

Supplement of

Quantification and evaluation of atmospheric ammonia emissions with different methods: A case study for the Yangtze River Delta region, China

Yu Zhao, Mengchen Yuan, Xin Huang, Feng Chen, Jie Zhang

*Corresponding author: Yu Zhao (yuzhao@nju.edu.cn)

Number of tables: 8

Number of figures: 4

Table list

Table S1. The annual numbers of livestock and poultry by prefectural city in YRD (10 000)

Table S2. Fertilizer consumption by type and prefectural city in the YRD region, and the provincial-level total consumption from statistics (metric tons).

Table S3. The NH₃ emission factors used in E1.

Table S4. Relevant parameters used for correction of emission factors of fertilizer activity in E2.

Table S5. Parameters used in estimates of annual TAN excretion per cattle.

Table S6. The temperature-dependant emission factors by stage/phase in livestock farming in E2.

Table S7. Model performance for meteorological parameters in D2.

Table S8. Inter-annual change in SO₂ and NO₂ VCDs for the YRD region 2012-2014 (%)

Figure list

Figure S1. Corrected NH₃ volatilization of urea and ABC application in E2.

Figure S2. Total ammoniacal nitrogen (TAN) for three main raising systems (taken from Huang et al., 2012).

Figure S3. Daily NH₃ concentration at JS-PAES for October 2014 from observation and simulation with E1 and E2.

Figure S4. The NH₃ volatilization rates under different soil pH values for urea (a) and ABC fertilizer (b). The blue dots indicate the values by grid in the YRD region in E2, while the red dots indicate the results from the field measurements by Zhong et al. (2006) (a) and Zhang et al. (2002) (b). The solid lines and equations were obtained from linear regression for the gridded values in the YRD region in E2.

Tables

Table S1. The annual numbers of livestock and poultry by prefectural city in YRD (10 000)

	Beef	Cow	Sow	Hog	Goat	Sheep	Layer	Laying Duck	Broiler	Meat duck	Goose	Horse	Donkey	Mule	Rabbit	Cattle	Buffalo
Shanghai	0.10	5.80	13.90	243.13	26.70	1.40	379.05	37.61	23.81	1051.68	140.00	—	—	—	9.60	—	—
Nanjing	—	1.75	—	—	11.73	—	491.68	119.51	12.57	562.96	108.92	—	—	—	17.49	0.01	1.08
Wuxi	—	0.67	—	74.77	3.67	0.09	174.87	42.51	6.31	282.49	54.65	—	—	—	2.23	—	—
Xuzhou	—	—	—	—	218.91	8.63	3783.84	919.73	96.01	4301.53	832.24	0.13	1.59	0.87	—	—	—
Changzhou	0.01	0.54	49.10	83.62	6.05	0.14	166.90	40.57	18.98	850.37	164.53	—	—	—	—	—	—
Suzhou	—	2.39	76.69	103.46	4.36	6.37	278.52	67.70	9.82	440.12	85.15	—	—	—	9.32	—	—
Nantong	—	0.75	270.76	394.71	227.33	—	2984.22	725.37	47.46	2126.22	411.37	—	—	—	—	—	0.32
Lianyungang	10.89	6.36	20.03	300.50	18.04	—	612.70	148.93	13.86	620.76	120.10	—	0.49	—	—	—	—
Huai'an	2.21	0.82	15.68	267.11	23.29	—	849.60	206.51	29.18	1307.15	252.90	—	—	—	—	—	3.06
Yancheng	—	1.97	41.79	735.62	138.68	—	7591.19	486.72	81.09	3633.09	702.91	—	0.02	—	228.00	1.38	1.10
Yangzhou	0.41	0.47	5.12	135.72	6.26	—	651.50	391.05	19.44	870.89	168.49	—	—	—	11.09	—	—
Zhenjiang	—	—	40.58	42.30	4.17	—	164.29	39.93	8.07	361.71	69.98	—	—	—	—	—	—
Taizhou	0.35	0.97	17.77	297.09	15.82	—	7.40	1.48	12.90	578.07	111.84	—	—	—	—	—	—
Suqian	5.92	7.77	146.14	260.75	29.67	—	923.53	224.48	26.05	1167.05	225.79	—	—	—	80.56	—	—
Hangzhou	1.23	0.74	15.45	333.26	23.12	—	959.79	102.66	17.97	804.96	155.74	—	—	—	86.57	—	—
Ningbo	0.71	0.80	9.56	165.95	—	—	537.08	57.45	10.54	472.28	91.38	—	—	—	62.00	—	—
Wenzhou	2.47	0.74	6.55	112.60	14.16	—	318.72	34.09	8.65	387.40	74.95	—	—	—	131.64	—	—
Jiaxing	—	—	6.65	374.83	—	58.91	429.45	45.93	22.84	1023.31	197.99	—	—	—	—	—	—
Huzhou	0.26	—	8.34	131.62	35.22	—	421.54	45.09	27.78	1244.38	240.76	—	—	—	61.35	—	—
Shaoxing	0.93	0.11	12.13	193.84	11.06	—	306.56	32.79	10.05	450.24	87.11	—	—	—	50.23	—	—

