Supplement of

High resolution inventory of re-estimating ammonia emissions from agricultural fertilizer in China from 1978 to 2008

P. Xu et al.

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9 **Section S1: Methodology**

10

11 Text S1: Equations for ammonia (NH_3) emissions from each source

12   The equations for NH_3 emissions from synthetic fertilizer, rural excrement and cake fertilizer

13 were found in Xu et al. (2015). The equations for livestock manure spreading and straw returning

14 are as follows:

15 1．Livestock manure spreading

16
$$E_{LM} = \frac{17}{14} \cdot \sum_{L} \sum_{R} LN_{(L,R)} \cdot EF_{(L,R)} \qquad (1)$$

17 where

18
$$EF_{(L,R)} = LM_{(L,R)} \cdot (1 - v_{(H)}) \cdot (1 - v_{(ST)}) \cdot v_{(MS)} \qquad (2)$$

19
$$LM_{(L,R)} = LN_{(L,R)} \cdot N_{er(L,R)} \cdot FD_{(L)} / 1000 \qquad (3)$$

20 and E_{LM} is the NH_3 emission from livestock manure spreading in region R, $TgNH_3 \ yr^{-1}$; $LN_{(L)}$ is

21 the number of head of species/category L in region R; $EF_{(L)}$ is the emission factor for NH_3

22 emissions from livestock manure spreading for livestock species/category L in region R, kg

23 $NH_3\text{-N·kg}^{-1}$; and $LM_{(L)}$ is the annual average N excretion in manure management for livestock

24 species/category L in region R, kg $N·yr^{-1}$. The livestock species/category considered in this study

25 included pigs, beef cattle, dairy cattle, sheep, poultry, horses, donkeys and mules. $v_{(H)}$, $v_{(ST)}$ and

26 $v_{(MS)}$ are the N volatilization rates of animal housing, manure storage and manure spreading,

27 respectively. $N_{er(L)}$ is the N excretion rate for livestock species/category L in region R, g

28 $N·head^{-1}·day^{-1}$. $FD_{(L)}$ is the number of feeding days for livestock species/category L, and 17/14 is

29 the conversion coefficient of NH_3-N emissions to NH_3 emissions (Zhou et al., 2014; Yang, 2008).

30   2. Straw returning

31

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

32   where E_{CR} is the NH$_3$ emission from straw returning in region R, TgNH$_3$ yr^{-1}; $NSR_{(i,R)}$ is the annual

33   nitrogen quantity of straw returning for crop i in region R, kgN· kg^{-1}yr^{-1}; and EF_{CR} is the emission

34   factor for NH$_3$ emission from straw returning for crop types i in region R, kg NH$_3$ ·kg^{-1}·N^{-1}. The

35   crop types considered in this study were maize, rice, wheat, barley, sugarcane, millet, potatoes,

36   other tubers, soybeans, peanuts, rapeseed, sesame, other oil crops, sorghum, highland barley, other

37   beans and alfalfa. $NSR_{(i,R)}$ was derived from Zhou et al. (2014)

Section S2: Data on activity data, RPs and EFs

39

40   Table S1 NH$_3$ EFs of livestock manure spreading for 34 provinces in China and of straw returning

41

| | EFs: kgNH$_3$ head^{-1}yr^{-1} | | | | | |
Province	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 Mongalia	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

Province							
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: $kgNH_3\ head^{-1}month^{-1}$

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 Mongalia	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: $kgNH_3\ head^{-1}month^{-1}$ | | | | | | | |
| | Dairy cattle | | | | Sheep | | | |
Province	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 Mongalia	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: $kgNH_3\ head^{-1}month^{-1}$ | | | | | | | |
| | Horses/Donkeys/Mules | | | | Poultry | | | |
Province	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 Mongalia	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	$kgNH_3\ kg^{-1}N^{-1}$							
Straw returning [a]	0.158							

42

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

44   Note: NH_3 EFs of synthetic fertilizer application, rural excrement and cake fertilizer could be

45   found in Xu et al.;[1] Mar, Apr and May (Spring), Jun, Jul and Aug (Summer), Sep, Oct and Nov

46   (Autumn) and Dec, Jan and Feb (Winter).

