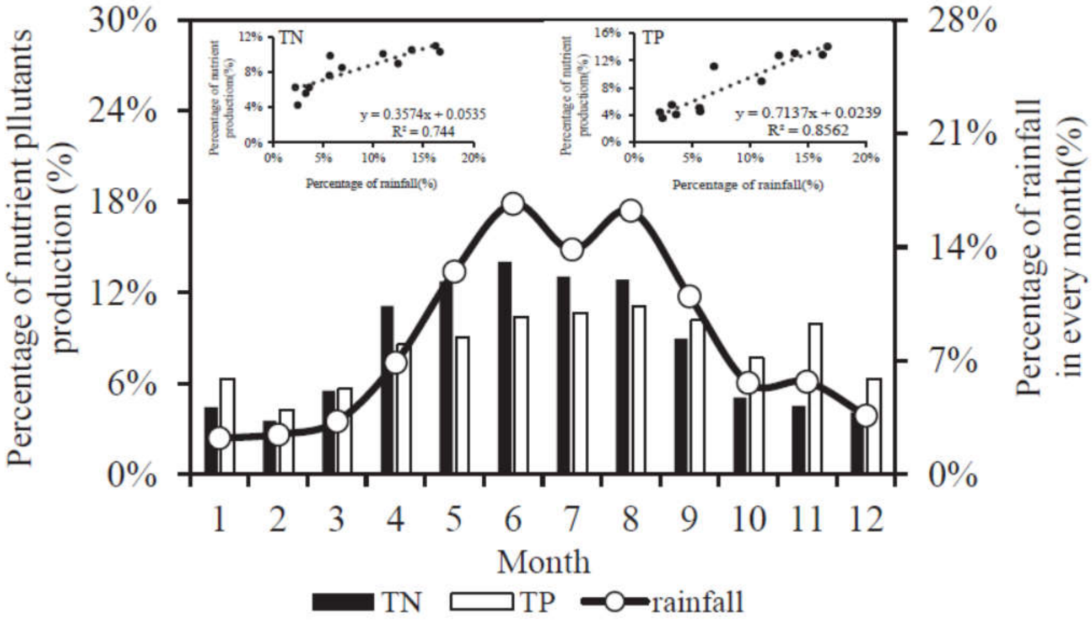


Figure S5 Temporal changes of dissolved pollutant in 2015

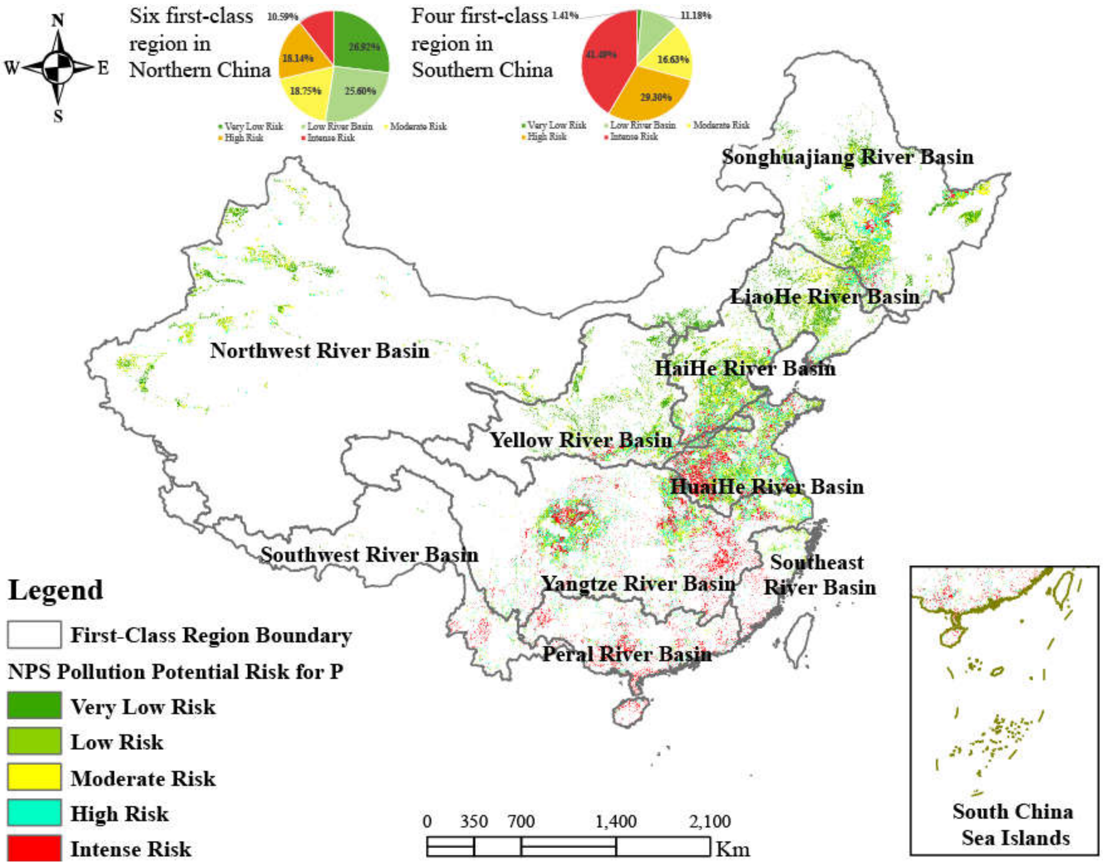


Figure S6 Potential pollution risk of NPS in 2015 (P pollution)

Supplementary Tables

Table S1 Description of data used in the study

Data type	Scale	Description	Data source
Land Cover	1:250 000	Land cover data in 2015	Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Science
Geographic	Grid:30m ×30m	Digital elevation model	Geospatial Data Cloud http://www.gscloud.cn
Soil map	1:1 000 000	Soil classifications, soil particle size (the content of sand, silt and clay) and chemical character (the content of total nitrogen, total phosphorus, and soil organic matter) of each type.	The soil Database of China for Land Surface Modeling from Land-Atmosphere Interaction Research Group at Sun Yat-sen University
Vegetation	1:1 000 000	Vegetation coverage rate in 2015	Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Science
Meteorological	National	Daily precipitation Rainfall density in 739 national reference climatological stations or basic synoptic stations in 2015.	National Meteorological Information Center
Rural and agricultural data	National	Rural population, livestock and poultry, fertilizing amount and crop yield and so on.	The statistical Yearbook 2015 in 299 cities of China

Table S2 The weighting factors of different indices for N/P pollution risk assessment

	SL	EP	DEP	DS	SI
Weighting	0.5	1	0.5	1	1.5

Table S3. Ratio of nutrient in compound fertilizer(Li and Jin, 2011)

Region	Province	Ratio of N, P, K in

		compound fertilizer
Northeast China	Liaoning, Jilin, Heilongjiang	1:2.0:0.2
North China	Beijing, Tianjin, Hebei, Shanxi, Shandong, Henan	1:1.5:0.4
Northwest China	Inner-Mongolia, Shaanxi, Gansu, Qinghai, Xinjiang, Ningxia	1:1.5:0.4
Central and Southern China region	Shanghai, Jiangsu, Zhejiang, Anhui, Hubei, Hunan, Jiangxi	1:1:0.8
Southwest China	Chongqing, Sichuan, Yunnan, Guizhou, Tibet	1:1:0.8
South China	Guangdong, Guangxi, Hainan, Fujian	1:1:0.8

Table S4. The N/P excretion coefficients for different livestock categories in China(Wang *et al.*, 2014)