Continued Table S1

	Beef	Cow	Sow	Hog	Goat	Sheep	Layer	Laying duck	Broiler	Meat duck	Goose	Horse	Donkey	Mule	Rabbit	Cattle	Buffalo
Jinhua	2.46	1.96	11.96	265.22	8.27	—	373.58	39.96	10.31	461.69	89.33	—	—	—	10.24	—	—
Zhoushan	0.03	—	—	22.32	—	—	—	—	0.93	41.66	8.06	—	—	—	6.10	—	—
Taizhou	—	—	5.80	92.15	6.40	—	244.76	26.18	10.94	489.99	94.80	—	—	—	58.17	—	—
Lishui	—	—	4.55	86.66	7.92	—	89.22	9.54	40.32	1806.19	349.45	—	—	—	11.85	—	—
Quzhou	1.36	—	17.23	400.45	4.53	—	163.31	17.47	13.13	588.25	113.81	—	—	—	—	—	—
Hefei	5.07	3.59	15.25	285.48	8.54	0.45	861.96	209.46	75.69	3390.88	656.05	—	—	—	3.76	—	—
Huaibei	3.05	0.84	—	68.23	100.15	0.65	580.11	140.97	8.17	365.95	70.80	0.004	0.06	0.002	21.37	—	—
Anqing	7.09	—	17.89	284.97	7.69	—	1631.00	444.51	29.94	—	—	—	—	—	2.00	—	—
Boshou	13.53	—	—	304.66	126.99	—	438.70	106.60	11.57	518.27	100.27	0.04	0.01	0.03	—	—	—
Suzhou	15.76	—	—	483.34	241.66	3.26	1657.91	402.88	23.10	1034.75	200.20	0.005	0.01	0.003	—	—	—
Bengbu	26.67	—	—	202.87	73.70	0.47	470.98	114.45	31.43	1408.07	272.43	—	—	—	—	—	—
Fuyang	31.60	—	—	560.37	138.92	—	909.21	220.94	26.42	1183.72	229.02	0.02	0.12	0.02	—	—	—
Huainan	5.05	—	—	50.67	13.06	—	427.94	103.99	11.30	506.30	97.96	—	—	—	—	—	—
Chuzhou	12.72	—	—	335.62	37.21	0.23	721.84	175.41	28.99	1298.97	251.32	—	—	—	—	—	—
Liu'an	11.37	—	—	405.70	46.48	—	774.29	188.16	56.55	2533.44	490.16	—	—	—	—	—	—
Ma'anshan	0.20	—	—	37.06	6.72	—	132.64	32.23	15.78	707.14	136.81	—	—	—	—	—	—
Wuhu	2.70	—	—	83.15	2.92	—	516.96	125.62	25.95	1162.41	224.90	—	—	—	—	—	—
Xuancheng	2.30	—	—	103.72	4.66	—	314.63	76.46	43.47	1947.60	376.81	—	—	—	—	—	—
Tongling	0.11	—	—	10.24	—	—	64.32	15.63	—	—	—	—	—	—	—	—	—
Chizhou	1.07	—	—	73.93	1.41	—	237.85	57.80	8.54	382.44	73.99	—	—	—	—	—	—
Huangshan	0.81	—	—	96.38	0.49	0.03	137.06	33.31	2.14	95.73	18.52	0.03	0.003	0.004	—	—	—

Table S2. Fertilizer consumption by type and prefectural city in the YRD region, and the provincial-level total consumption from statistics (metric tons).

	Urea	ABC	AN	AS	DAP	NPK	Other	Total	Statistics
Shanghai	38522.2	1819.4	44.4	6.2	2033.7	62064.8	42.4	104533.1	89000
Nanjing	55146.7	2871.5	91.3	12.7	2645.4	62403.7	87.1	123258.4	
Wuxi	52579.2	3148.5	101.4	14.1	2347.9	65723.9	96.8	124011.7	
Xuzhou	172364.1	10517.6	354.8	49.4	5962.7	158593.2	338.6	348180.3	
Changzhou	34264.2	2074.6	93.4	13.0	1245.9	31052.6	89.2	68832.9	
Suzhou	37217.3	2230.9	57.6	8.0	1687.8	48076.9	55.0	89333.4	
Nantong	148372.9	7212.5	256.6	35.8	6750.4	138864.2	245.0	301737.4	
Lianyungang	97247.4	5461.8	121.6	17.0	3958.0	100200.1	116.1	207121.9	
Huai'an	119622.4	6238.7	170.0	23.7	4602.1	112485.5	162.3	243304.7	
Yancheng	222512.3	9824.1	347.0	48.4	8286.0	236255.7	331.2	477604.6	
Yangzhou	74431.4	3258.3	99.9	13.9	2118.3	73909.4	95.4	153926.5	
Zhenjiang	48497.0	2494.6	83.8	11.7	1828.2	40582.9	80.0	93578.1	
Taizhou	114868.5	5766.9	185.9	25.9	4691.2	106345.4	177.5	232061.3	
Suqian	112064.1	6159.3	162.5	22.6	3864.9	108296.8	155.1	230725.2	
Jiangsu total	1289187.3	67259.1	2125.7	296.2	49988.8	1282790.1	2029.1	2693676.3	2609000
Hangzhou	33929.4	3299.9	30.1	4.2	547.9	58816.2	28.7	96656.4	
Ningbo	32399.1	3521.5	35.6	5.0	831.4	56213.0	34.0	93039.5	
Wenzhou	21741.3	3051.4	1.2	0.2	169.6	33691.6	1.1	58656.3	
Jiaxing	37957.0	4132.5	35.5	5.0	803.5	57789.3	33.9	100756.7	
Huzhou	28860.3	3742.5	26.0	3.6	576.3	35435.9	24.8	68669.4	
Shaoxing	33122.2	3801.7	35.2	4.9	768.2	46923.1	33.6	84688.9	
Jinhua	28634.6	3062.7	30.3	4.2	689.9	39802.2	28.9	72252.8	
Zhoushan	2736.0	238.6	4.1	0.6	84.7	5136.9	4.0	8204.9	
Taizhou	27127.9	2912.9	31.8	4.4	500.1	46903.3	30.4	77510.8	
Lishui	20953.3	2403.5	36.6	5.1	563.7	32309.7	34.9	56306.8	
Quzhou	27943.3	3597.7	21.1	2.9	461.3	33127.1	20.2	65173.5	
Zhejiang total	295404.4	33764.7	287.6	40.1	5996.5	446148.3	274.5	781916.0	717000
Hefei	89980.7	3144.9	32.4	4.5	4567.2	115602.5	31.0	213363.1	
Huaibei	26884.4	678.8	12.2	1.7	897.6	42907.1	11.7	71393.5	

