

Evaluation of nitrogen fertilizer fates and related environmental risks for main cereals in China's croplands from 2004 to 2018

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1. Materials and Methods

1.1 Definition and estimates N emissions

1.1.1 N₂O-N emissions

N₂O is calculated based on the Intergovernmental Panel on Climate Change (IPCC) methodology [1], specific parameters of the direct emissions and indirect emissions are derived from the investigation on compiling the works of the Provincial Agricultural Greenhouse Gas Inventories in China [2]. The parameters of N₂O-N concentration per unit of economic yield based on previous studies are summarized in Table S5 [3–37]. The equation is shown as follows:

$$N_{N_2O} = N_{input} \times N_{2O_{emission}} \quad (1)$$

where the N_{input} (kg N ha⁻¹) is the sum of chemical fertilizer N (N_{fert}), manure fertilizer N from livestock and humans (N_{man}), straw N returned to the soil (N_{straw}), N fixed via symbiotic and non-symbiotic pathways (N_{fix}), dry and wet N deposition from the atmosphere (N_{depo}) and N from irrigation water (N_{irri}). $N_{2O_{emission}}$ is emission factor of total N input.

1.1.2 NH₃-N emissions

NH₃-N is calculated by the chemical fertilizers and organic manures application and regional emission factors, these parameters are derived from Table S5 [3–37]. Since inputs of chemical fertilizers and organic manures are only considered in this study, the equation is shown as follows:

$$N_{NH_3} = N_{fert} \times NH_{3_{volatilization}} + N_{man} \times NH_{3_{volatilization}} \quad (2)$$

where NH₃-N is total NH₃ volatilization from cropland, (N_{fert}) is chemical fertilizer N, N_{man} is manure fertilizer N from livestock and humans NH₃ volatilization is emission factor of fertilizers volatilized as NH₃.

Table S1. Summary of main arable crops and soil types at six regions of China in 2004–2018

Region ¹	Province ²	Maize	Wheat	Single rice	Double rice	Main soil types
Northeast (NE)	HLJ, JL, LN	HLJ, JL, LN	HLJ	HLJ, JL, LN		Black soil, cinnamon soil, meadow soil
North central (NC)	BJ, HEB, HEN, SX, SD, TJ	BJ, HEB, HEN, SX, SD, TJ	BJ, HEB, HEN, SX, SD, TJ	HEB, HEN, SD, TJ		Fluvo-aquic soil, cinnamon soil, saline-alkali soil, brown soil
Northwest (NW)	GS, NMG, NX, QH, SNX, XJ	GS, NMG, NX, SNX, XJ	GS, NMG, NX, QH, SNX, XJ	NMG, NX, SNX		Loess soil, desert soil, irrigation-silting soil, fluvo-aquic soil, chestnut soil, gray calcareous soil
The middle and lower reaches of Yangtze River (MLYR)	AH, HUB, HUN, JS, JX, SH, ZJ	AH, JS, HUB	AH, JS, HUB, SH	AH, HUB, HUN, JS, JX, SH, ZJ	AH, HUB, JX, ZJ	Fluvo-aquic soil, yellow brown soil, red soil, paddy soil
Southeast (SE)	FJ, GD, GX, HN	GX		FJ	FJ, GD, GX, HN	Red soil, yellow brown soil, purple soil, paddy soil
Southwest (SW)	CQ, GZ, SC, XZ, YN	CQ, GZ, SC, YN	CQ, GZ, SC, YN	CQ, GZ, SC, YN		Red soil, yellow brown soil, purple soil, paddy soil

¹Six agricultural regions were grouped based on geographical locations and China's administrative divisions: NE, northeast; NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

²AH, Anhui; BJ, Beijing; CQ, Chongqing; FJ, Fujian; GD, Guangdong; GS, Gansu; GX, Guangxi; GZ, Guizhou; HEB, Hebei; HEN, Henan; HLJ, Heilongjiang; HN, Hainan; HUB, Hubei; HUN, Hunan; JL, Jilin; JS, Jiangsu; JX, Jiangxi; LN, Liaoning; NMG, Inner Mongolia; NX, Ningxia; QH, Qinghai; SC, Sichuan; SD, Shandong; SH, Shanghai; SNX, Shaanxi; SX, Shanxi; TJ, Tianjin; XJ, Xinjiang; XZ, Tibet; YN, Yunnan; ZJ, Zhejiang.