			Northeast China	North China	Eastern China	South China	Southwest China	Northwest China
Wine	Weaners	N	26.03	20.4	11.35	19.83	10.97	21.49
		P	3.05	3.48	1.44	2.51	1.94	2.78
	Fattening pigs	N	57.7	33.23	25.4	44.73	19.74	36.77
		P	6.16	6.06	3.21	5.99	4.84	4.88
	Pregnant sows	N	78.67	43.66	39.6	51.13	22.02	40.79
		P	11.05	9.93	5.11	11.18	6.55	5.24
Beef Cattle	Adult	N	150.81	72.74	153.47	65.93	104.1	104.1
		P	17.06	13.69	19.85	10.52	10.17	10.17
	Young	N	61.91	20.33	53.55	30.59	38.27	38.27
		P	7	3.83	6.93	4.88	3.74	3.74
Horse, mule, and donkey	Adult	N	150.81	72.74	153.47	65.93	104.1	104.1
		P	17.06	13.69	19.85	10.52	10.17	10.17
	Young	N	61.91	20.33	53.55	30.59	38.27	38.27
		P	7	3.83	6.93	4.88	3.74	3.74
Sheep and goats	Adult	N	25.62	11.57	22.16	12.66	15.84	15.84
		P	2.9	2.18	2.87	2.20	1.55	1.55
	Young	N	40.76	18.41	35.26	20.14	25.19	25.19
		P	4.61	3.46	4.56	3.21	2.46	2.46
Rabbits		N	1.85	1.27	1.02	0.71	0.71	1.85
		P	0.48	0.3	0.5	0.06	0.06	0.46
Laying hens and		N	1.12	1.42	1.06	1.16	1.16	1.12

		Northeast China	North China	Eastern China	South China	Southwest China	Northwest China
broilers	P	0.23	0.42	0.51	0.23	0.23	0.23

Table S5. Parameter values for calculating the straw incorporation inputs(ZHANG *et al.*, 2017)

Crop category	Ratio of straw to grains	N content in straw (%)	P content in straw (%)
Rice	0.9	0.91	0.13
Corn	1.2	0.92	0.15
Wheat	1.1	0.65	0.08
Soybean	1.6	1.81	0.196
Tubers	0.5	2.5	0.28
Peanut	0.8	1.82	0.16
Rapeseed	2.5	0.87	0.14
Sunflower	2.2	0.82	0.111
Cotton	0.24	1.24	0.15
Sugarcane	1.1	1.1	0.14
Sugar beet	1	0.25	/

Table S6. Ratio of straw returning to the field in different provinces(ZHANG *et al.*, 2017)

Province	ri（%）	Province	ri（%）
Liaoning	11.2	Inner-Mongolia	15.0
Jilin	15.0	Shaanxi	33.2
Heilongjiang	10.2	Gansu	13.5
Beijing	88.6	Qinghai	0.8
Tianjin	29.1	Ningxia	15.0

Province	ri（%）	Province	ri（%）
Hebei	80.8	Xinjiang	15.0
Shanxi	55.5	Chongqing	58.0
Shandong	77.8	Sichuan	49.7
Henan	60.8	Guizhou	30.0
Shanghai	47.2	Yunnan	22.0
Jiangsu	26.8	Tibet	0.8
Zhenjiang	47.2	Fujian	26.8
Anhui	30.0	Guangdong	38.0
Hubei	28.7	Guangxi	45.3
Hunan	43.1	Hainan	29.1
Jiangxi	40.0		

Table S7 Parameters for estimating cake fertilizer input

	Rate of cake（%）	Rate of crop to oil（%）	N in cake（%）	P in cake（%）	ratio of cake returning to the field（%）
Soybeans	85	50	6.68	0.44	30
Peanut	50	43	6.92	0.55	30
Rapeseeds	95	55	5.25	0.79	90
Cotton	80	94	4.29	0.54	90
Sunflower	76.8	35	4.76	1.70	30

Table S8. Coefficients for estimating the seed input(Peng *et al.*, 2004)

Crop category	seeding rate($kg•hm^{-2}$)	N content in seeds (%)	P content in seeds (%)
Rice	112.5	1.40	0.01
Corn	30.0	1.60	0.22
Wheat	225.0	2.10	0.44
Soybean	67.5	5.30	0.47

Crop category	seeding rate($kg \cdot hm^{-2}$)	N content in seeds (%)	P content in seeds (%)
Tubers	1312.5	0.32	0.03
Peanut	300.0	4.40	0.11
Rapeseed	1.1	4.00	0.00
Cotton	52.5	3.00	0.24
Sunflower	30.0	2.61	0.3

Table S9. Coefficients for estimating the crop uptake output(Li and Jin, 2011)