Continued Table S2

Anqing	104403.0	2946.8	34.1	4.7	5869.2	120970.4	32.5	234260.7	
Bozhou	112979.8	2989.4	68.5	9.6	5221.0	182035.0	65.4	303368.7	
Suzhou	104919.7	2576.0	35.6	5.0	3810.2	151179.4	34.0	262559.9	
Bengbu	72865.7	1953.0	36.4	5.1	4875.7	119840.0	34.8	199610.6	
Fuyang	139596.0	3727.8	75.8	10.6	5704.0	223240.0	72.4	372426.6	
Huainan	29430.1	845.1	14.8	2.1	1092.6	48097.9	14.2	79496.8	
Chuzhou	115575.2	2824.0	53.9	7.5	5953.7	176377.9	51.5	300843.7	
Liuan	112162.7	2719.9	36.0	5.0	3482.5	155355.5	34.4	273796.0	
Maanshan	28821.6	721.0	10.6	1.5	1237.6	39867.0	10.1	70669.3	
Wuhu	46288.4	1159.8	17.4	2.4	2845.1	61312.2	16.6	111641.8	
Xuancheng	41839.9	1015.9	15.7	2.2	1652.2	56933.7	15.0	101474.6	
Tongling	5782.1	141.1	1.9	0.3	319.6	7754.7	1.8	14001.5	
Chizhou	26140.2	559.0	4.6	0.6	1588.7	29744.6	4.4	58042.0	
Huangshan	15024.7	364.5	8.3	1.2	594.2	19994.1	7.9	35994.8	
Anhui total	1072694.0	28366.9	458.3	63.9	49711.1	1551211.9	437.5	2702943.6	2731025
`YRD total	2695807.8	131210.1	2916.1	406.4	107730.2	3342215.0	2783.4	6283069.0	6146025

Note: ABC, AN, AS, DAP, and NPK represent ammonium bicarbonate, ammonium nitrate, ammonium sulfate, diammonium phosphate and complex-fertilizer, respectively.

Table S3. The NH₃ emission factors used in E1.

Source	EFs	Unit	Reference	Source	EFs	Unit	Reference
Fertilizer Application				waste incineration	0.21	kg/t	Zheng et al. (2012)
urea	17.4	NH ₃ -N/%	Dong et al.(2009)	waste compost	1.275	kg/t	Zhou et al. (2015)
Ammonium bicarbonate	21.3	NH ₃ -N/%		sewage treatment	0.28	g/m ³	Kang et al. (2016)
ammonium sulfate	2	NH ₃ -N/%		Transportation			
ammonium nitrate	8	NH ₃ -N/%		light duty gasoline vehicle	43.1	mg/km	Huang et al. (2012)
others	4	NH ₃ -N/%		light duty diesel vehicle	4.1	mg/km	Zheng et al. (2012)
diammoniumphosphate	7.3	NH ₃ -N/%	Zhou et al. (2015)	heavy duty gasoline vehicle	28	mg/km	Zhou et al. (2015)
NPK Compound-Fertilizer	5	NH ₃ -N/%	Zhou et al. (2015)	heavy duty diesel vehicle	16.9	mg/km	Kang et al. (2016)
Livestock				motorcycle	7	mg/km	Yin et al. (2010)
beef cattle	19.8	kg/a	Zheng et al. (2012)	Fuel Combustion			
dairy cow	30.53	kg/a	Zhou et al. (2015)	industrial coal combustion	0.02	kg/t	Huang et al. (2012)
sow	14.53	kg/a	Yin et al. (2010)	industrial oil combustion	0.1	kg/m ³	Zheng et al. (2012)
hog	2.87	kg/a	Dong et al. (2010)	industrial gas combustion	51.3	kg/10 ⁶ m ³	Zhou et al. (2015)
goat	5.27	kg/a	Pan et al. (2015)	domestic coal combustion	0.9	kg/t	Kang et al. (2016)
sheep	5.23	kg/a	Shen et al. (2014)	domestic oil combustion	0.12	kg/m ³	Yin et al. (2010)
layer	0.46	kg/a	Liu et al. (2015)	domestic gas combustion	320.51	kg/10 ⁶ m ³	
laying duck	0.35	kg/a	Yang et al. (2008)	Industry Sources			
broiler	0.14	kg/a		ammonium synthesis	1	kg/t	Huang et al. (2012)
duck	0.03	kg/a		nitrogenous fertilizer	5	kg/t	Zheng et al. (2012)
goose	0.23	kg/a		phosphate fertilizer	0.07	kg/t	Zhou et al. (2015)
horse	17.26	kg/a		coking	0.07	kg/t	Kang et al. (2016)
donkey	17.26	kg/a		Human Being			
mule	17.26	kg/a		human breath	3.64	g/(cap·yr)	Yin et al. (2010)

Table S4. Relevant parameters used for correction of emission factors of fertilizer activity in E2.

Fertilizer Categories	pH		T		CF _{rate}	CF _{method}
	pH slope	pH intercept	T _{base}	T slope		
Urea	6.265	-25.029	27.6	0.35	1.18	0.32
ABC	6.147	-14.994	27.6	0.44	1.18	0.32
AN	1.793	-10.45	13	0.06	1.18	0.32
AS	0	0.8	0	0.01	1.18	0.32
Other nitrogen fertilizers	0	0.8	0	0.01	1.18	0.32
DAP	0	0.8	0	0.01	1.18	0.32
NPK	2.806	-10.158	13	0.01	1.18	0.32

Table S5. Parameters used in estimates of annual TAN excretion per cattle.