Table S2. N content in air-dry grain and straw, ratio of grain to straw and ratio of straw returning for maize, wheat, single rice and double rice in China [38–55].

Crop	Region	N content in grain (%)	N content in straw (%)	Grian /straw	Ratio of straw returning (%)
Maize	NE	2.25	0.89	0.90	25
	NW	2.25	0.89	0.77	50
	NC	2.25	0.89	0.81	80
	MLYR	2.25	0.89	0.77	80
	SE	2.25	0.89	0.79	50
	SW	2.25	0.89	0.78	30
Wheat	NE	2.12	0.57	0.64	85
	NW	2.12	0.57	0.91	82
	NC	2.12	0.57	0.75	90
	MLYR	2.12	0.57	0.72	88
	SW	2.12	0.57	0.67	85
Single rice	NE	1.30	0.82	1.10	45
	NW	1.30	0.82	0.97	55
	NC	1.30	0.82	1.00	70
	MLYR	1.30	0.82	0.93	72
	SE	1.30	0.82	1.08	72
	SW	1.30	0.82	1.05	50
Double rice	MLYR	1.30	0.82	0.93	72
	SE	1.30	0.82	1.08	72

¹NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

Table S3. N input parameters of non-fixation N, deposition N and irrigation N for maize, wheat, single rice and double rice [51–54, 56–58].

Crop	Region ¹	Non-fixation (kg ha ⁻¹)	Deposition (kg ha ⁻¹)	Irrigation (kg ha ⁻¹)
Maize	NE	15.0	10.8	3.5
	NW	15.0	7.2	5.0
	NC	15.0	18.2	6.9
	MLYR	15.0	26.6	5.2
	SE	15.0	26.2	3.4
	SW	15.0	15.3	3.1
Wheat	NE	15.0	9.4	7.9
	NW	15.0	9.4	7.9
	NC	15.0	12.9	9.9
	MLYR	15.0	14.5	5.8
	SW	15.0	14.5	5.8
Single rice	NE	33.0	9.4	7.7
	NW	33.0	9.4	7.7
	NC	33.0	12.9	9.9
	MLYR	33.0	14.5	5.8
	SE	33.0	14.5	5.8
	SW	33.0	14.5	5.8
Double rice	MLYR	44.0	19.3	7.7
	SE	44.0	19.3	7.7

¹NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

Table S4. Livestock feeding period, daily excretion, N content and percentage for field application (based on fresh) [2,57].

Livestock	Feeding phase	Population Structure (%)	Feeding Period (d)	Daily excretion/urine and N content ¹			Rate to field (%)	
				Excretion (kg d)	Urine (kg d)	Content (%)	Urine (%)	2000s–2010s
Human		100	365	0.31	1.60	1.16	0.53	33
Dairy	Adult cattle	53	365	37.5	18.8	0.43	0.87	30
	Young dairy cattle	37	365	21.4	10.7	0.43	0.87	30
	Calf	10	180	5.80	2.9	0.43	0.87	30
Beef	Heifers for slaughter	73	365	18.1	9.1	0.38	0.51	30
	Fattening cattle	16	270	12.1	6.0	0.38	0.51	30
	Calf	11	120	4.00	2.0	0.38	0.51	30
Pig	Breeding pigs	11	365	5.50	3.0	0.55	0.17	65
	Fattening pigs	56	180	4.00	3.0	0.55	0.17	65
	Piglets	37	90	1.50	3.0	0.55	0.17	65
Sheep/Goat	Adult sheep/goat	70	365	2.60	-	0.89	-	33
	Lambs	30	180	1.40	-	0.89	-	33
Poultry		100	210	0.13	-	0.94	-	45
Horse		100	365	10.0	5.0	0.42	0.69	44
Donkey		100	365	10.0	5.0	0.45	0.71	44
Mule		100	365	10.0	5.0	0.33	0.60	44

¹ The N contents in daily excretion and urine were evaluated together based on parameters availability.