	N in grain （kg/t）	P in grain （kg/t）	N in straw (%)	P in straw (%)
Rice	14.6	6.2	0.91	0.13
Corn	25.8	9.8	0.87	0.15
Wheat	24.6	8.5	0.65	0.08
Soybean	81.4	23.0	1.81	0.20
Tubers	4.45	1.0	2.50	0.28
Peanut	43.7	10.0	1.82	0.16
Rape	43.0	27.0	0.87	0.14
Sunflower	69.0	20.0	0.82	0.11
Cotton	12.6	4.6	1.24	0.15
Sugarcane	1.81	0.36	1.10	0.14
Sugar beet	4.8	1.42	/	/
Vegetables	4.32	1.42	/	/
Fruit	4.95	2.95	/	/

Table S10. Coefficients for estimating the runoff and leaching output(Wang *et al.*, 2014)

	N runoff coefficient ($kg\ ha^{-1}\ yr^{-1}$)			N leaching coefficient ($kg\ ha^{-1}\ yr^{-1}$)		
	Northern China	Northwest China	Southern China	Northern China	Northwest China	Southern China
Rice	6.41	0.05	16.31	7.29	7.29	7.29
Wheat	3.36	0.12	7.41	19.34	4.23	26.12

	N runoff coefficient (kg ha^{-1} yr^{-1})			N leaching coefficient (kg ha^{-1} yr^{-1})		
	Northern China	**Northwest China**	**Southern China**	**Northern China**	**Northwest China**	**Southern China**
Corn	2.88	0.12	7.95	16.02	4.23	34.86
Soybeans	2.64	0.12	13.55	18.75	3.17	34.86
Vegetables	8.60	7.98	18.18	48.98	30.22	28.50
Fruits	0.65	7.98	8.16	16.02	3.96	10.68

Table S11. The N inputs in the 31provinces of China in 2015 (10^4 tons)

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	dry deposition	Biological N fixation	Irrigation	Seed	Total
Liaoning	79.88	2.03	0.41	36.15	6.25	1.70	13.71	0.64	0.65	141.42
Jilin	106.02	4.95	0.56	36.83	7.08	2.09	15.33	0.56	0.63	174.05
Heilongjiang	105.96	6.18	3.71	37.28	29.01	1.95	46.83	2.15	1.79	234.85
Beijing	6.07	0.50	0.01	3.09	0.21	0.08	0.73	0.06	0.02	10.76
Tianjin	11.70	0.46	0.09	3.75	0.42	0.25	1.17	0.13	0.07	18.04
Hebei	175.69	23.99	1.98	41.04	5.96	3.26	18.30	1.71	1.94	273.86
Shanxi	50.48	5.99	0.25	10.99	4.20	0.67	10.41	0.67	0.56	84.23
Shandong	214.55	33.13	3.34	63.61	10.05	5.82	21.42	2.11	3.17	357.20
Henan	320.42	32.17	5.09	56.34	7.73	7.14	27.04	2.11	4.55	462.57
Shanghai	5.98	0.37	0.03	5.40	0.41	0.15	1.11	0.11	0.04	13.60
Jiangsu	184.70	7.37	3.53	15.02	9.01	3.55	18.18	2.27	1.64	245.28
Zhejiang	51.68	3.17	0.90	11.68	4.00	0.61	7.81	0.81	0.29	80.94
Anhui	156.54	9.39	5.30	22.83	12.30	3.02	25.53	2.08	2.20	239.19
Hubei	165.10	7.66	7.65	37.58	12.72	2.40	17.83	1.47	1.37	253.78
Hunan	120.75	11.71	5.94	32.27	9.94	1.34	17.00	1.58	1.00	201.52