	Raising cycle	Excretion (kg/cattle/day)		Nitrogen content (%)		TAN (%)	Annual TAN (tons/cattle)		
		Urine	Excrement	Urine	Excrement		Urine	Excrement	Total
Beef	365	7.5	13.5	0.9	0.38	60	0.0148	0.0112	0.0260
Cow	365	12	23.5	0.9	0.38	60	0.0237	0.0196	0.0432
Horse	365	6.5	15	1.4	0.2	60	0.0199	0.0066	0.0265
Donkey	365	6.5	15	1.4	0.2	60	0.0199	0.0066	0.0265
Mule	365	6.5	15	1.4	0.2	60	0.0199	0.0066	0.0265
Sow	365	5.7	2.1	0.4	0.34	70	0.0058	0.0018	0.0076
Hog	75	3.2	1.5	0.4	0.34	70	0.0007	0.0003	0.0009
Goat	365	0.705	2.05	1.35	0.75	55	0.0019	0.0031	0.0050
Sheep	365	0.705	2.05	1.35	0.75	55	0.0019	0.0031	0.0050
Layer	365	—	0.12	—	1.63	70	—	0.0005	0.0005
Laying duck	365	—	0.13	—	1.1	70	—	0.0004	0.0004
Broiler	50	—	0.09	—	1.63	70	—	5×10^{-5}	5×10^{-5}
Meat duck	55	—	0.1	—	1.1	70	—	4×10^{-5}	4×10^{-5}
Goose	70	—	0.1	—	0.55	70	—	3×10^{-5}	3×10^{-5}
Rabbit	55	0.3	0.15	0.15	1.72	45	—	6×10^{-5}	7×10^{-5}
Cattle	365	10	18	0.9	0.6	50	0.0164	0.01971	0.0361
Buffalo	365	10	18	0.9	0.6	45	0.0148	0.0177	0.0325

Table S6. The temperature-dependant emission factors by stage/phase in livestock farming in E2 (% TAN).

	EF _{outdoors}	EF _{Housing} (liquid)			EF _{Housing} (solid)			EF _{Storage} (liquid)				EF _{Storage} (solid)				EF _{spreading} (liquid)	EF _{spreading} (solid)
		<10°C	10-20°C	>20°C	<10°C	10-20°C	>20°C	NH ₃	N ₂ O	NO	N ₂	NH ₃	N ₂ O	NO	N ₂		
Free-range																	
Beef	53	7	10.5	14	7	10.5	14	20	1	0	0.3	27	8	1	30	55	79
Cow	41.5	7	10.5	14	7	10.5	14	20	1	0	0.3	27	8	1	30	55	79
Horse	0	9.3	14	18.7	9.3	14	18.7	35	0	0	0.3	35	8	1	30	90	81
Donkey	0	9.3	14	18.7	9.3	14	18.7	35	0	0	0.3	35	8	1	30	90	81
Mule	0	9.3	14	18.7	9.3	14	18.7	35	0	0	0.3	35	8	1	30	90	81
Sow	0	9.2	14.7	20.2	9.2	14.7	20.2	14	0	0	0.3	45	5	1	30	40	81
Hog	0	6.2	10.2	14.2	6.2	10.2	14.2	14	0	0	0.3	45	5	1	30	40	81
Goat	64	7	10.5	14	7	10.5	14	24	4	0	0.3	27.5	7.5	1	30	72.5	80
Sheep	64	7	10.5	14	7	10.5	14	24	4	0	0.3	27.5	7.5	1	30	72.5	80
Layer	69	24.9	45.2	56.5	24.9	45.2	56.5	0	0	0	0	14	4	1	30	0	63
Laying duck	54	24.9	45.2	56.5	24.9	45.2	56.5	0	0	0	0	24	3	1	30	0	63
Broiler	66	22.2	40.3	50.4	22.2	40.3	50.4	0	0	0	0	17	3	1	30	0	63
Meat duck	54	22.2	40.3	50.4	22.2	40.3	50.4	0	0	0	0	24	3	1	30	0	63
Goose	54	22.2	40.3	50.4	22.2	40.3	50.4	0	0	0	0	24	3	1	30	0	63
Intensive																	
Beef	53	7	10.5	14	7	10.5	14	16	1	0	0.3	4.2	8	1	30	55	79
Cow	41.5	7	10.5	14	7	10.5	14	16	1	0	0.3	4.2	8	1	30	55	79
Horse	0	9.3	14	18.7	9.3	14	18.7	16	0	0	0.3	4.2	8	1	30	90	81
Donkey	0	9.3	14	18.7	9.3	14	18.7	16	0	0	0.3	4.2	8	1	30	90	81
Mule	0	9.3	14	18.7	9.3	14	18.7	16	0	0	0.3	4.2	8	1	30	90	81