Table S5. Nitrogen losses coefficients (%) for different components for each crop and region in China's croplands [3–37].

Crop	Region ¹	N ₂ O emission		NO emission		N ₂ emission		NH ₃ volatilization		N runoff		N leaching	
		2004– 2008	2014– 2018	2004– 2008	2014– 2018	2004– 2008	2014– 2018	2004– 2008	2014– 2018	2004– 2008	2014– 2018	2004– 2008	2014– 2018
Maize	NE	0.65	0.65	0.52	0.52	9.70	9.71	6.69	6.67	0.28	0.29	2.69	2.7
	NW	0.68	0.69	0.54	0.55	10.14	10.31	6.47	6.44	0.27	0.28	5.97	6.05
	NC	1.66	1.61	1.33	1.29	24.94	24.14	11.74	11.81	0.83	0.80	20.67	21.12
	MLYR	0.89	0.91	0.71	0.73	13.31	13.71	7.60	7.58	1.05	1.09	11.27	11.38
	SE	0.87	0.88	0.70	0.71	13.10	13.26	7.61	7.60	1.06	1.08	10.7	10.94
	SW	0.85	0.84	0.68	0.67	12.69	12.64	7.63	7.64	0.95	0.94	8.57	8.72
Wheat	NE	0.32	0.32	0.26	0.26	4.80	4.80	8.41	8.41	0.28	0.28	4.71	4.7
	NW	0.36	0.37	0.29	0.30	5.36	5.56	7.73	7.67	0.27	0.29	8.27	8.95
	NC	0.46	0.47	0.37	0.38	6.91	7.06	7.53	7.49	0.83	0.88	19.33	19.51
	MLYR	1.39	1.48	1.11	1.18	20.78	22.17	12.83	12.84	1.05	1.11	7.26	8.03
	SW	1.06	1.07	0.85	0.85	15.87	15.99	12.76	12.76	0.95	0.97	4.61	4.68
Single rice	NE	0.25	0.25	0.20	0.20	3.78	3.80	11.12	11.21	1.24	1.22	2.61	2.62
	NW	0.26	0.26	0.20	0.20	3.84	3.83	10.94	10.96	1.33	1.32	2.65	2.64
	NC	0.27	0.27	0.22	0.21	4.10	3.98	10.76	10.82	1.53	1.44	2.83	2.75
	MLYR	0.83	0.83	0.67	0.66	12.48	12.40	19.87	19.87	3.90	3.88	2.67	2.66
	SE	0.74	0.76	0.59	0.61	11.03	11.47	18.44	18.42	2.37	2.38	2.61	2.61
	SW	1.00	0.88	0.80	0.70	14.95	13.20	18.27	18.33	2.43	2.40	2.63	2.62
Double rice	MLYR	0.72	0.73	0.58	0.58	10.81	10.95	19.82	19.83	3.38	3.43	2.97	2.98
	SE	0.58	0.61	0.46	0.48	8.65	9.09	18.62	18.58	2.35	2.35	2.97	2.97

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

Table S6. Nitrogen (N) input (kg N ha⁻¹) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	284.9±61.2c	292.0±36.7b	200.2±15.3c	198.7±8.8c	343.1±92.7c	317.2±59.4e	- ²	-
NW	379.1±35.0ab	397.3±36.6a	346.6±77.8b	378.4±102.7b	406.9±68.9b	401.0±43.7bc	-	-
NC	411.1±72.6a	395.9±62.3a	429.8±83.7a	454.3±64.7a	501.9±65.8a	466.3±37.3a	-	-
MLYR	388.5±27.6ab	397.3±34.9a	351.9±36.8b	371.7±46.8b	421.3±57.7b	417.3±49.2b	604.3±36.4	637.5±37.4
SE	380.3±17.2ab	388.4±32.5a	-	-	343.5±6.28c	349.5±5.86de	610.4±49.2	625.9±43.6
SW	362.8±44.9b	360.3±37.6a	250.4±42.9c	254.0±58.2c	389.2±45.2bc	370.8±36.8cd	-	-
China	367.8±64.4	371.9±57.0	346.1±95.1	366.6±105.7	414.0±79.6	398.8±63.6	607.0±42.2	632.3±40.2