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	dry deposition	Biological N fixation	Irrigation	Seed	Total
Jiangxi	59.04	7.35	2.57	19.04	8.37	0.62	13.41	1.06	0.86	112.32
Inner-Mongolia	115.47	4.45	2.35	10.27	5.15	0.37	21.19	1.16	0.91	161.31
Shaanxi	118.85	3.59	1.34	13.02	5.77	1.16	12.72	0.48	0.82	157.74
Gansu	47.69	1.49	1.17	5.62	2.88	0.25	10.42	0.55	0.74	70.81
Qinghai	5.15	0.01	0.74	3.24	0.28	0.00	1.23	0.08	0.08	10.82
Ningxia	21.32	0.50	0.04	1.31	0.71	0.17	3.48	0.23	0.16	27.92
Xinjiang	119.91	3.74	10.77	7.26	1.01	0.17	12.08	2.26	0.99	158.17
Chongqing	55.01	6.35	1.39	8.35	6.10	0.79	8.41	0.25	0.59	87.22
Sichuan	136.59	16.38	6.67	37.16	11.50	1.04	27.64	1.31	1.88	240.16
Guizhou	59.27	3.94	2.36	25.37	8.41	0.89	10.75	0.54	0.78	112.29
Yunnan	127.42	8.96	1.69	28.95	6.59	1.23	13.99	0.93	0.85	190.60
Tibet	2.65	0.00	0.16	4.72	0.18	0.00	0.72	0.13	0.02	8.59
Fujian	56.02	1.67	0.33	16.43	2.43	0.49	6.37	0.54	0.40	84.67
Guangdong	123.64	8.59	0.65	40.76	6.76	1.38	13.00	0.99	0.96	196.72
Guangxi	102.04	26.74	0.47	29.05	10.04	1.18	13.53	0.84	0.78	184.66

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	dry deposition	Biological N fixation	Irrigation	Seed	Total
Hainan	22.00	1.19	0.06	16.36	1.62	0.75	2.03	0.12	0.13	44.26
Total	2927.57	243.99	71.54	680.74	197.07	44.48	413.36	29.93	30.84	4639.51

Table S12. The phosphorus inputs in the 31provinces of China in 2015 (10^4 tons)

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	Irrigation	Seed	Total
Liaoning	47.52	0.31	0.03	5.44	0.16	0.07	0.03	53.56
Jilin	88.71	0.78	0.04	4.99	0.22	0.06	0.04	94.84
Heilongjiang	94.51	0.91	0.25	5.63	0.51	0.23	0.14	102.16
Beijing	2.75	0.08	0.00	0.69	0.01	0.01	0.00	3.53
Tianjin	6.46	0.07	0.01	0.83	0.01	0.01	0.01	7.40
Hebei	98.61	3.39	0.21	8.52	0.21	0.18	0.28	111.40
Shanxi	43.53	0.91	0.02	2.27	0.13	0.07	0.09	47.03
Shandong	154.12	4.55	0.34	15.77	0.24	0.23	0.44	175.69
Henan	254.51	4.32	0.57	10.07	0.26	0.23	0.62	270.57
Shanghai	1.93	0.05	0.00	0.82	0.01	0.01	0.01	2.83

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	Irrigation	Seed	Total
Jiangsu	72.19	1.02	0.48	5.35	0.15	0.19	0.24	79.61
Zhejiang	17.75	0.43	0.12	2.37	0.06	0.07	0.02	20.82
Anhui	87.45	1.27	0.67	6.39	0.19	0.17	0.30	96.42
Hubei	91.60	1.09	1.11	6.37	0.17	0.12	0.15	100.60
Hunan	50.16	1.68	0.87	5.62	0.13	0.13	0.03	58.62
Jiangxi	40.15	1.02	0.35	4.67	0.10	0.09	0.02	46.40
Inner-Mongolia	72.95	0.67	0.26	1.18	0.30	0.12	0.12	75.60
Shaanxi	63.82	0.52	0.19	1.75	0.13	0.05	0.13	66.59
Gansu	32.25	0.21	0.16	0.80	0.17	0.06	0.12	33.77
Qinghai	3.64	0.00	0.11	0.36	0.02	0.01	0.01	4.16
Ningxia	11.82	0.08	0.00	0.18	0.04	0.02	0.02	12.16
Xinjiang	87.27	0.48	1.35	0.93	0.17	0.24	0.16	90.59
Chongqing	25.02	0.87	0.19	1.48	0.08	0.02	0.05	27.71
Sichuan	66.00	2.29	0.96	5.70	0.22	0.11	0.19	75.46
Guizhou	21.20	0.55	0.35	4.81	0.15	0.05	0.08	27.16