Continued Table S6

	EF _{outdoors}	EF _{Housing} (liquid)			EF _{Housing} (solid)			EF _{Storage} (liquid)				EF _{Storage} (solid)				EF _{spreading} (liquid)	EF _{spreading} (solid)
		<10°C	10-20 °C	>20 °C	<10°C	10-20 °C	>20 °C	NH ₃	N ₂ O	NO	N ₂	NH ₃	N ₂ O	NO	N ₂		
		Intensive															
Sow	0	8.9	14.3	19.7	8.9	14.3	19.7	3.8	0	0	0.3	4.6	5	1	30	40	81
Hog	0	11.3	18.5	25.7	11.3	18.5	25.7	3.8	0	0	0.3	4.6	5	1	30	40	81
Goat	64	7	10.5	14	7	10.5	14	16	4	0	0.3	4.2	7.5	1	30	72.5	80
Sheep	64	7	10.5	14	7	10.5	14	16	4	0	0.3	4.2	7.5	1	30	72.5	80
Layer	69	0	0	0	19.7	35.9	44.9	0	0	0	0	3.7	4	1	30	0	63
Laying duck	54	0	0	0	19.7	35.9	44.9	0	0	0	0	3.7	3	1	30	0	63
Broiler	66	0	0	0	22.2	40.3	50.4	0	0	0	0	0.8	3	1	30	0	63
Meat duck	54	0	0	0	22.2	40.3	50.4	0	0	0	0	0.8	3	1	30	0	63
Goose	54	0	0	0	22.2	40.3	50.4	0	0	0	0	0.8	3	1	30	0	63

Table S7. Model performance for meteorological parameters in D2.

Parameters	Indicator	January	April	July	October	Benchmark
Wind speed	Mean OBS (m/s)	2.50	2.62	2.52	2.64	
	Mean SIM (m/s)	2.55	2.52	2.38	2.63	
	Bias (m/s)	0.05	-0.10	-0.14	-0.009	$\leq \pm 0.5$
	RMSE (m/s)	0.27	0.29	0.33	0.27	≤ 2.0
	IOA	0.95	0.94	0.94	0.97	≥ 0.6
	R ²	0.82	0.82	0.82	0.89	
	NMB (%)	1.91	-3.94	-5.53	-0.32	
	NME (%)	8.37	9.10	10.53	7.75	
Wind direction	Mean OBS (°)	184.29	163.29	169.28	174.34	
	Mean SIM (°)	165.37	147.09	159.80	155.66	
	Bias (°)	-18.91	-15.68	-9.17	-18.68	$\leq \pm 10$
	RMSE (°)	33.88	27.82	23.15	30.93	
	IOA	0.87	0.88	0.87	0.92	
	R ²	0.69	0.74	0.63	0.81	
	NMB (%)	-10.26	-9.92	-5.60	-10.72	
	NME (%)	13.95	13.41	9.62	13.91	
Temperature	Mean OBS (°C)	5.60	16.30	27.33	18.69	
	Mean SIM (°C)	7.09	16.03	26.64	18.75	
	Bias (°C)	1.49	-0.26	-0.66	0.06	≤ 0.5
	RMSE (°C)	1.76	0.95	1.27	0.69	
	IOA	0.95	0.98	0.95	0.99	≥ 0.7
	R ²	0.95	0.94	0.89	0.96	
	NMB (%)	26.60	-1.62	-2.51	0.34	
	NME (%)	28.00	4.70	3.57	2.94	
Relative humidity	Mean OBS (%)	65.16	75.69	79.81	71.96	
	Mean SIM (%)	61.87	76.21	82.24	70.80	
	Bias (%)	-3.29	0.51	2.36	-1.16	
	RMSE (%)	6.61	3.86	4.24	4.07	
	IOA	0.97	0.98	0.95	0.98	≥ 0.7
	R ²	0.92	0.95	0.89	0.94	
	NMB (%)	-5.06	0.69	3.05	-1.61	
	NME (%)	8.42	4.23	4.29	4.15	

Note: OBS and SIM indicate the results from observation and simulation, respectively. The Bias, IOA, RMSE, NMB and NME were calculated using following equations (P and O indicates the results from modeling prediction and observation, respectively):

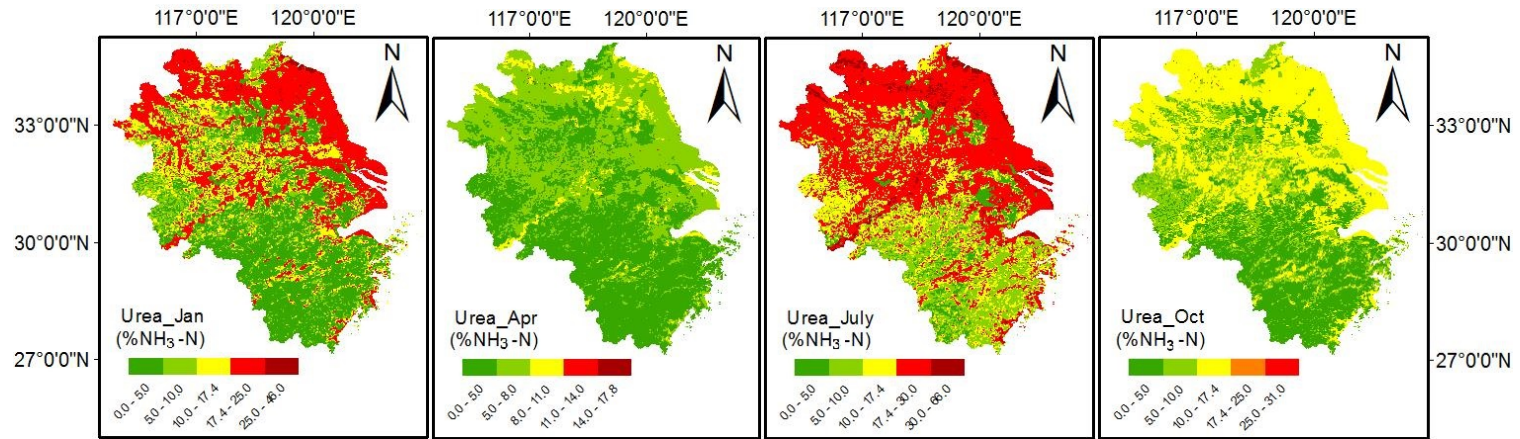
$$Bias = \frac{1}{n} \sum_{i=1}^n (P_i - O_i); IOA = 1 - \frac{\sum_{i=1}^n (P_i - O_i)^2}{\sum_{i=1}^n \left(\left| P_i - \bar{O} \right| + \left| O_i - \bar{O} \right| \right)^2}; RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (P_i - O_i)^2}$$

$$NMB = \frac{\sum_{i=1}^n (P_i - O_i)}{\sum_{i=1}^n O_i} \times 100\%; NME = \frac{\sum_{i=1}^n |P_i - O_i|}{\sum_{i=1}^n O_i} \times 100\%$$

