



*Supplement of*

## **High-resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008**

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# Supporting Information

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## **Section S1: Methodology**

Text S1: Equations for ammonia ( $\text{NH}_3$ ) emissions from each source

The equations for  $\text{NH}_3$  emissions from synthetic fertilizer, rural excrement and cake fertilizer were found in Xu et al. (2015). The equations for livestock manure spreading and straw returning are as follows:

1. Livestock manure spreading

$$E_{LM} = \frac{17}{14} \cdot \sum_L \sum_R LN_{(L,R)} \cdot EF_{(L,R)} \quad (1)$$

where

$$EF_{(L,R)} = LM_{(L,R)} \cdot (1 - \nu_{(H)}) \cdot (1 - \nu_{(ST)}) \cdot \nu_{(MS)} \quad (2)$$

$$LM_{(L,R)} = LN_{(L,R)} \cdot N_{er(L,R)} \cdot FD_{(L)} / 1000 \quad (3)$$

and  $E_{LM}$  is the  $\text{NH}_3$  emission from livestock manure spreading in region R,  $\text{TgNH}_3 \text{ yr}^{-1}$ ;  $LN_{(L)}$  is the number of head of species/category  $L$  in region R;  $EF_{(L)}$  is the emission factor for  $\text{NH}_3$  emissions from livestock manure spreading for livestock species/category  $L$  in region R,  $\text{kg NH}_3 \cdot \text{N} \cdot \text{kg}^{-1}$ ; and  $LM_{(L)}$  is the annual average N excretion in manure management for livestock species/category  $L$  in region R,  $\text{kg N} \cdot \text{yr}^{-1}$ . The livestock species/category considered in this study included pigs, beef cattle, dairy cattle, sheep, poultry, horses, donkeys and mules.  $\nu_{(H)}$ ,  $\nu_{(ST)}$  and  $\nu_{(MS)}$  are the N volatilization rates of animal housing, manure storage and manure spreading, respectively.  $N_{er(L)}$  is the N excretion rate for livestock species/category  $L$  in region R, g

$\text{N}\cdot\text{head}^{-1}\cdot\text{day}^{-1}$ .  $FD_{(L)}$  is the number of feeding days for livestock species/category  $L$ , and 17/14 is the conversion coefficient of  $\text{NH}_3\text{-N}$  emissions to  $\text{NH}_3$  emissions (Zhou et al., 2014; Yang, 2008).

## 2. Straw returning

$$E_{CR} = \sum_i \sum_R NSR_{(i,R)} \cdot EF_{CR} \quad (4)$$

where  $E_{CR}$  is the  $\text{NH}_3$  emission from straw returning in region R,  $\text{TgNH}_3 \text{ yr}^{-1}$ ;  $NSR_{(i,R)}$  is the annual nitrogen quantity of straw returning for crop  $i$  in region R,  $\text{kgN} \cdot \text{kg}^{-1} \text{yr}^{-1}$ ; and  $EF_{CR}$  is the emission factor for  $\text{NH}_3$  emission from straw returning for crop types  $i$  in region R,  $\text{kg NH}_3 \cdot \text{kg}^{-1} \cdot \text{N}^{-1}$ . The crop types considered in this study were maize, rice, wheat, barley, sugarcane, millet, potatoes, other tubers, soybeans, peanuts, rapeseed, sesame, other oil crops, sorghum, highland barley, other beans and alfalfa.  $NSR_{(i,R)}$  was derived from Zhou et al. (2014)

## Section S2: Data on activity data, RPs and EFs

Table S1  $\text{NH}_3$  EFs of livestock manure spreading for 34 provinces in China and of straw returning

Province	EFs: $\text{kgNH}_3 \text{ head}^{-1} \text{yr}^{-1}$					
	Pigs	Cattle	Dairy cattle	Sheep	Horses/Donkeys/Mules	Poultry
Beijing	1.458	6.891	25.820	0.636	3.611	0.061
Tianjin	1.458	6.891	25.820	0.636	3.611	0.061
Hebei	1.458	6.891	25.820	0.636	3.611	0.061
Shanxi	1.458	6.891	25.820	0.636	3.611	0.061
Inner Mongolia	1.458	6.891	25.820	0.636	3.611	0.061
Liaoning	2.531	14.287	24.263	0.815	4.626	0.089
Jilin	2.531	14.287	24.263	0.815	4.626	0.089
Heilongjiang	2.531	14.287	24.263	0.815	4.626	0.089
Shanghai	1.114	14.539	20.197	0.591	3.355	0.049
Jiangsu	1.114	14.539	20.197	0.591	3.355	0.049
Zhejiang	1.114	14.539	20.197	0.591	3.355	0.049
Anhui	1.114	14.539	20.197	0.591	3.355	0.049
Taiwan	1.114	14.539	20.197	0.591	3.355	0.049
Jiangxi	1.114	14.539	20.197	0.591	3.355	0.049
Shandong	1.114	14.539	20.197	0.591	3.355	0.049
Henan	1.962	6.246	33.275	0.702	3.986	0.034
Hubei	1.962	6.246	33.275	0.702	3.986	0.034
Hunan	1.962	6.246	33.275	0.702	3.986	0.034
Guangdong	1.962	6.246	33.275	0.702	3.986	0.034
Guangxi	1.962	6.246	33.275	0.702	3.986	0.034
Hainan	1.962	6.246	33.275	0.702	3.986	0.034
Sichuan	0.866	9.862	20.197	0.712	4.041	0.034
Yunnan	0.866	9.862	20.197	0.712	4.041	0.034
Guizhou	0.866	9.862	20.197	0.712	4.041	0.034