	Mineral fertilizer	Straw	Cake fertilizer	Excrements of animal	wet deposition	Irrigation	Seed	Total
Yunnan	51.47	1.21	0.23	3.99	0.20	0.08	0.09	57.26
Tibet	1.90	0.00	0.02	0.71	0.01	0.01	0.00	2.66
Fujian	27.92	0.22	0.03	4.47	0.04	0.05	0.02	32.73
Guangdong	49.19	1.11	0.05	7.20	0.08	0.08	0.03	57.75
Guangxi	61.53	3.48	0.04	5.22	0.14	0.07	0.03	70.50
Hainan	11.48	0.15	0.00	2.64	0.02	0.01	0.00	14.31
Total	1739.37	33.70	9.01	127.20	4.32	2.84	3.47	1919.90

Table S13. The N outputs in the 31provinces of China in 2015 (10^4 tons)

	Crop uptake	Denitrification	Ammonia volatilization	Leaching	Runoff	Volatilization	Total
Liaoning	77.98	13.86	15.53	1.38	1.60	4.01	114.35
Jilin	128.34	16.96	19.16	1.55	1.88	5.30	173.19
Heilongjiang	233.18	17.53	19.05	4.13	4.64	12.03	290.57
Beijing	2.99	1.03	1.25	0.16	0.08	0.16	5.66
Tianjin	7.81	1.78	2.09	0.43	0.19	0.46	12.76

	Crop uptake	Denitrification	Ammonia volatilization	Leaching	Runoff	Volatilization	Total
Hebei	150.89	23.72	29.69	7.64	3.14	8.19	223.27
Shanxi	47.66	6.48	8.50	2.72	1.04	3.07	69.47
Shandong	217.58	30.09	38.20	10.62	4.31	10.90	311.69
Henan	262.80	41.65	51.50	13.23	5.30	13.78	388.26
Shanghai	4.01	1.83	1.38	0.55	0.42	0.28	8.46
Jiangsu	125.27	31.90	24.37	11.64	9.17	7.33	209.67
Zhejiang	27.97	11.11	7.43	3.72	3.00	2.08	55.31
Anhui	136.20	27.11	22.34	12.15	9.86	8.59	216.24
Hubei	104.00	30.65	25.22	11.32	8.80	7.41	187.40
Hunan	100.50	25.04	18.47	12.88	11.06	7.94	175.89
Jiangxi	63.53	12.66	9.46	8.02	7.44	5.11	106.21
Inner-Mongolia	122.18	13.00	17.47	2.63	0.34	6.33	161.94
Shaanxi	49.53	14.98	17.91	1.74	0.52	3.70	88.37
Gansu	45.82	5.51	7.40	1.54	0.49	3.37	64.14
Qinghai	4.54	0.94	1.14	0.16	0.05	0.40	7.23
Ningxia	14.99	2.87	2.99	0.44	0.18	0.92	22.38

	Crop uptake	Denitrification	Ammonia volatilization	Leaching	Runoff	Volatilization	Total
Xinjiang	85.98	13.02	17.70	2.03	0.44	5.00	124.18
Chongqing	38.16	8.24	8.29	5.07	3.78	3.12	66.66
Sichuan	122.27	23.37	22.49	13.00	9.84	8.73	199.70
Guizhou	41.88	10.96	11.07	6.94	5.00	4.40	80.25
Yunnan	71.24	19.75	20.63	8.49	6.34	5.57	132.02
Tibet	1.36	0.89	0.98	0.14	0.09	0.09	3.56
Fujian	24.17	11.54	8.84	3.96	3.10	2.09	53.69
Guangdong	57.10	25.04	20.40	8.12	6.49	4.45	121.59
Guangxi	91.22	18.97	16.57	9.14	7.26	5.55	148.70
Hainan	8.31	5.10	4.98	1.42	1.11	0.76	21.67
Total	2469.47	467.54	472.47	166.93	116.97	151.09	3844.46

Table S14. the nutrient balance of the farmland in the 31provinces in 2015

	N input (10^4tons)	N output (10^4tons)	N balance (10^4tons)	N density (t km^{-2})	P input (10^4tons)	P output (10^4tons)	P balance (10^4tons)	P density (t km^{-2})
Liaoning	141.42	53.56	27.07	4.97	114.35	36.68	16.88	3.10