47

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

49

Categories	animal housing [b]	manure storage [b]	manure spreading [b]
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%[c]
	N volatilization rate of manure storage	normal	50%[c]
	N volatilization rate of manure spreading	normal	50%[c]
Straw returning	Emission Factors	normal	50%[c]
	Other beans	uniform	20%[d]
	Other tubers	uniform	20%[d]

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
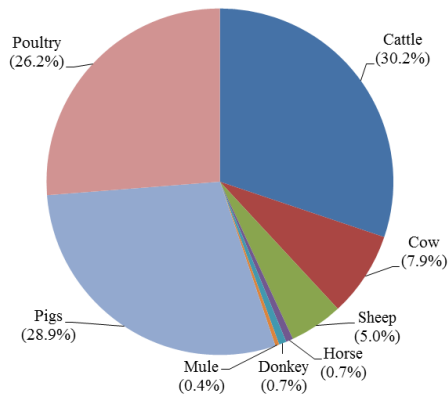
Figure S1 Relative share of animal types in the total ammonia emissions from livestock of China in 2008.
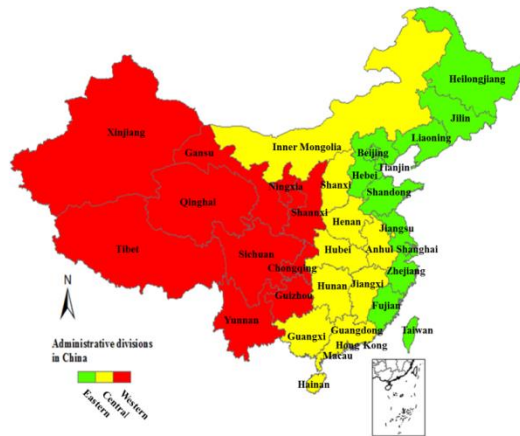
66
67   Figure S2 Administrative divisions (regions and provinces) of China.
68   Note: The Administrative divisions (regions and provinces) of China are shown in Figure S2
69   (http://en.wikipedia.org/wiki/Administrative_divisions_of_China and http://www.data.ac.cn/).
70
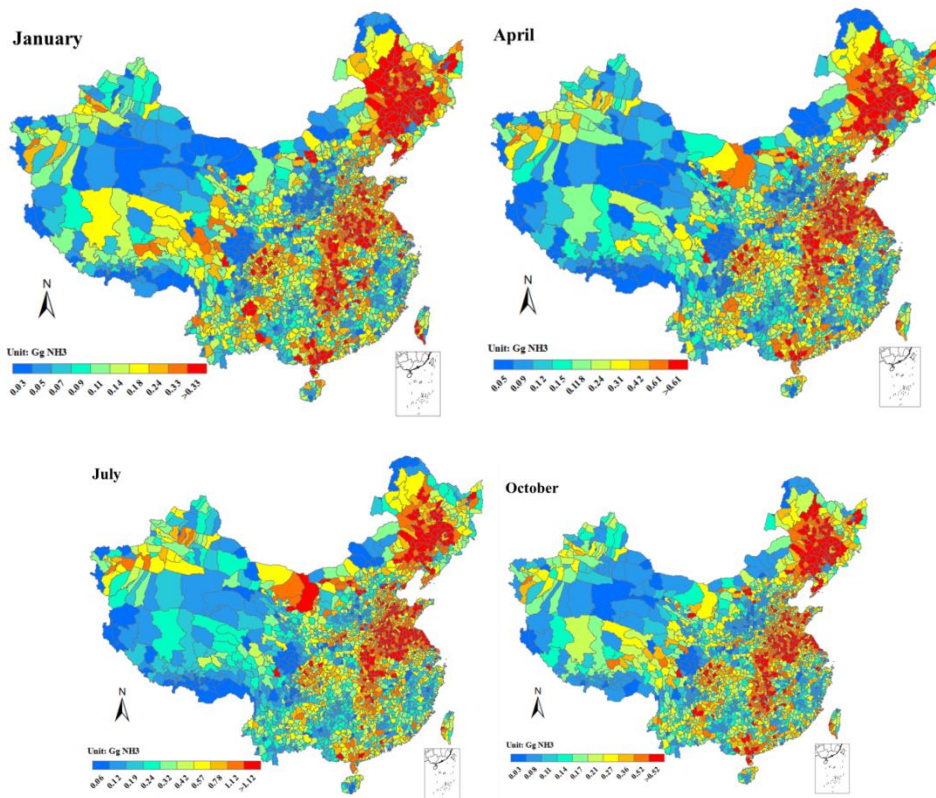
71