Chongqing	0.866	9.862	20.197	0.712	4.041		0.034
Tibet	0.866	9.862	20.197	0.712	4.041		0.034
Ningxia	1.613	9.862	17.502	0.644	3.657		0.089
Xinjiang	1.613	9.862	17.502	0.644	3.657		0.089
Qinghai	1.613	9.862	17.502	0.644	3.657		0.089
Shannxi	1.613	9.862	17.502	0.644	3.657		0.089
Gansu	1.613	9.862	17.502	0.644	3.657		0.089
Fujian	1.114	14.539	20.197	0.591	3.355		0.049
Macao	1.114	14.539	20.197	0.591	3.355		0.049
Hong Kong	1.114	14.539	20.197	0.591	3.355		0.049

EFs: kgNH<sub>3</sub> head<sup>-1</sup>month<sup>-1</sup>

Province	Pigs				Cattle			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
Beijing	0.109	0.140	0.134	0.103	0.515	0.662	0.632	0.488
Tianjin	0.109	0.140	0.134	0.103	0.515	0.662	0.632	0.488
Hebei	0.109	0.140	0.134	0.103	0.515	0.662	0.632	0.488
Shanxi	0.109	0.140	0.134	0.103	0.515	0.662	0.632	0.488
Inner Mongolia	0.109	0.140	0.134	0.103	0.515	0.662	0.632	0.488
Liaoning	0.189	0.243	0.232	0.179	1.067	1.373	1.311	1.011
Jilin	0.189	0.243	0.232	0.179	1.067	1.373	1.311	1.011
Heilongjiang	0.189	0.243	0.232	0.179	1.067	1.373	1.311	1.011
Shanghai	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Jiangsu	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Zhejiang	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Anhui	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Taiwan	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Jiangxi	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Shandong	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Henan	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Hubei	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Hunan	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Guangdong	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Guangxi	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Hainan	0.147	0.189	0.180	0.139	0.467	0.600	0.573	0.442
Sichuan	0.065	0.083	0.079	0.061	0.737	0.948	0.905	0.698
Yunnan	0.065	0.083	0.079	0.061	0.737	0.948	0.905	0.698
Guizhou	0.065	0.083	0.079	0.061	0.737	0.948	0.905	0.698
Chongqing	0.065	0.083	0.079	0.061	0.737	0.948	0.905	0.698
Tibet	0.065	0.083	0.079	0.061	0.737	0.948	0.905	0.698
Ningxia	0.121	0.155	0.148	0.114	0.737	0.948	0.905	0.698
Xinjiang	0.121	0.155	0.148	0.114	0.737	0.948	0.905	0.698
Qinghai	0.121	0.155	0.148	0.114	0.737	0.948	0.905	0.698
Shannxi	0.121	0.155	0.148	0.114	0.737	0.948	0.905	0.698
Gansu	0.121	0.155	0.148	0.114	0.737	0.948	0.905	0.698

Fujian	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Macao	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029
Hong Kong	0.083	0.107	0.102	0.079	1.086	1.397	1.334	1.029

EFs: kgNH<sub>3</sub> head<sup>-1</sup>month<sup>-1</sup>

Province	Dairy cattle				Sheep			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
Beijing	1.929	2.481	2.369	1.827	0.048	0.061	0.058	0.045
Tianjin	1.929	2.481	2.369	1.827	0.048	0.061	0.058	0.045
Hebei	1.929	2.481	2.369	1.827	0.048	0.061	0.058	0.045
Shanxi	1.929	2.481	2.369	1.827	0.048	0.061	0.058	0.045
Inner Mongolia	1.929	2.481	2.369	1.827	0.048	0.061	0.058	0.045
Liaoning	1.813	2.332	2.227	1.717	0.061	0.078	0.075	0.058
Jilin	1.813	2.332	2.227	1.717	0.061	0.078	0.075	0.058
Heilongjiang	1.813	2.332	2.227	1.717	0.061	0.078	0.075	0.058
Shanghai	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Jiangsu	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Zhejiang	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Anhui	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Taiwan	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Jiangxi	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Shandong	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Henan	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Hubei	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Hunan	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Guangdong	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Guangxi	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Hainan	2.486	3.198	3.053	2.354	0.052	0.068	0.064	0.050
Sichuan	1.509	1.941	1.853	1.429	0.053	0.068	0.065	0.050
Yunnan	1.509	1.941	1.853	1.429	0.053	0.068	0.065	0.050
Guizhou	1.509	1.941	1.853	1.429	0.053	0.068	0.065	0.050
Chongqing	1.509	1.941	1.853	1.429	0.053	0.068	0.065	0.050
Tibet	1.509	1.941	1.853	1.429	0.053	0.068	0.065	0.050
Ningxia	1.307	1.682	1.606	1.238	0.048	0.062	0.059	0.046
Xinjiang	1.307	1.682	1.606	1.238	0.048	0.062	0.059	0.046
Qinghai	1.307	1.682	1.606	1.238	0.048	0.062	0.059	0.046
Shannxi	1.307	1.682	1.606	1.238	0.048	0.062	0.059	0.046
Gansu	1.307	1.682	1.606	1.238	0.048	0.062	0.059	0.046
Fujian	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Macao	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042
Hong Kong	1.509	1.941	1.853	1.429	0.044	0.057	0.054	0.042