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

Table S7. Nitrogen (N) output (kg N ha⁻¹) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	241.8±23.3d	260.5±28.4e	146.1±20.1d	173.0±6.3c	255.3±21.7e	269.3±25.4d	- ²	-
NW	298.8±43.5c	336.4±45.6c	217.6±34.1c	244.0±44.2b	273.2±23.5de	286.0±13.8cd	-	-
NC	385.9±45.2a	393.3±38.2a	337.8±42.3a	388.5±26.8a	300.5±28.0bc	318.6±24.8b	-	-
MLYR	339.7±25.3b	379.0±30.1b	301.4±23.6b	331.5±29.3b	363.7±22.4a	382.5±23.2a	560.0±20.4	615.6±22.1
SE	312.2±23.0c	320.2±23.6d	-	-	304.4±9.7cd	319.5±8.9c	509.8±38.1	535.0±39.8
SW	315.0±22.9bc	335.7±27.6cd	169.4±27.0d	177.0±28.7c	352.4±25.5ab	354.4±19.4b	-	-
China	351.2±65.2	373.8±62.2	255.3±62.4	285.8±69.7	320.4±33.3	234.3±33.8	537.5±33.8	579.5±41.2

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

Table S8. Global warming potential (GWP) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	139.2±28.4b	228.4±58.9b	4.9±0.24c	2.0±0.60c	25.4±10.1c	34.6±25.2b	- ²	-
NW	65.8±45.0b	112.2±77.2bc	23.7±14.5c	29.4±15.4c	3.1 ±0.43c	3.2±0.82b	-	-
NC	313.7±192.4a	373.9±199.4a	110.3±76.5b	128.9±82.1b	7.5±6.79c	7.6±8.07b	-	-
MLYR	52.4±12.3b	79.6±19.5c	181.4±86.3a	257.4±112.4a	122.3±83.1a	124.9±88.1a	204.7±121.6	204.9±126.2
SE	54.7±5.5b	63.6±4.8c	-	-	71.6±4.5b	23.7±1.18b	147.3±93.1	140.7±97.8
SW	88.8±40.1b	107.3±46.5bc	52.5±32.7bc	35.7±20.4c	139.1±77.4a	112.0±61.5a	-	-
China	145.3±85.9	191.8±107.4	83.1±55.2	101.2±76.2	72.7±61.5	67.7±56.1	179.2±106.2	176.4±102.6

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

Table S9. Acidification potential (AP) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	8.73±1.78b	14.3±3.68a	0.77±0.04b	0.32±0.10b	6.80±2.70cd	9.28±6.76bc	- ²	-
NW	3.82±2.68c	6.37±4.60b	3.11±1.90b	3.69±2.46b	0.80±0.12d	0.82±0.20d	-	-
NC	13.4±10.1a	16.6±12.5a	10.9±10.46a	12.4±12.35a	1.79±1.75d	1.87±2.10d	-	-
MLYR	2.72±0.64c	4.00±0.98b	10.2±7.53a	13.56±10.06a	17.71±12.1a	18.20±12.9a	34.2±27.8	33.7±29.5
SE	2.89±0.29c	3.32±0.25b	-	-	10.89±0.68bc	3.46±0.25cd	28.9±17.4	26.2±17.3
SW	4.85±2.19bc	5.90±3.18b	3.84±3.31b	2.59±2.65b	15.46±8.54ab	14.15±7.74ab	-	-
China	7.11±6.92	9.70±8.74	6.72±7.65	7.71±9.47	10.31±10.47	10.24±9.93	31.8±23.8	30.4±25.1