	N input (10^4tons)	N output (10^4tons)	N balance (10^4tons)	N density (t km^{-2})	P input (10^4tons)	P output (10^4tons)	P balance (10^4tons)	P density (t km^{-2})
Jilin	174.05	94.84	0.86	0.12	173.19	59.69	35.15	4.98
Heilongjiang	234.85	102.16	-55.72	-3.50	290.57	91.36	10.80	0.68
Beijing	10.76	3.53	5.09	14.38	5.66	1.76	1.77	5.01
Tianjin	18.04	7.40	5.28	11.31	12.76	4.14	3.27	7.00
Hebei	273.86	111.40	50.59	6.87	223.27	72.09	39.31	5.34
Shanxi	84.23	47.03	14.76	3.31	69.47	24.77	22.26	4.98
Shandong	357.20	175.69	45.51	5.46	311.69	106.02	69.67	8.36
Henan	462.57	270.57	74.32	8.93	388.26	137.61	132.97	15.97
Shanghai	13.60	2.83	5.13	24.86	8.46	2.04	0.78	3.80
Jiangsu	245.28	79.61	35.61	7.30	209.67	58.84	20.77	4.26
Zhejiang	80.94	20.82	25.63	10.00	55.31	14.09	6.73	2.62
Anhui	239.19	96.42	22.95	3.69	216.24	63.27	33.15	5.33
Hubei	253.78	100.60	66.37	11.57	187.40	56.99	43.61	7.60
Hunan	201.52	58.62	25.63	5.32	175.89	47.88	10.74	2.23
Jiangxi	112.32	46.40	6.11	1.79	106.21	30.55	15.85	4.65

	N input (10⁴tons)	N output (10⁴tons)	N balance (10⁴tons)	N density (t km^{-2})	P input (10⁴tons)	P output (10⁴tons)	P balance (10⁴tons)	P density (t km^{-2})
Inner-Mongolia	161.31	75.60	-0.64	-0.07	161.94	53.17	22.43	2.41
Shaanxi	157.74	66.59	69.37	14.41	88.37	30.00	36.59	7.60
Gansu	70.81	33.77	6.66	1.18	64.14	21.67	12.10	2.15
Qinghai	10.82	4.16	3.60	6.05	7.23	2.56	1.60	2.68
Ningxia	27.92	12.16	5.54	4.13	22.38	7.57	4.59	3.42
Xinjiang	158.17	90.59	33.99	5.85	124.18	43.63	46.96	8.08
Chongqing	87.22	27.71	20.56	7.61	66.66	17.94	9.77	3.62
Sichuan	240.16	75.46	40.46	5.42	199.70	56.09	19.37	2.59
Guizhou	112.29	27.16	32.04	6.81	80.25	19.41	7.75	1.65
Yunnan	190.60	57.26	58.58	7.47	132.02	33.15	24.11	3.07
Tibet	8.59	2.66	5.03	11.31	3.56	1.04	1.61	3.63
Fujian	84.67	32.73	30.98	14.69	53.69	14.52	18.21	8.63
Guangdong	196.72	57.75	75.13	19.33	121.59	28.93	28.82	7.41
Guangxi	184.66	70.50	35.96	6.55	148.70	37.88	32.62	5.95
Hainan	44.26	14.31	22.59	13.71	21.67	5.54	8.78	5.33

Table S15. Comparison of the N budget in different research(Ti *et al.*, 2012; Gu *et al.*, 2015; He *et al.*, 2018)

Tg N yr^{-1}	Gu *et al.*, 2015 in 2010	Ti *et al.*, 2012 in 2007	He *et al.*, 2018 in 2010	This Study in 2015
Chemical fertilizer	28.9	27.8	30.7	29.3
Organic fertilizer	8.0	-	16.1	10.0
Deposition	2.7	2.3	2.9	2.4
Biological Nitrogen Fixation	4.6	-	3.7	4.1
Irrigation	0.6	-	0.8	0.3
Seed	-	-	0.4	0.3
Total input	45.8	-	54.7	46.4
Crop uptake	18.0	30.1[a]	21.6	24.7
Denitrification	7.9	-	8.2	4.7
Ammonia volatilization	7.7	6.7	7.0	4.7
leaching	2.1	2.1[b]	2.4	1.7
runoff	2.4	-	2.5	1.2
Total output	38.0	-	41.7	38.4

[a] This value included the crop uptake and denitrification

[b] This value included leaching and runoff

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