EFs: kgNH<sub>3</sub> head<sup>-1</sup>month<sup>-1</sup>

Province	Horses/Donkeys/Mules				Poultry			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
Beijing	0.270	0.347	0.331	0.256	0.005	0.006	0.006	0.004

Tianjin	0.270	0.347	0.331	0.256	0.005	0.006	0.006	0.004
Hebei	0.270	0.347	0.331	0.256	0.005	0.006	0.006	0.004
Shanxi	0.270	0.347	0.331	0.256	0.005	0.006	0.006	0.004
Inner Mongolia	0.270	0.347	0.331	0.256	0.005	0.006	0.006	0.004
Liaoning	0.346	0.445	0.424	0.327	0.007	0.009	0.008	0.006
Jilin	0.346	0.445	0.424	0.327	0.007	0.009	0.008	0.006
Heilongjiang	0.346	0.445	0.424	0.327	0.007	0.009	0.008	0.006
Shanghai	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Jiangsu	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Zhejiang	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Anhui	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Taiwan	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Jiangxi	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Shandong	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Henan	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Hubei	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Hunan	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Guangdong	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Guangxi	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Hainan	0.298	0.383	0.366	0.282	0.003	0.003	0.003	0.002
Sichuan	0.302	0.388	0.371	0.286	0.003	0.003	0.003	0.002
Yunnan	0.302	0.388	0.371	0.286	0.003	0.003	0.003	0.002
Guizhou	0.302	0.388	0.371	0.286	0.003	0.003	0.003	0.002
Chongqing	0.302	0.388	0.371	0.286	0.003	0.003	0.003	0.002
Tibet	0.302	0.388	0.371	0.286	0.003	0.003	0.003	0.002
Ningxia	0.273	0.351	0.336	0.259	0.007	0.009	0.008	0.006
Xinjiang	0.273	0.351	0.336	0.259	0.007	0.009	0.008	0.006
Qinghai	0.273	0.351	0.336	0.259	0.007	0.009	0.008	0.006
Shannxi	0.273	0.351	0.336	0.259	0.007	0.009	0.008	0.006
Gansu	0.273	0.351	0.336	0.259	0.007	0.009	0.008	0.006
Fujian	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Macao	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
Hong Kong	0.251	0.322	0.308	0.237	0.004	0.005	0.005	0.003
EFs			kgNH <sub>3</sub>	kg <sup>-1</sup> N <sup>-1</sup>				
Straw returning <sup>a</sup>			0.158					

a Values are derived from Wang et al. (2013) and Zhou et al. (2014)

Note: NH<sub>3</sub> EFs of synthetic fertilizer application, rural excrement and cake fertilizer could be found in Xu et al.<sup>1</sup> Mar, Apr and May (Spring), Jun, Jul and Aug (Summer), Sep, Oct and Nov (Autumn) and Dec, Jan and Feb (Winter).

Table S2 N volatilization rate of animal housing, manure storage and manure spreading

Categories	animal housing <sup>b</sup>	manure storage <sup>b</sup>	manure spreading <sup>b</sup>
------------	-----------------------------	-----------------------------	-------------------------------

Pigs	19.8%	13.6%	26.2%
Cattle	10.9%	10.2%	26.7%
Dairy cattle	11.4%	10.2%	26.7%
Sheep	5.6%	0.0%	27.8%
Horses/Donkeys/Mules	8.9%	0.0%	12.0%
Poultry	27.2%	3.8%	26.9%

b Values are derived from Yang (2008), Pain et al. (1998) and Webb (2001).

Table S3 Coefficients of variations (CVs) for each type of activity data and selected parameters

Emission sources	Parameter description	Distribution	CV
Livestock manure	N volatilization rate of animal housing	normal	50% <sup>c</sup>
	N volatilization rate of manure storage	normal	50% <sup>c</sup>
	N volatilization rate of manure spreading	normal	50% <sup>c</sup>
Straw returning	Emission Factors	normal	50% <sup>c</sup>
	Other beans	uniform	20% <sup>d</sup>
	Other tubers	uniform	20% <sup>d</sup>

c Values are derived from Beusen et al. (2008).

Note: The other CVs for each type of activity data and selected parameters could be found in Xu et al. (2015) and Zhou et al. (2014).

### Section S3: Supplement results

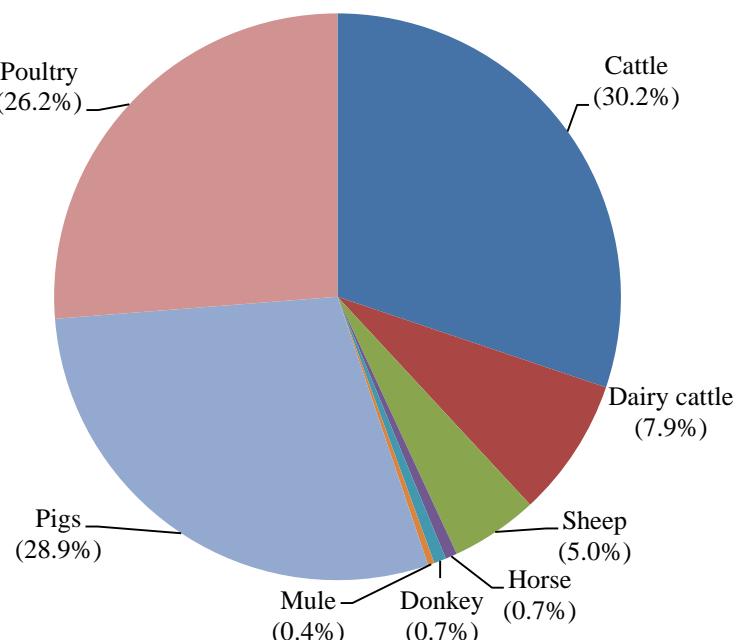


Figure S1 Relative share of animal types in the total ammonia emissions from livestock of China in 2008.