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

Table S10. The water pollution level of surface runoff emissions (WPL-SR) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	0.0034±0.0018b	0.0050±0.0018b	0.0001±0.0000b	0.0000±0.0001b	0.0058±0.0008b	0.0070±0.0023b	- ²	-
NW	0.0028±0.0019b	0.0043±0.0028b	0.0024±0.0026b	0.0018±0.0015b	0.0128±0.0173ab	0.0119±0.0158ab	-	-
NC	0.0379±0.0206a	0.0428±0.0244a	0.0363±0.0250a	0.0383±0.0276a	0.0065±0.0019b	0.0069±0.0035b	-	-
MLYR	0.0021±0.0007b	0.0030±0.0009b	0.0035±0.0042b	0.0079±0.0068b	0.0242±0.0237a	0.0235±0.0241a	0.0138±0.0079	0.0135±0.0089
SE	0.0007±0.0001b	0.0008±0.0001b	0.0031±0.0046b	0.0003±0.0002b	0.0038±0.0004b	0.0010±0.0000b	0.0081±0.0020	0.0072±0.0024
SW	0.0014±0.0003b	0.0017±0.0003b	-	-	0.0049±0.0017b	0.0043±0.0012b	-	-
China	0.0120±0.0192	0.0141±0.0218	0.0123±0.0204	0.0130±0.0220	0.0125±0.0169	0.0121±0.0168	0.0113±0.0066	0.0107±0.0075

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

Table S11. The water pollution level of leaching emissions (WPL-LEA) for maize, wheat, single rice and double rice between 2004–2008 and 2014–2018 at the region scale in China farmlands, respectively.

Region ¹	Maize		Wheat		Single rice		Double rice	
	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018	2004–2008	2014–2018
NE	0.0045±0.0021b	0.0066±0.0019b	0.0004±0.0000b	0.0002±0.0001b	0.0017±0.0003b	0.0020±0.0005ab	- ²	-
NW	0.0023±0.0011b	0.0038±0.0018b	0.0065±0.0055b	0.0059±0.0035b	0.0006±0.0005b	0.0006±0.0005c	-	-
NC	0.0680±0.0313a	0.0772±0.0405a	0.0578±0.0306a	0.0727±0.0428a	0.0008±0.0004b	0.0010±0.0007bc	-	-
MLYR	0.0076±0.0024b	0.0116±0.0034b	0.0072±0.0050b	0.0105±0.0078b	0.0030±0.0022a	0.0029±0.0022a	0.0007±0.0004	0.0006±0.0004
SE	0.0032±0.0003b	0.0038±0.0003b			0.0008±0.0001b	0.0003±0.0001c	0.0006±0.0001	0.0005±0.0002
SW	0.0066±0.0025b	0.0074±0.0021b	0.0007±0.0003b	0.0003±0.0002b	0.0013±0.0007b	0.0011±0.0006bc	-	-
China	0.0221±0.0255	0.0259±0.0272	0.0199±0.0164	0.0245±0.0181	0.0017±0.0015	0.0014±0.0011	0.0006±0.0003	0.0006±0.0003

¹ NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

² “-” indicates that the crop is not planted in the area.

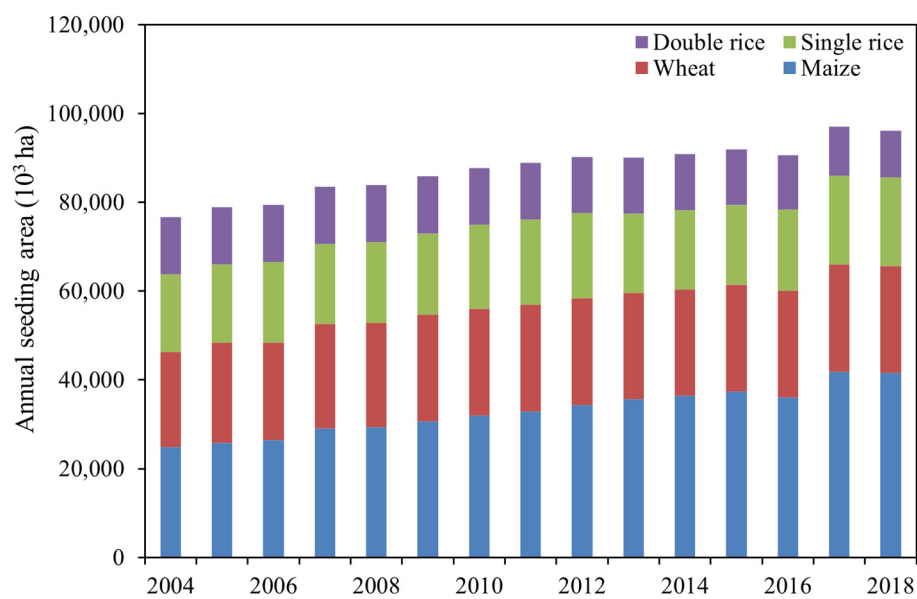


Figure. S1. Annual seeding area of main crops from 2004 to 2018 in China farmlands.