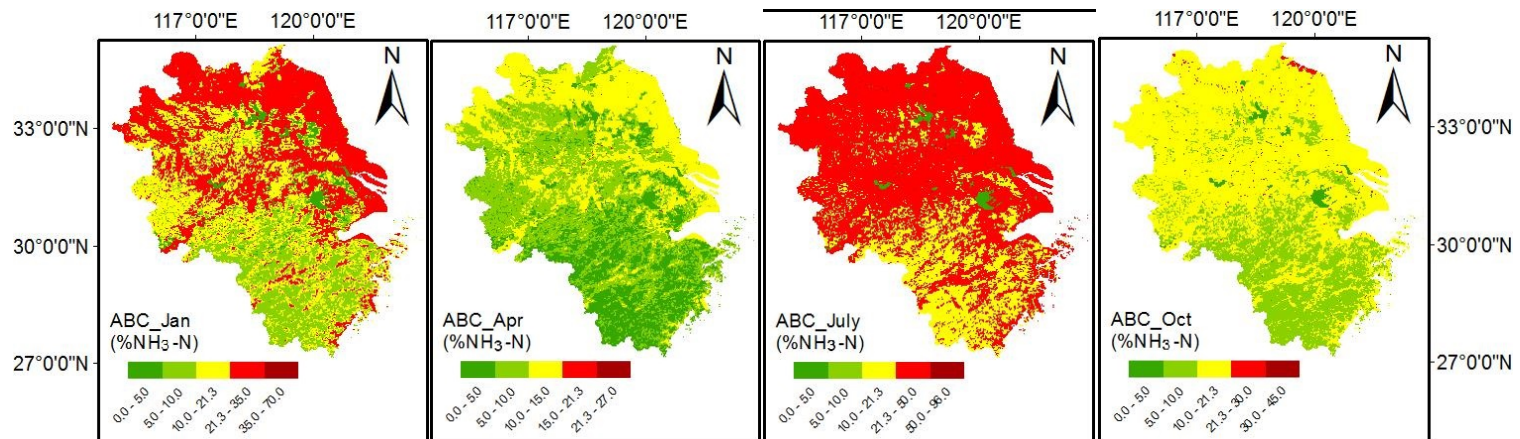
Table S8. Inter-annual change in SO₂ and NO₂ VCDs for the YRD region 2012-2014 (%)

	Jiangsu	Zhejiang	Anhui	Shanghai	Total YRD
SO ₂	-49.94	-29.53	-51.38	-56.72	-47.84
NO ₂	-29.12	-26.22	-35.69	-29.48	-30.91

Figure S1. Corrected NH_3 volatilization of urea and ABC application in E2



(a) NH_3 volatility of urea



(b) NH_3 volatility of ABC

Figure S2. Total ammoniacal nitrogen (TAN) for three main raising systems (taken from Huang et al., 2012)

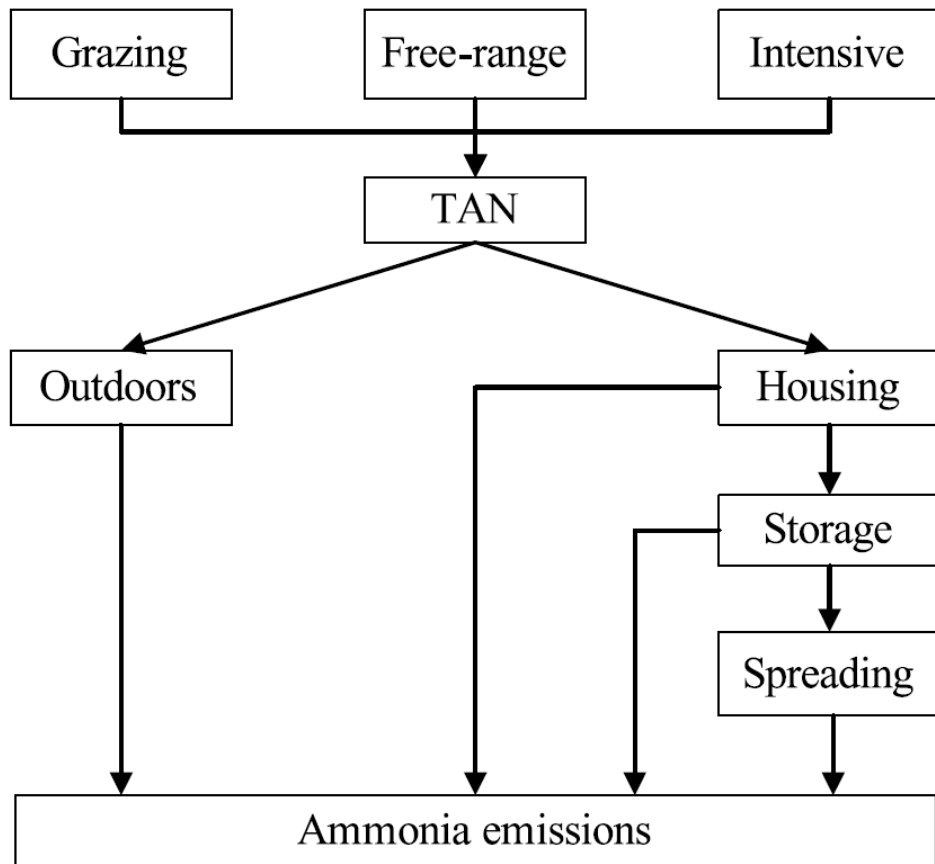


Figure S3. Daily NH₃ concentration at JSPAES for October 2014 from observation and simulation with E1 and E2.

