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Improved gridded ammonia emission inventory in China

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Improved gridded ammonia emission inventory in China

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Section S1 Sources of fertilization application timing and frequency

We evaluated the fertilizer application timing and frequency for rice, maize, and wheat crops in different regions by collecting data from a large number of studies and the technical guidelines for field management in each province in 2016. Table S5 lists the main data sources for the fertilization application timing and frequency. Most of the collected reports/websites were published by the national or provincial governments in 2016.

Section S2 WRF-Chem model configuration

A fully coupled online Weather Research and Forecasting with Chemistry model (WRF-Chem v3.7) is used to evaluate the accuracy of different NH_3 emission inventories. The WRF-Chem model is designed to cover most parts of North, East, Central, and South China at the horizontal resolutions of 27 km (Fig.2). The vertical dimension is resolved by 46 full sigma levels, with 18 layers located in the bottom 2 km for finer resolution in the planetary boundary layer; the height of the first layer averaged over the analyzed region is about 30 m.

Meteorological initial and lateral boundary conditions used in the WRF-Chem model are taken from the NCEP (National Center for Environmental Prediction) (Final) Operational Global Analysis data with a spatial resolution of 1° × 1°. The forecasts from the MOZART-4 global chemical transport model are processed to provide the chemical initial and boundary conditions for the WRF-Chem model (Emmons et al., 2010).

Air pollutants emissions (including SO_2, NO_x, CO, CO_2, NMVOC, BC, OC, $PM_{2.5}$, and PM_{10}) of 2016 were obtained from Multi-resolution Emission Inventory for China (MEIC) (http://meicmodel.org/), with the horizontal resolution of 0.25° (Li et al., 2017). The emission rate of each species for each hour is based on Gao et al. (2015). The biogenic emissions are calculated online using the MEGANv2.04 (Model of Emission of Gases and Aerosol from Nature v2.04) model (Guenther et al., 2006). Biomass-burning emissions are obtained from the GFEDv3 (Global Fire Emissions Database v3) (Randerson et al., 2005). Dust emissions and sea salt emissions are calculated online using algorithms proposed by Shao (2004) and Gong et al. (1997), respectively.

The Carbon Bond Mechanism Z (CBM-Z) is selected as the gas-phase chemical mechanism (Zaveri and Peters, 1999), and the full 8-bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol module with aqueous chemistry is used to simulate aerosol evolution (Zaveri et al., 2008). The photolysis rates are calculated by the Fast-J scheme (Wild et al., 2000). Aerosol radiation is simulated using RRTMG (Rapid Radiative Transfer Model for GCMs) for both shortwave (SW) and longwave (LW) radiation (Zhao et al., 2011). Other major physical parameterizations used in this study are listed in Table S6.

Table S1. Basal and topdressing fertilization dates for maize, wheat, and rice in mainland China (2016)

Province	Crop	Basal	Topdressing	
			First	Second
Beijing	Winter wheat	September 21-October 20	March 21-April 10	April 21-May 10
	Summer maize	June 1-20	July 11-31	August 1-20
Tianjin	Spring wheat	April 11-20	June 1-20	July 1-10
	Single-season rice	June 1-10	June 21-July 20	August 1-10
	Winter wheat	September 21-October 20	March 21-April 10	April 21-May 10
	Summer maize	June 1-20	July 11-31	August 1-20
Hebei	Single-season rice	June 1-10	June 21-July 20	August 1-10
	Winter wheat	September 21-October 20	March 21-April 10	April 21-May 10
	Summer maize	June 1-20	July 11-31	August 1-20
Shanxi	Spring maize	April 21-May 10	June 11-July 20	July 11-August 20
	Winter wheat	September 21-October 10	March 21-April 10	April 21-May 10
	Summer maize	June 1-20	July 11-31	August 1-20
Inner Mongolia	Spring wheat	April 11-20	June 1-20	July 1-10
	Spring maize	April 21-May 10	June 11-July 20	July 11-August 20
	Single-season rice	May 1-20	June 11-20	July 11-31
Liaoning	Spring maize	April 21-May 10	June 11-July 20	July 11-August 20
	Single-season rice	May 1-20	June 11-20	July 11-31
Jilin	Spring maize	April 21-May 10	June 11-July 20	July 11-August 20
	Single-season rice	May 1-20	June 11-20	July 11-31
Heilongjiang	Spring wheat	April 11-20	June 1-20	July 1-10
	Spring maize	April 21-May 10	June 11-July 20	July 11-August 20
	Single-season rice	May 1-20	June 11-20	July 11-31
Shanghai	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-31	March 21-April 10	April 21-May 10
	middle rice	June 1-20	July 11-20	August 1-20
Jiangsu	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Summer maize	June 1-20	July 11-31	August 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Zhejiang	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Late rice	July 11-31	August 1-20	September 1-20
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Anhui	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Late rice	July 11-31	August 1-20	September 1-20
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20