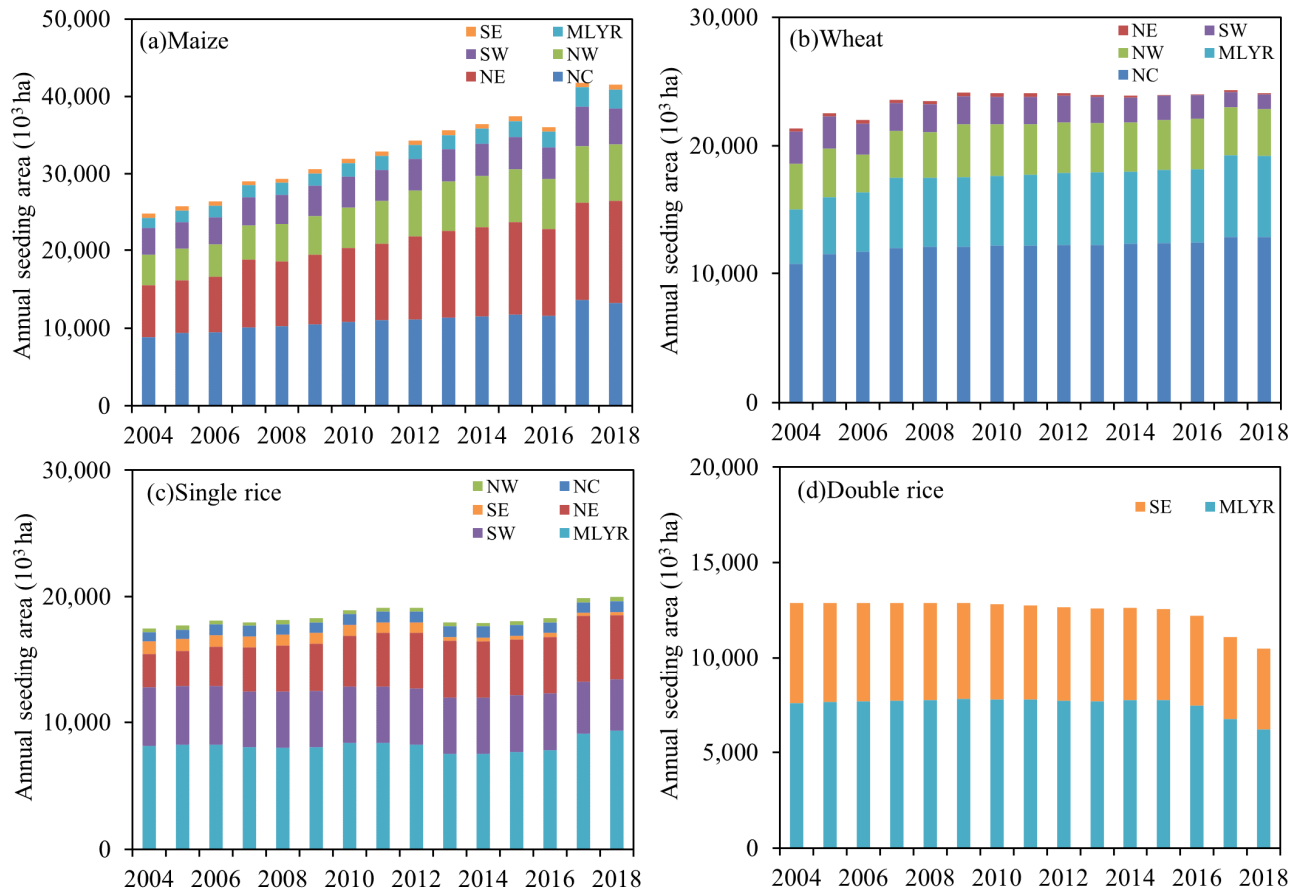


Figure S2. Annual seeding area of (a) maize, (b) wheat, (c) single rice and (d) double from 2004 to 2018 in China farmlands, respectively. NE, northeast; NC, north central; NW, northwest; MLYR, the middle and lower reaches of the Yangtze River; SE, southeast; SW, southwest.