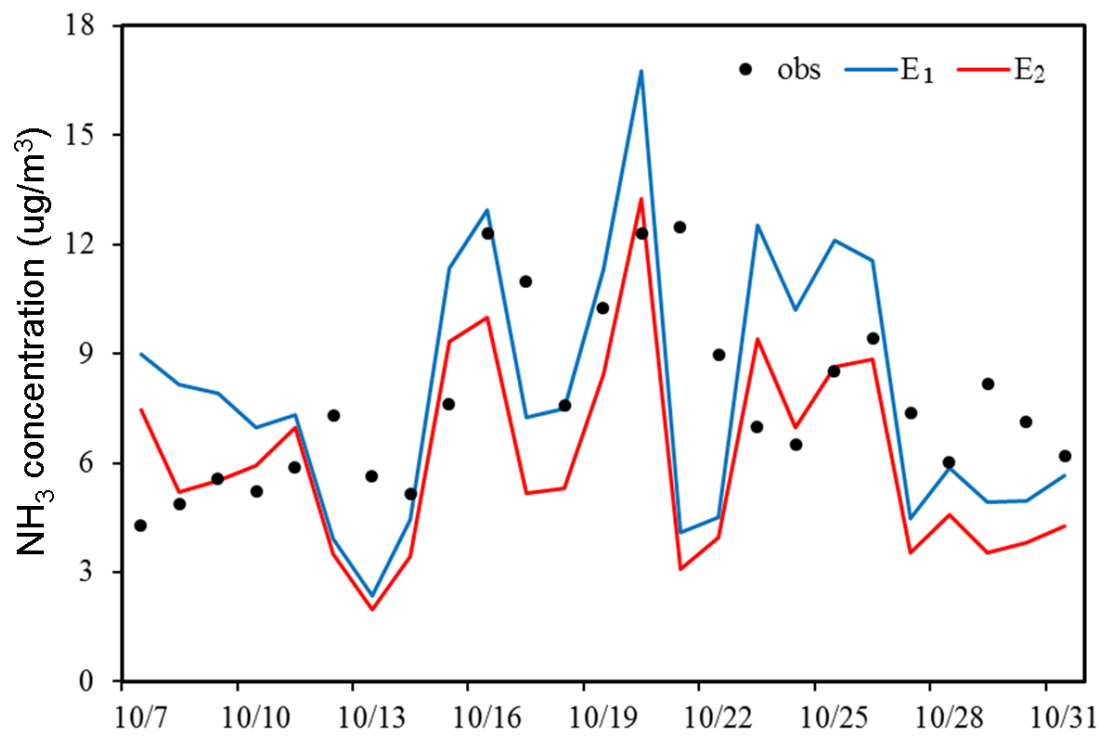
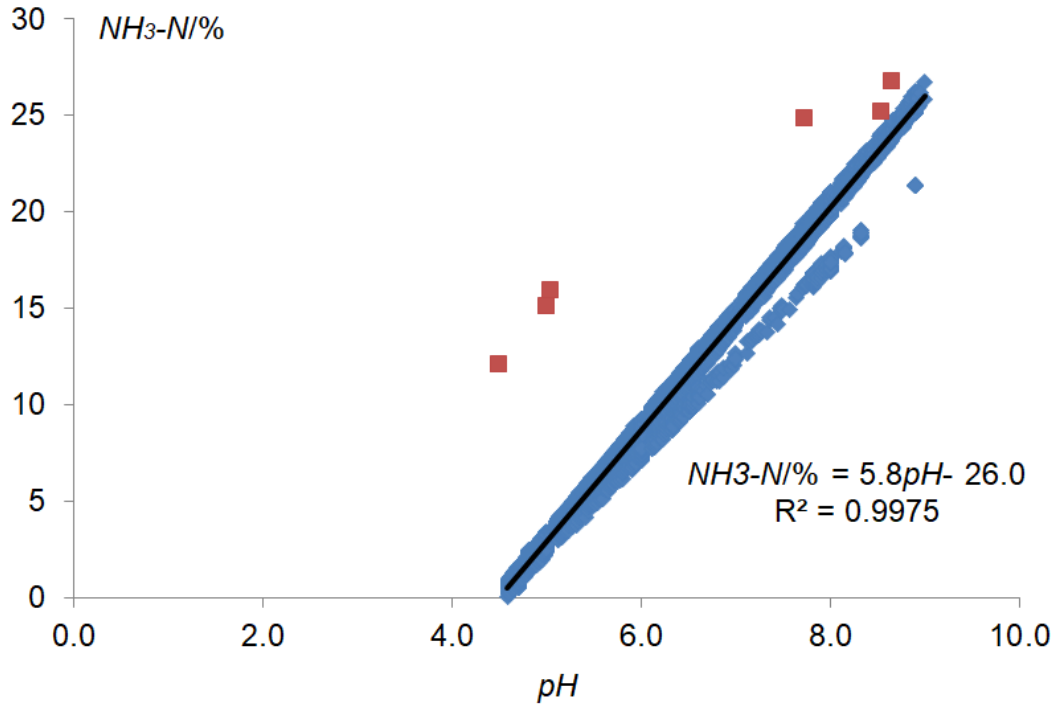
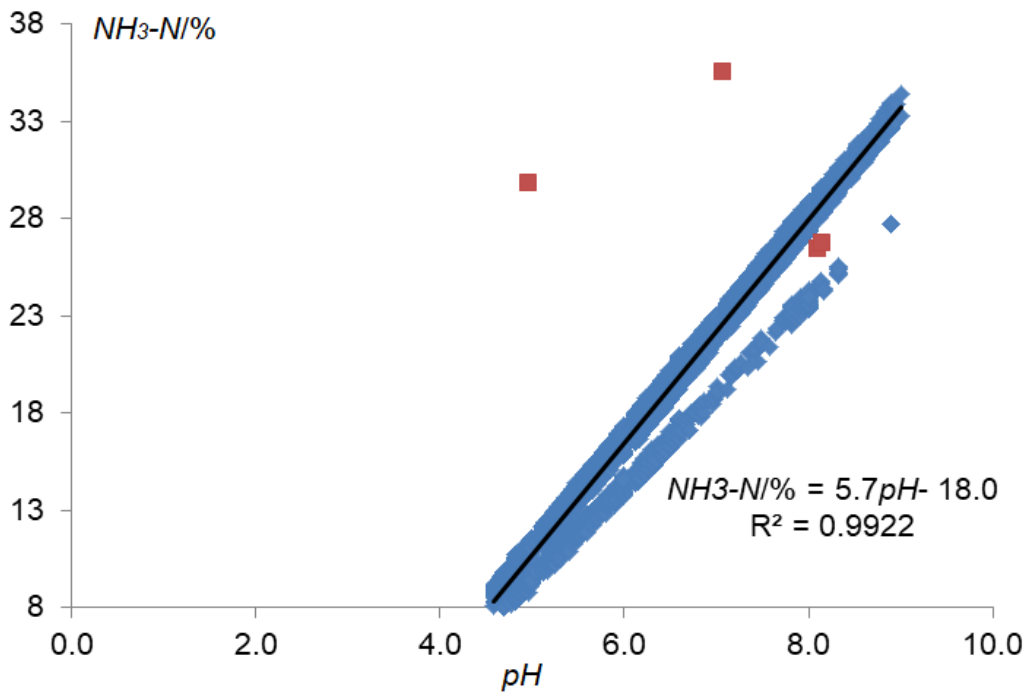