Fujian	Single-season rice	May 11-31	June 11-30	July 11-31
	Late rice	July 11-31	August 1-20	September 1-20
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Jiangxi	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Late rice	July 11-31	August 1-20	September 1-20
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Shandong	Single-season rice	June 1-10	June 21-July 20	August 1-10
	Winter wheat	September 21-October 20	March 21-April 10	April 21-May 10
	Summer maize	June 11-30	July 11-31	August 1-20
Henan	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-31	March 21-April 10	April 21-May 10
	Summer maize	June 1-20	July 11-31	August 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Hubei	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Late rice	July 11-31	August 1-20	September 11-30
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Hunan	Single-season rice	May 11-31	June 11-30	July 11-31
	Winter wheat	October 11-November 20	March 11-April 20	April 11-May 20
	Late rice	July 11-31	August 1-20	September 11-30
	Summer maize	June 1-20	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Guangdong	Late rice	July 11-31	August 1-20	September 11-30
	Summer maize	June 21-30	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
Guangxi	Single-season rice	May 11-31	June 11-30	July 11-31
	Late rice	July 11-31	August 1-20	September 11-30
	Summer maize	June 21-30	July 11-31	August 1-20
	Early rice	April 21-May 10	May 11-31	June 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Hainan	Late rice	July 11-31	August 1-20	September 11-30
	Early rice	April 21-May 10	May 11-31	June 1-20
Chongqing	Spring maize	March 11-April 20	May 21-31	June 1-20
	Single-season rice	May 1-20	June 11-30	July 11-31
	Winter wheat	October 21-November 20	January 11-Febuary 20	February 10-March 20
	middle rice	June 1-20	July 11-20	August 1-20

Sichuan	Spring maize	March 11-April 20	May 21-31	June 1-20
	Single-season rice	May 1-20	June 11-30	July 11-31
	Winter wheat	October 21-November 20	January 11-Febuary 20	February 10-March 20
	middle rice	June 1-20	July 11-20	August 1-20
Guizhou	Spring maize	March 11-April 20	May 21-31	June 1-20
	Single-season rice	May 1-20	June 11-30	July 11-31
	Winter wheat	October 21-November 20	January 11-Febuary 20	February 10-March 20
	middle rice	June 1-20	July 11-20	August 1-20
Yunnan	Spring maize	March 11-April 20	May 21-31	June 1-20
	Single-season rice	May 1-20	June 11-30	July 11-31
	Winter wheat	October 21-November 20	January 11-Febuary 20	February 10-March 20
	Late rice	August 11-20	September 1-10	October 11-20
	Early rice	February 11-20	March 11-20	April 1-20
	middle rice	June 1-20	July 11-20	August 1-20
Tibet	Winter wheat	October 21-November 20	January 11-Febuary 20	February 10-March 20
Shaanxi	Spring maize	April 21-May 10	June 11-30	July 11-31
	Single-season rice	May 1-10	June 11-20	July 11-31
	Winter wheat	September 11-October 20	March 11-31	April 11-30
Gansu	Spring wheat	March 11-April 20	May 21-31	June 11-30
	Spring maize	April 21-May 10	June 11-30	July 11-31
	Winter wheat	September 11-October 20	March 11-31	April 11-30
Qinghai	Spring wheat	March 11-April 20	May 21-31	June 11-30
	Spring maize	April 21-May 10	June 11-30	July 11-31
Ningxia	Spring wheat	March 11-April 20	May 21-31	June 11-30
	Spring maize	April 21-May 10	June 11-30	July 11-31
	Single-season rice	May 1-10	June 11-20	July 11-31
	Winter wheat	September 11-October 20	March 11-31	April 11-30
Xinjiang	Spring wheat	March 11-April 20	May 21-31	June 11-30
	Spring maize	April 11-May 20	June 11-30	July 11-31
	Single-season rice	May 1-20	June 11-20	July 11-31
	Winter wheat	September 11-October 20	March 11-31	April 11-30