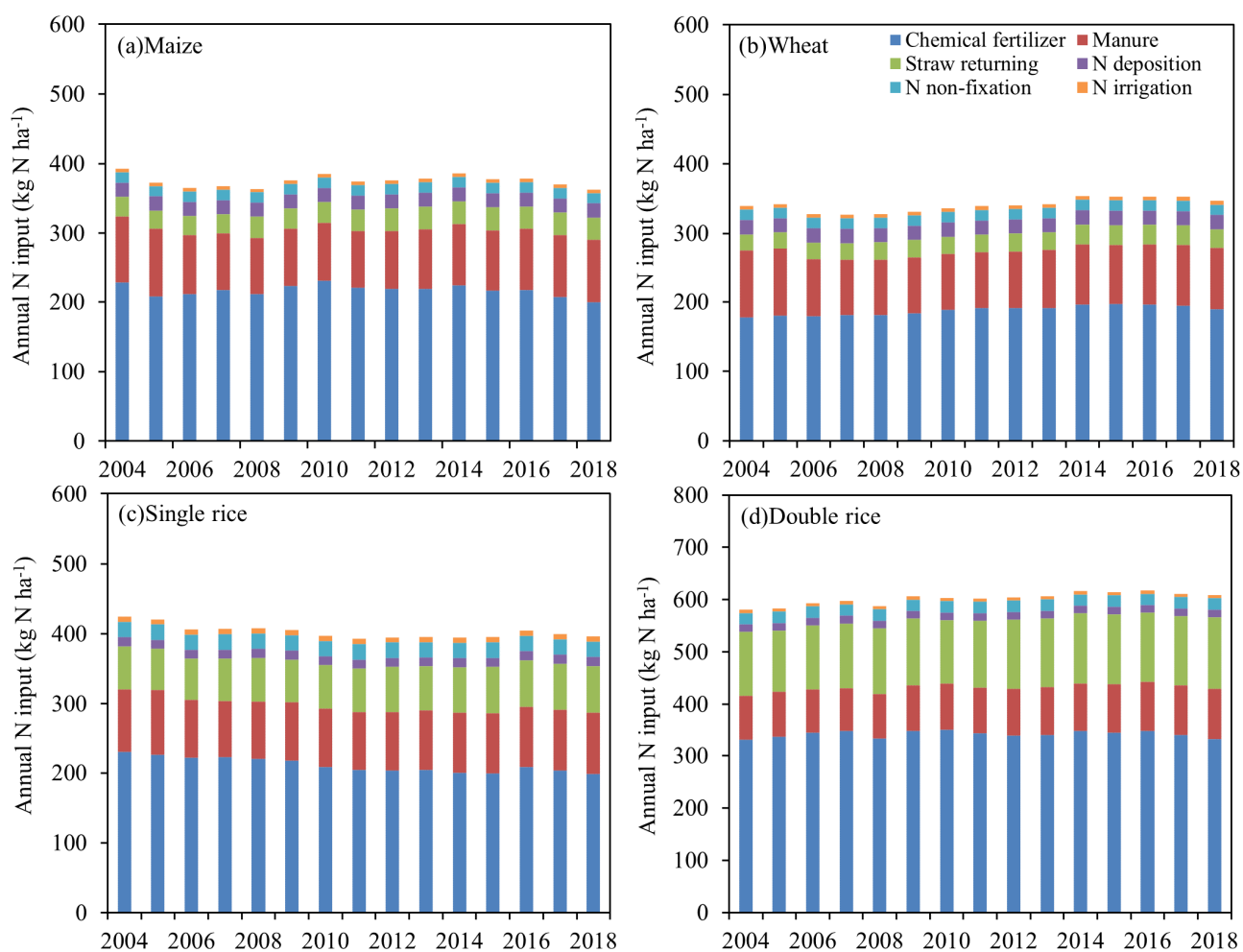


Figure S3. Composition of nitrogen (N) input (kg N ha^{-1}) from 2004 to 2018 for (a) maize, (b) wheat, (c) single rice and (d) double from 2004 to 2018 in China farmlands, respectively.