Figure S2 Administrative divisions (regions and provinces) of China.

Note: The Administrative divisions (regions and provinces) of China are shown in Figure S2 ([http://en.wikipedia.org/wiki/Administrative\\_divisions\\_of\\_China](http://en.wikipedia.org/wiki/Administrative_divisions_of_China) and <http://www.data.ac.cn/>).

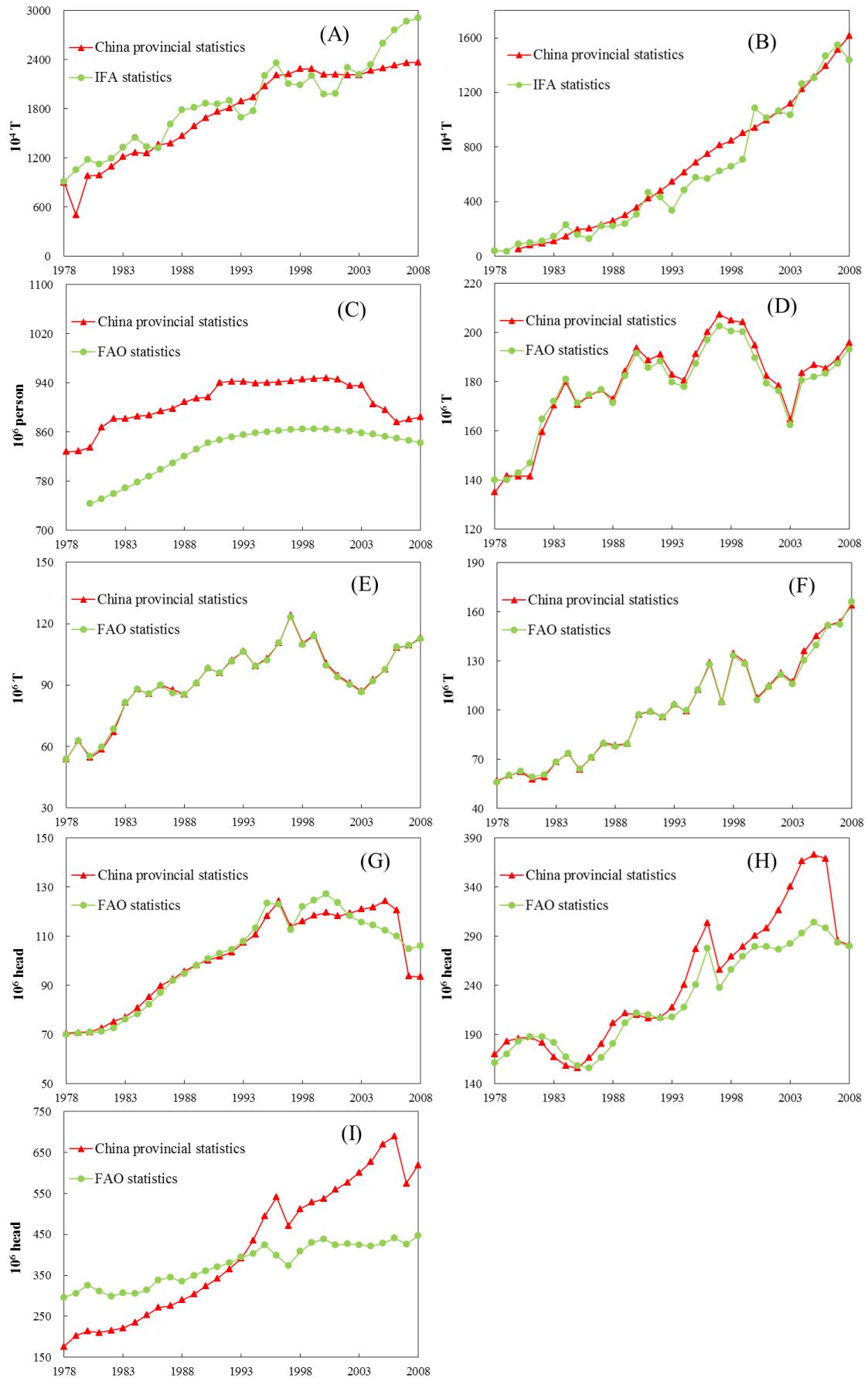
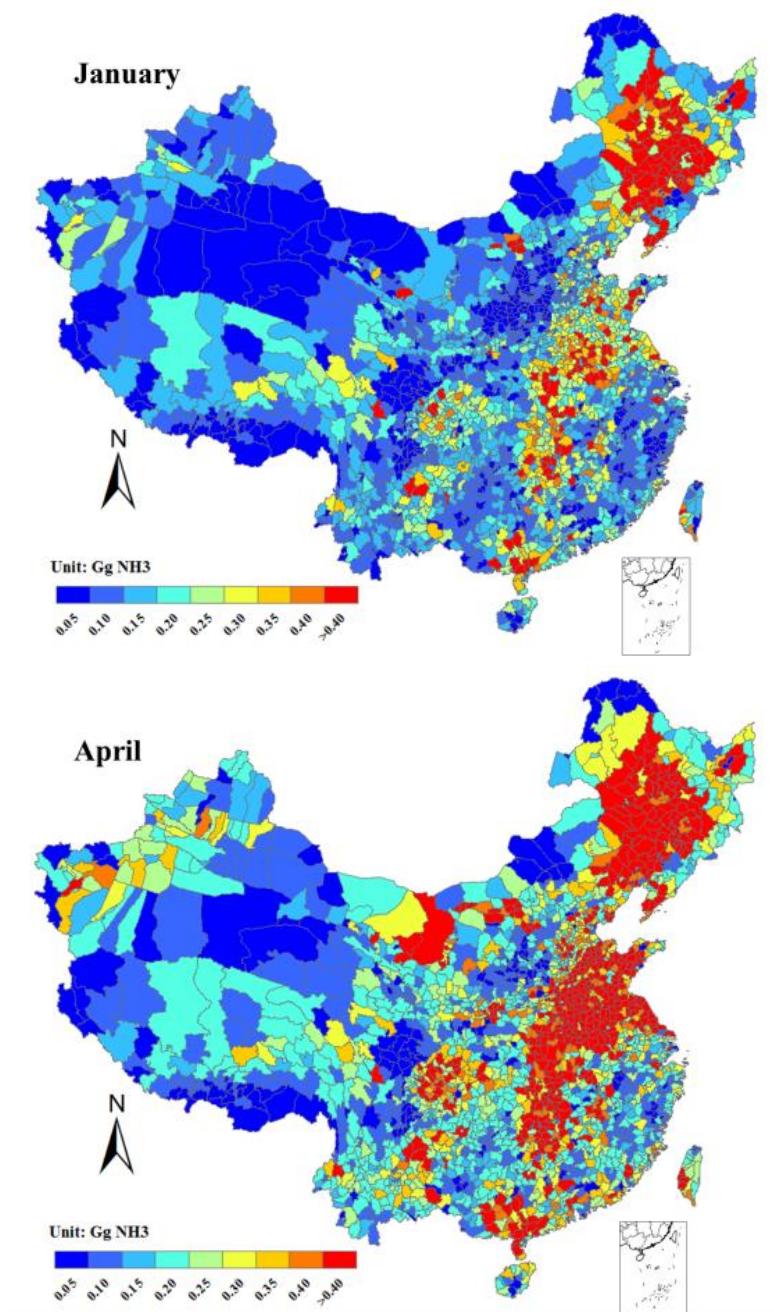