Note: Farmers often fertilize crops according to their planting experiences and available time, resulting in some uncertainty around the fertilization dates.

Table S2. Emission factor index for fertilizer application

Factor		Value	References
EF_0	urea	17.4 % NH3-N	(Cai et al., 2002; Dong et al., 2009; Zhou et al., 2016; Zhao et al., 2020)
	ammonium bicarbonate	21.3% NH3-N	
	diammonium phosphate	7.3% NH3-N	
	NPK compound fertilizer	5% NH3-N	
	other	4% NH3-N	
Crop	Upland crops	-0.045	(Zhang et al., 2018)
	Flooded crops	0	
Method	Basal	-1.139	(Huang et al., 2012)
	Top	0	
Soil pH		$0.067\times pH^2-0.69\times pH+0.68$	(Bouwman et al., 2002; Zhang et al., 2018)
Soil CEC	CEC ≤16	0.088	(Zhang et al., 2018)
	16＜CEC≤24	0.012	
	24＜CEC≤32	0.163	
	CEC＞32	0	

Note: for soil pH, a second-order function is applied to fit the segmental values given in (Bouwman et al., 2002; Zhang et al., 2018).

Table S3. The parameters used in estimates of annual TAN excretion per cattle

	Raising cycle	Excretion (kg/cattle/day)		Nitrogen content (%)		TAN (%)
		Urine	Excrement	Urine	Excrement	
beef cattle	365	10	20	0.9	0.38	60
dairy cow	365	19	40	0.9	0.38	60
goat	365	0.75	2.6	1.35	0.75	50
sheep	365	0.75	2.6	1.35	0.75	50
rabbit	55	0.3	0.15	0.15	1.72	45
horse/donkey/mule	365	6.5	15	1.4	0.2	60
sow	365	5.7	2.1	0.4	0.34	70
fattening pig	75	3.2	1.5	0.4	0.34	70
camel	365	6.5	15	1.4	0.2	60
meat duck	55	-	0.1	-	1.1	70
meat goose	70	-	0.1	-	0.55	70
broilers	50	-	0.09	-	1.63	70
laying hen	365	-	0.12	-	1.63	70
laying duck	365	-	0.13	-	1.1	70

Table S4. The EF and spatial allocation methods of NH_3 emission sources

Category	Subcategory	EF	Spatial allocation
Transportation	light-duty gasoline vehicles	0.026 g/km	Road density [a]
	heavy-duty gasoline vehicles	0.028 g/km	
	light-duty diesel vehicles	0.004 g/km	
	heavy-duty diesel vehicles	0.017 g/km	
	motorcycles	0.007 g/km	
Residential & commercial	human excrement	787 g/capita	Rural population [b]
	indoor firewood combustion	1.3 g/kg	
	indoor wheat burning	0.52 g/kg	
	indoor rice burning	0.37 g/kg	
	Indoor maize burning	0.68 g/kg	
	domestic coal combustion	1 g/kg	Population [b]
	domestic oil combustion	120 g/10^3L	
	domestic gas combustion	3203.8 g/10^4m^3	
Industry	synthetic ammonia	787 g/t	Sub-national fuel data disaggregation method using POI (SDMP) [c]
	nitrogen fertilizers production	5000 g/t	
	wastewater treatment	30 g/10^4 m^3	
	waste landfill	0.0073 kg/kg CH4	
	waste incineration	210 g/t	
	coal combustion	10 g/t	
	oil combustion	100 g/10^3L	
	gas combustion	512.6 g/10^4m^3	
Others	agricultural soil	1800 g/ha	Cropland [d]
	nitrogen-fixing plants (soybean)	1050 g/ha	Soybean harvest areas [e]
	nitrogen-fixing plants (peanuts)	1200 g/ha	Groundnut harvest areas [e]
	outdoor straw burning	0.53 g/kg	based on the gridded burned area in cropland [f]
	Forest fires	2.9 g/kg	Gridded burned areas in forest [f]
	Grassland fires	0.7 g/kg	Gridded burned areas in grass [f]

[a] The road networks were derived from the OpenStreet data (http://www.geofabrik.de).