Figure S4. The NH_3 volatilization rates under different soil pH values for urea (a) and ABC fertilizer (b). The blue dots indicate the values by grid in the YRD region in E2, while the red dots indicate the results from the field measurements by Zhong et al. (2006) (a) and Zhang et al. (2002) (b). The solid lines and equations were obtained from linear regression for the gridded values in the YRD region in E2.



(a) Urea



(b) ABC

References

- Dong, W., Xin, J., Wang, S.: Temporal and spatial distribution of anthropogenic ammonia emissions in China: 1994–2006, *Environ. Sci.*, 31, 1457–1463, 2010 (in Chinese).
- Dong, Y., Chen, C., Huang, C., Wang, H., Li, L., Dai, P., Jia, J.: Anthropogenic emissions and distribution of ammonia over the Yangtze River Delta, *Acta Scientiae Circumstantiae*, 29, 1611-1617, 2009.
- Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., Zhang, H.: A high-resolution ammonia emission inventory in China. *Global Biogeochem. Cy.*, 26, GB1030, doi: 10.1029/2011GB004161, 2012.
- Liu, C., Yao, L.: Agricultural ammonia emission inventory and its distribution characteristics in Jiangsu Province, *Journal of Anhui Agri. Sci.*, 44, 70-74, 2016 (in Chinese).
- Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., Zhu, T.: High-resolution ammonia emissions inventories in China from 1980 to 2012, *Atmos. Chem. Phys.*, 16, 2043–2058, 2016.
- Pan, T., Xue, N., Sun, C., Liu, G., Zhang, J., Li, S.: Distribution characteristics of ammonia emission from livestock farming industry in Beijing, *Environ. Sci. Technol.*, 38, 159-162, 2015.
- Shen, X., Yin, S., Zheng, J., Lu, Q., Zhong, L.: Anthropogenic ammonia emission inventory and its mitigation potential in Guangdong Province, *Acta Scientiae Circumstantiae*, 34, 43-53, 2014.
- Yang, Z.: Estimation of ammonia emission from livestock in China based on mass-flow method and regional comparison, Master thesis, Peking University, Beijing, China, 2008.
- Yin, S., Zheng, J., Zhang, L., Zhong, L.: Anthropogenic ammonia emission inventory and characteristics in the Pearl River Delta region, *Environ. Sci.*, 31, 1146-1151, 2010.
- Zhang, Q., Zhang, M., Yang, Y., Lu, J.: Volatilization of ammonium bicarbonate and urea in main soil of Shandong Province, *Chinese Journal of Soil Science*, 33, 32-34, 2002.
- Zheng, J., Yin, S., Kang, D., Che, W., Zhong, L.: Development and uncertainty analysis of a high-resolution NH₃ emission inventory and its implications with precipitation over the Pearl River Delta region, China, *Atmos. Chem. Phys.*, 12, 7041-7058, 2012.
- Zhong, N., Zeng, Q., Zhang, L., Liao, B., Zhou, X., Jiang, J.: Effects of acidity and alkalinity on urea transformation in soil, *Chinese Journal of Soil Science*, 37, 1123-1128, 2006.
- Zhou, F., Ciais, P., Hayashi, K., Galloway, J., Kim, D.-G., Yang, C., Li, S., Liu, B., Shang, Z., Gao, S.: Re-estimating NH₃ emissions from Chinese cropland by a new nonlinear model, *Environ. Sci. Technol.*, 50, 564-572, 2015.