Figure S3 Comparison of nitrogen fertilizer (A), compound fertilizer (B), rural population (C), rice (D), wheat (E), maize (F), cattle (G), sheep (H) and pigs (I) activity data for a period from 1978 to 2007 which are the major  $\text{NH}_3$  emission sources in this study from NBSC (2009a and 2009b) provincial statistics (sums of the provincial data), IFA and FAO (national data) (<http://www.fertilizer.org/> and <http://faostat.fao.org/>).



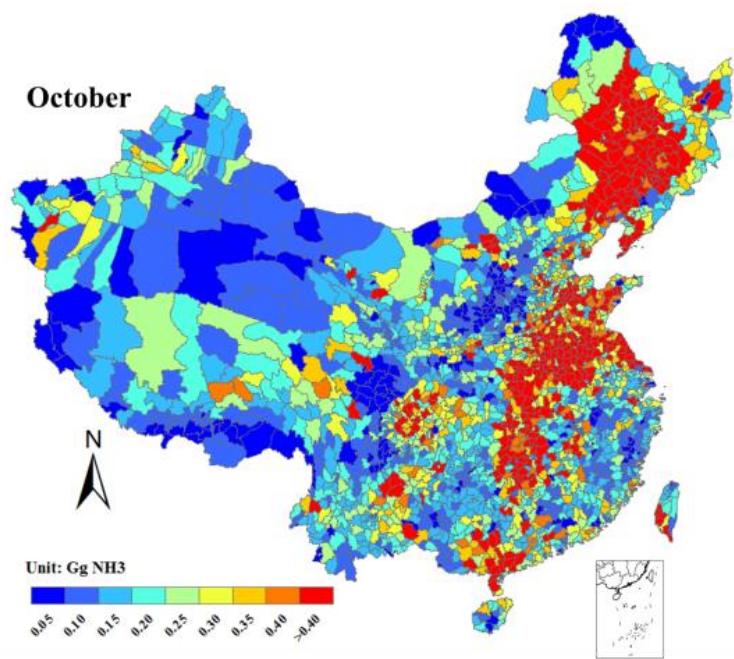
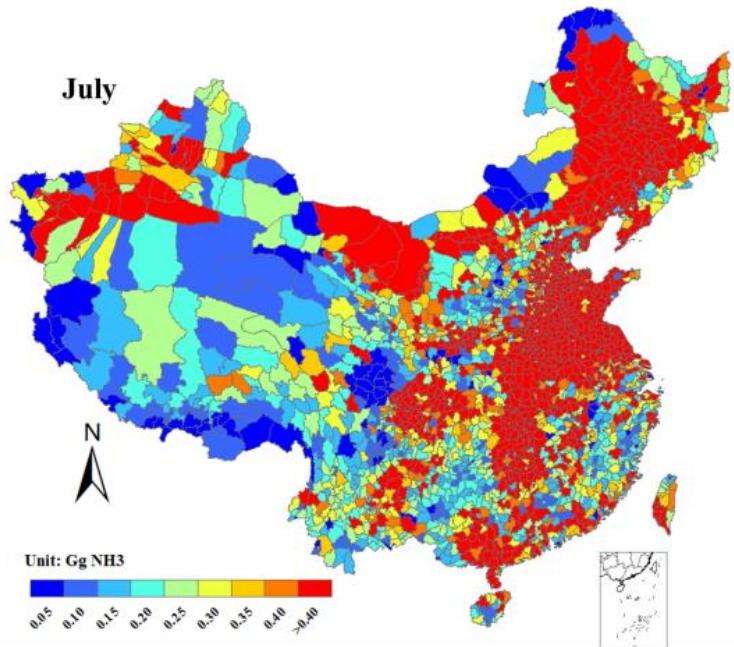
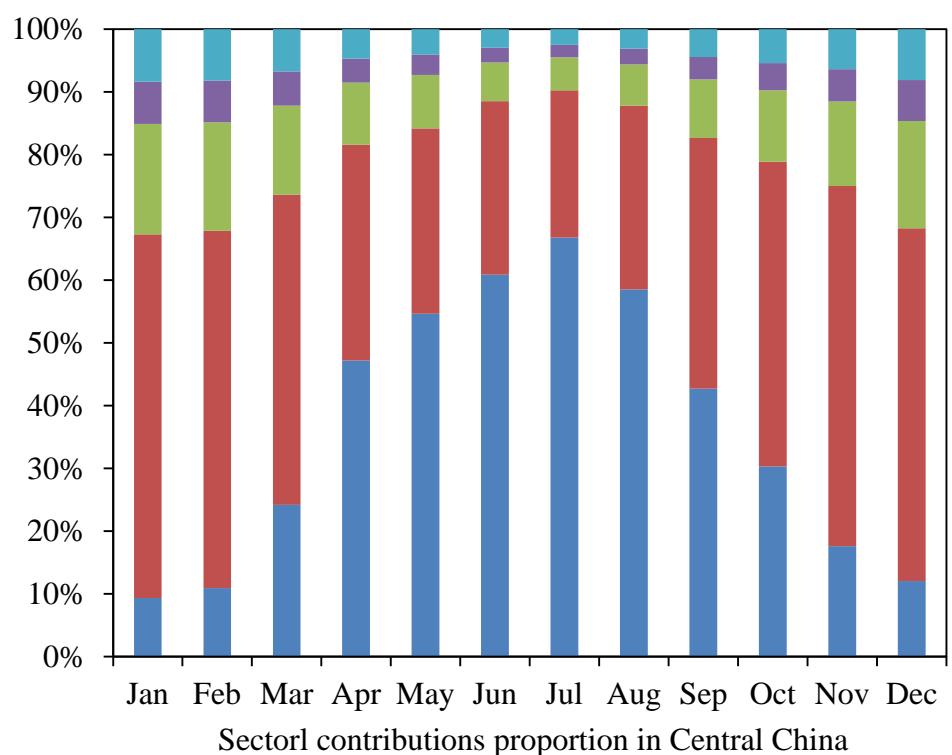
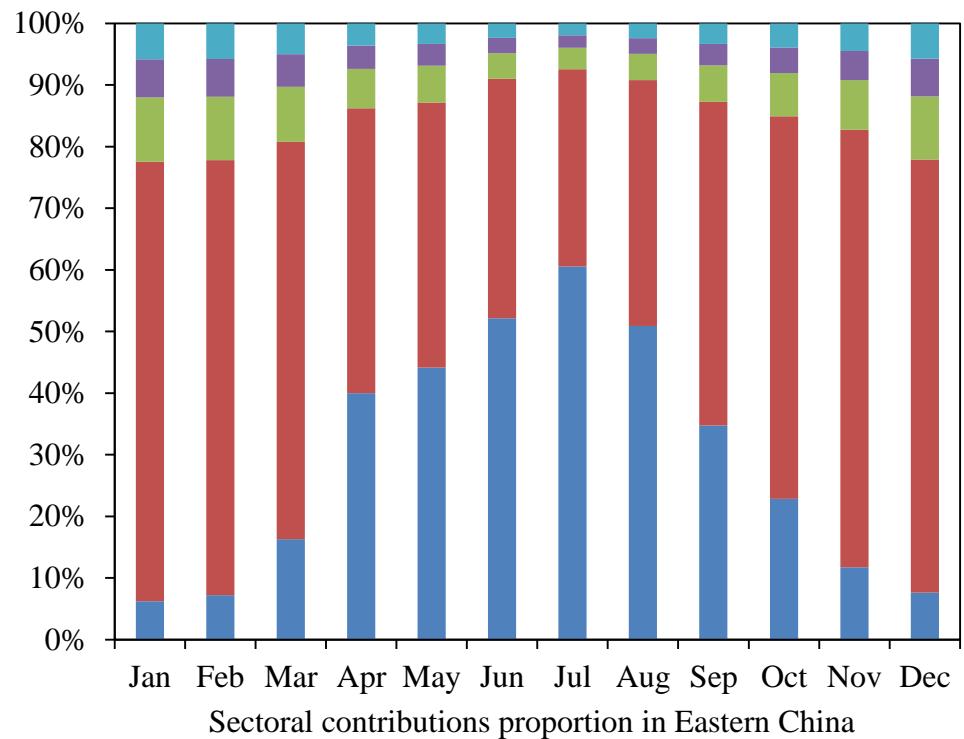


Figure S4 The spatial distributions of the agricultural fertilizer  $\text{NH}_3$  emissions for January, April, July and October.