[b] The population data in 2016 were downloaded at https://landscan.ornl.gov/download.

[c] Details about the DPOI allocation method were previously discussed (Li et al., 2019).

[d] The cropland was derived from the Resource and Environment Science and Data Center. (http://www.resdc.cn/data.aspx?DATAID=184).

[e] The harvest areas of each crop were derived from the EarthStat dataset.

[f] The gridded burned area data were determined by coupling the MCD64A1 and MCD14ML fire products based on previous studies (Li et al., 2018; Qiu et al., 2016).

Table S5. Main sources of the fertilization application timing and frequency for the three main crops: rice, maize, and wheat.

Crops	Regions	Sources
Wheat	Major regions of China	http://www.moa.gov.cn/ztzl/ql/jszd/201602/t20160223_5024971.htm
		http://www.moa.gov.cn/ztzl/2016ncg/jszd/201603/t20160317_5058189.htm
		http://www.moa.gov.cn/gk/nszd_1/2016nszd/201609/t20160922_5282096.htm
	Southwest China	http://www.moa.gov.cn/ztzl/2016ncg/jszd/201602/t20160205_5009571.htm
	North of China	http://www.cma.gov.cn/2011xwzx/2011xqxxw/2011xqxyw/201609/t20160928_323755.html
	Hebei	http://nync.hebei.gov.cn/article/tzgg/201603/20160300000999.shtml
		http://nync.hebei.gov.cn/article/tzgg/201509/20150990059492.shtml
	Shandong	https://www.tuliu.com/read-29459.html
	Anhui	http://nync.ah.gov.cn/snzx/sxxx/11016591.html
		https://www.tuliu.com/read-24814.html
	Shanxi	https://www.tuliu.com/read-26184.html
	Henan	https://www.163.com/dy/article/F6NFH60I05322B99.html
	Jiangsu	https://www.sohu.com/a/291887309_99951299
	Hubei	http://www.ampcn.com/info/detail/22904.asp
	Sichuan	http://www.mynky.cn/Html/FuWuSanNong/NongJiBaiKe/202101/2Io4Me8Am4Ut41491796.html
	Shaanxi	https://baijiahao.baidu.com/s?id=1692466171471969276&wfr=spider&for=pc
	Gansu	https://www.longnan.gov.cn/4455585/15819653.html
	Xinjiang	http://nynct.xinjiang.gov.cn/nynct/ywnjtg/201603/a08b46a4c9f54dc7ada0ee185045f289.shtml
		https://www.sohu.com/a/345312778_100082336
Maize	Northeast China	http://www.moa.gov.cn/ztzl/ql/jszd/201605/t20160506_5120809.htm
		http://www.moa.gov.cn/gk/nszd_1/2016nszd/201607/t20160726_5218391.htm
	Major regions of China	http://www.moa.gov.cn/ztzl/2016ncg/jszd/201603/t20160317_5058189.htm
	Huang-Huai-Hai region	http://www.agri.cn/V20/ZX/nyyw/201606/t20160606_5162723.htm
	Southwest China	http://www.moa.gov.cn/ztzl/ql/jszd/201508/t20150824_4802201.htm
	Hebei	http://nync.hebei.gov.cn/article/tzgg/201607/20160700001943.shtml
		http://nync.hebei.gov.cn/article/tzgg/201607/20160700001943.shtml
	Shandong	https://www.sohu.com/a/164032835_266055
	Jiangsu	http://www.ampcn.com/info/content.asp?newsid=26871
	Liaoning	http://www.moa.gov.cn/xw/qg/201505/t20150522_4609518.htm
	Jilin	http://www.moa.gov.cn/xw/qg/202108/t20210827_6375068.htm
		http://www.jlates.cn/zwzp/144.html
		http://www.jlates.cn/zwzp/230.html
	Heilongjiang	http://www.agri.cn/V20/syjs/zzjs/201202/t20120206_2475334.htm
		https://www.tuliu.com/read-31938.html
	Henan	http://www.xxagri.org.cn/news/10_1745
		http://nynct.henan.gov.cn/2019/06-14/934805.html
	Hubei	http://www.jiangxia.gov.cn/msrd_22342/202102/t20210222_1637576.html
	Guangxi	http://nynct.gxzf.gov.cn/xwdt/gxlb/nn/t1837418.shtml
		http://www.gxny.gov.cn/xwdt/syjs/t617158.html
	Chongqing	http://www.cqaas.cn/nky/index/detail/id/314.html
	Yunnan	http://news.wugu.com.cn/article/830800.html