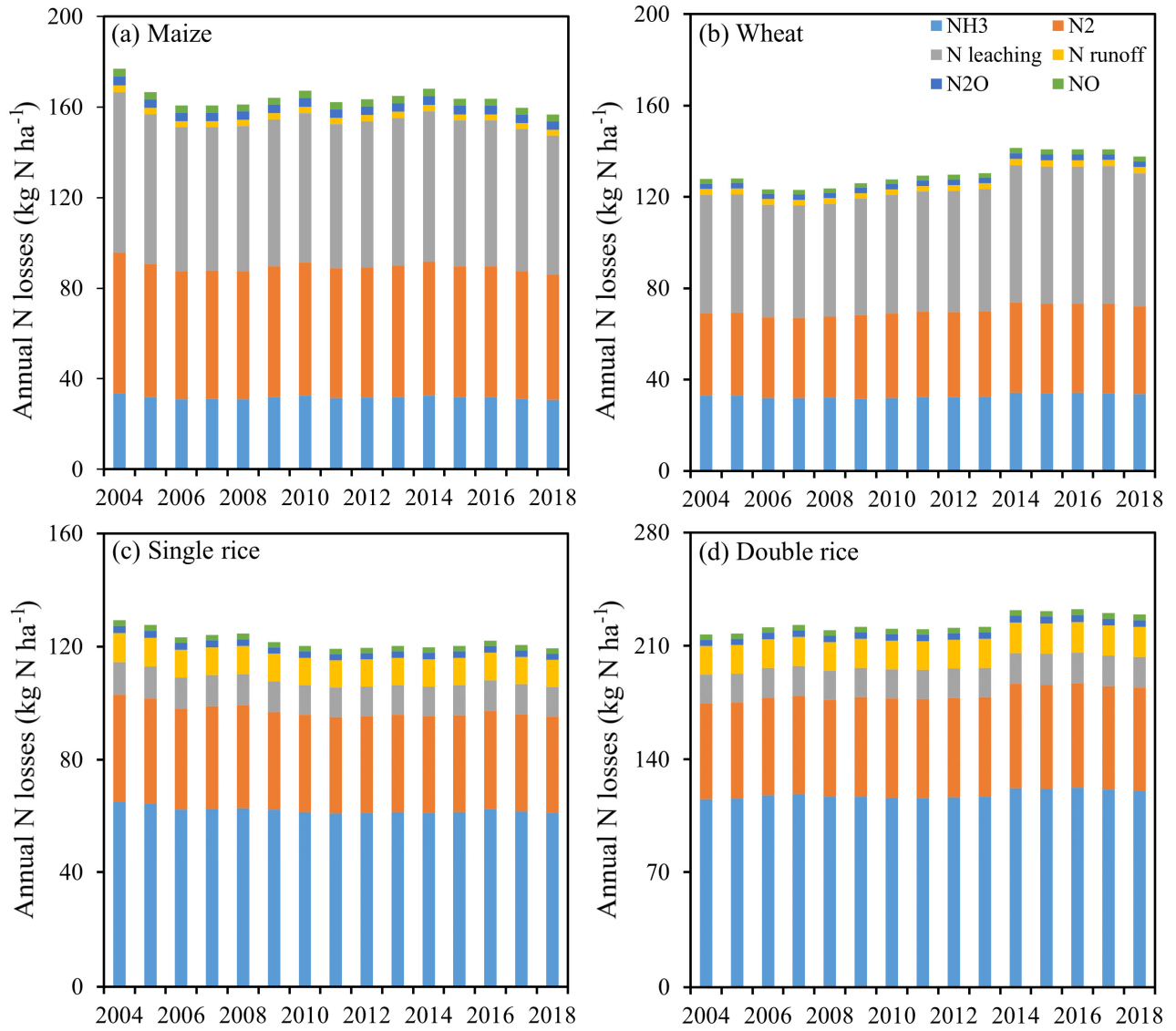


Figure S4. Composition of annual nitrogen (N) losses (kg N ha⁻¹) from 2004 to 2018 for (a) maize, (b) wheat, (c) single rice and (d) double from 2004 to 2018 in China farmlands, respectively.

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