72
73   Figure S3 The spatial distributions of the agricultural fertilizer NH$_3$ emissions for January, April,
74   July and October.
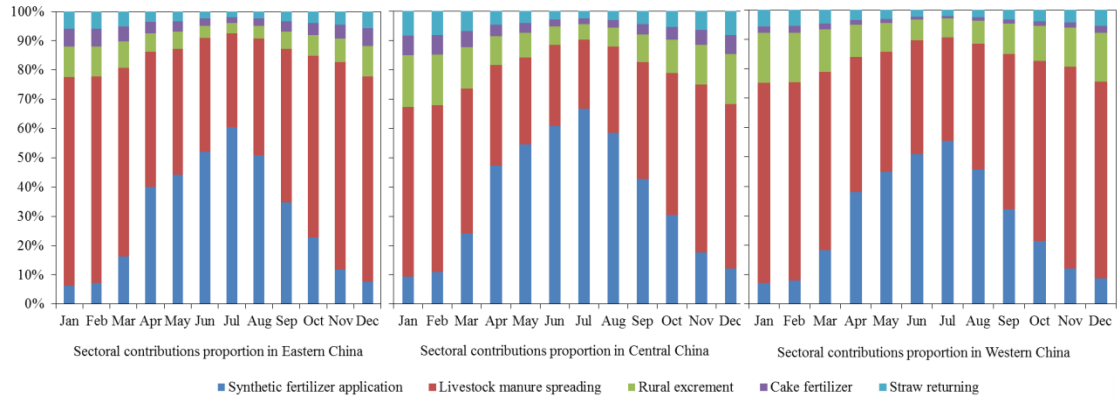
Figure S4 Sectoral contributions proportion of monthly ammonia emissions in different administrative divisions of China in 2008

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
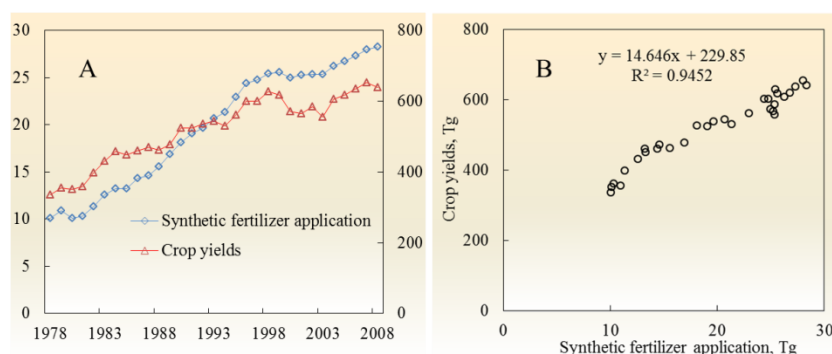
Figure S5 Synthetic fertilizer application and crop yields during the study period (A): Amount; (B): Correlation analysis.
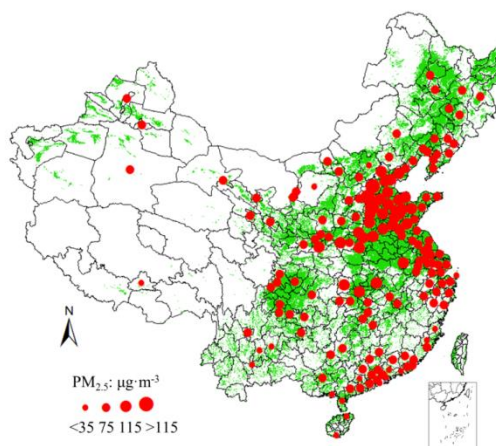
Figure S6 Spatial distribution of PM$_{2.5}$ concentration and agricultural areas. The data of PM$_{2.5}$ concentration is derived from (http://news.163.com/15/0121/20/AGGQM0F400014SEH.html).

Section S4: Supporting references

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