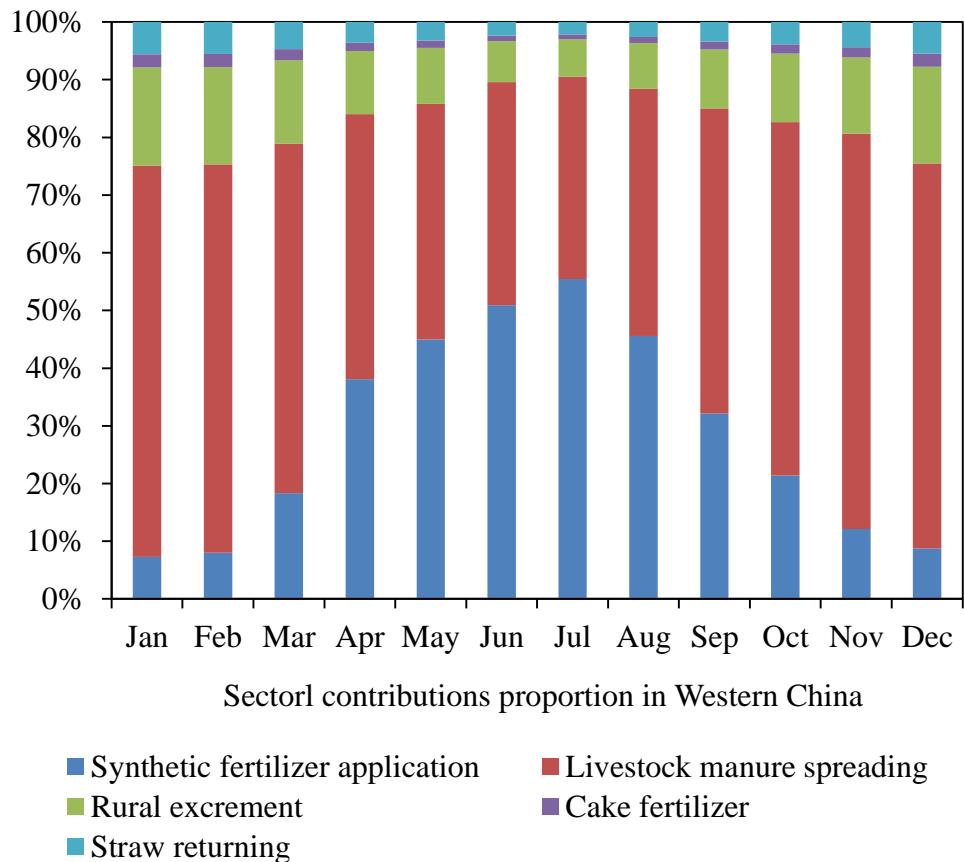
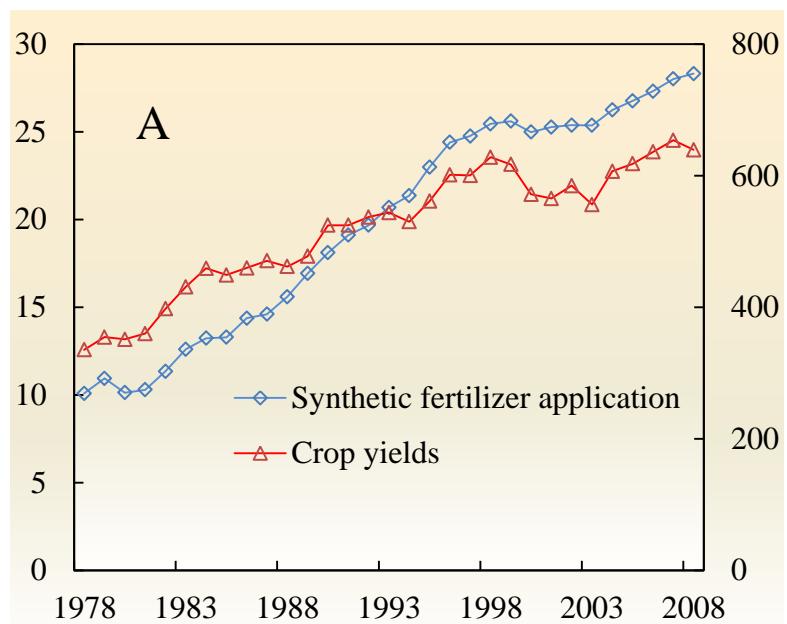


Figure S5 Sectoral contributions proportion of monthly ammonia emissions in different administrative divisions of China in 2008

Table S4 Sectoral contributions proportion of Livestock manure spreading

	Cattle	Dairy cattle	Sheep	Horses	Donkeys	Mules	Pigs	Poultry
1978	0.581	0.015	0.096	0.038	0.024	0.013	0.215	0.019
1979	0.558	0.011	0.100	0.037	0.023	0.013	0.239	0.019
1980	0.552	0.013	0.100	0.036	0.024	0.013	0.244	0.019
1981	0.558	0.014	0.100	0.035	0.025	0.013	0.236	0.020
1982	0.561	0.016	0.094	0.034	0.026	0.013	0.236	0.020
1983	0.564	0.018	0.085	0.033	0.027	0.013	0.238	0.023
1984	0.561	0.023	0.077	0.032	0.027	0.013	0.240	0.027
1985	0.554	0.026	0.071	0.030	0.027	0.013	0.242	0.039
1986	0.553	0.027	0.071	0.028	0.026	0.013	0.246	0.036
1987	0.545	0.031	0.074	0.026	0.025	0.012	0.239	0.048
1988	0.536	0.030	0.078	0.025	0.024	0.012	0.237	0.057
1989	0.532	0.033	0.079	0.023	0.024	0.012	0.241	0.057
1990	0.523	0.034	0.075	0.022	0.023	0.011	0.249	0.063
1991	0.510	0.035	0.071	0.021	0.022	0.011	0.253	0.077
1992	0.499	0.036	0.068	0.020	0.021	0.011	0.262	0.084
1993	0.487	0.037	0.067	0.019	0.019	0.010	0.264	0.097
1994	0.447	0.070	0.066	0.017	0.017	0.009	0.263	0.111

1995	0.431	0.069	0.068	0.015	0.015	0.008	0.272	0.122
1996	0.421	0.065	0.069	0.014	0.014	0.007	0.280	0.130
1997	0.416	0.067	0.064	0.013	0.014	0.007	0.266	0.153
1998	0.417	0.067	0.066	0.013	0.013	0.007	0.281	0.135
1999	0.412	0.067	0.066	0.013	0.013	0.006	0.283	0.141
2000	0.406	0.069	0.067	0.012	0.012	0.006	0.280	0.148
2001	0.395	0.075	0.068	0.011	0.011	0.006	0.286	0.148
2002	0.388	0.083	0.070	0.011	0.011	0.005	0.285	0.147
2003	0.374	0.095	0.072	0.010	0.010	0.005	0.285	0.149
2004	0.355	0.107	0.073	0.009	0.009	0.004	0.283	0.161
2005	0.350	0.112	0.072	0.009	0.009	0.004	0.293	0.151
2006	0.334	0.122	0.071	0.008	0.008	0.004	0.299	0.154
2007	0.325	0.101	0.068	0.010	0.009	0.004	0.307	0.176
2008	0.312	0.099	0.065	0.009	0.009	0.004	0.322	0.180



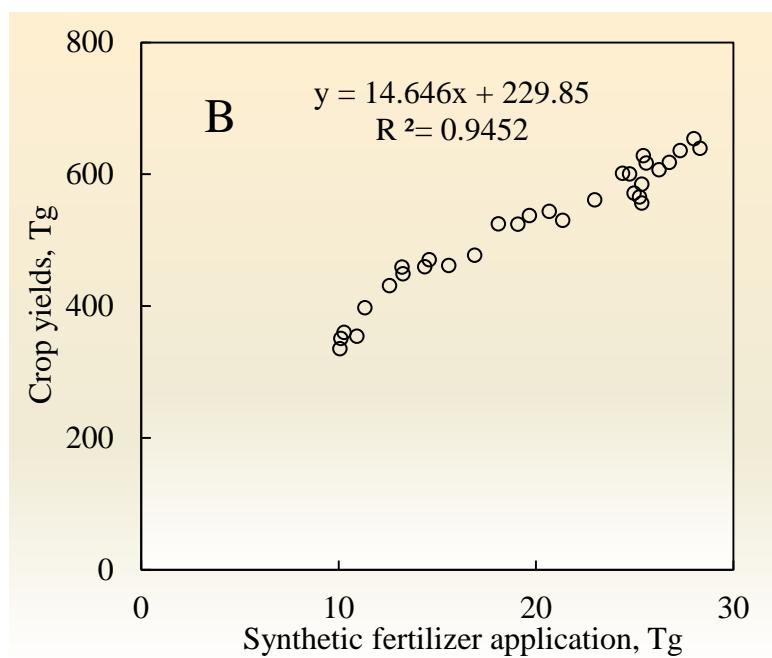


Figure S6 Synthetic fertilizer application and crop yields during the study period (A): Amount; (B): Correlation analysis.

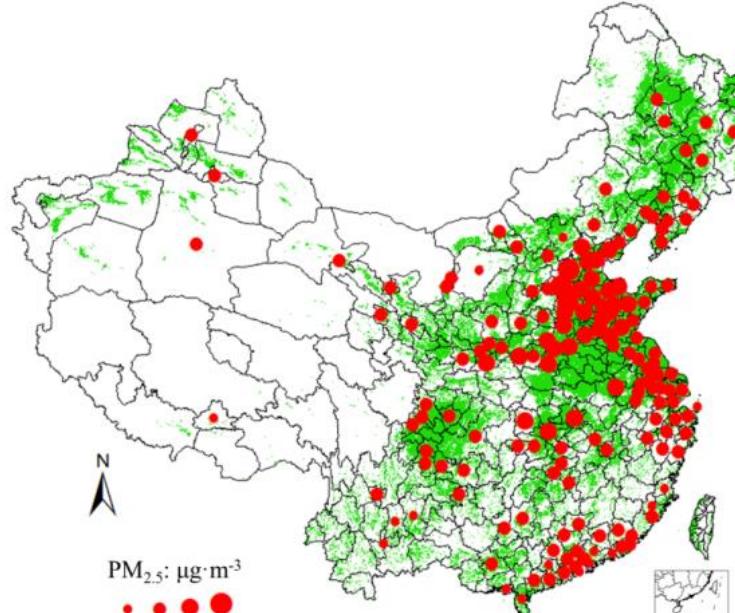


Figure S7 Spatial distribution of PM<sub>2.5</sub> concentration and agricultural areas. The data of PM<sub>2.5</sub> concentration is derived from (<http://news.163.com/15/0121/20/AGGQM0F400014SEH.html>).

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