	Gansu	http://www.moa.gov.cn/ztzl/ql/dtbd/200907/t20090709_1307301.htm
	Ningxia	http://www.nx12346.com/zxkt/zjwg/zjzzjs/zjls/202105/t20210529_398362.html
	Xinjiang	https://www.sohu.com/a/379144178_293364
Rice	Major regions of China	http://www.moa.gov.cn/ztzl/2016ncg/jszd/201603/t20160317_5058189.htm
	Middle and lower reaches of Yangtze River	http://www.moa.gov.cn/ztzl/2016ncg/jszd/201605/t20160512_5126581.htm
	Northeast China	https://news.cnhnb.com/rdzx/detail/385809/
	South of China	http://www.moa.gov.cn/gk/nszd_1/2016nszd/201604/t20160405_5083015.htm
		http://www.moa.gov.cn/gk/nszd_1/2016nszd/201606/t20160619_5180240.htm
		http://www.moa.gov.cn/gk/nszd_1/2016nszd/201607/t20160714_5206812.htm
		http://www.moa.gov.cn/xw/zwdt/201607/t20160728_5221628.htm
	Southwest China	http://www.moa.gov.cn/gk/nszd_1/2019/201907/t20190719_6321226.htm
	Jiangsu	http://www.agri.cn/kj/syjs/zzjs/201604/t20160418_5098223.htm
		http://nynct.jiangsu.gov.cn/art/2017/8/22/art_13469_6156717.html
	Henan	http://nynct.henan.gov.cn/2019/06-14/934805.html
	Anhui	http://nync.ah.gov.cn/public/7021/11273021.html
	Heilongjiang	https://www.tuliu.com/read-31938.html
	Jilin	http://www.jlates.cn/zwzp/111.html
		http://www.jlates.cn/zwzp/144.html
		http://www.jlates.cn/zwzp/230.html
	Zhejiang	http://www.ruian.gov.cn/art/2016/6/1/art_1229181511_1222768.html
		http://www.zjpy.gov.cn/art/2015/9/21/art_1229252500_3953190.html
		http://nyncj.ningbo.gov.cn/art/2016/8/22/art_1229058373_48548981.html
	Fujian	http://nynct.fujian.gov.cn/xxgk/gzdt/xxkd/201606/t20160621_2559895.htm
		http://nynct.fujian.gov.cn/xxgk/gzdt/xxkd/201607/t20160729_2558957.htm
	Jiangxi	http://nync.jiangxi.gov.cn/art/2019/6/10/art_27910_908458.html
		http://nync.jiangxi.gov.cn/art/2020/8/25/art_27910_2808929.html
	Hubei	http://www.jiangxia.gov.cn/msrd_22342/202102/t20210222_1637576.html
		http://nyncj.wuhan.gov.cn/xwzx_25/xxlb/202001/t20200103_494873.html
	Hunan	http://www.agri.cn/kj/nyhljc/qy/201603/t20160331_5078911.htm
	Guangxi	http://www.gxny.gov.cn/xwdt/gxlb/nn/t608354.html
		http://www.gxny.gov.cn/xwdt/syjs/t617158.html
	Chongqing	http://www.cqaas.cn/nky/index/detail/id/312.html
	Hainan	http://www.gov.cn/xinwen/2016-04/06/content_5061635.htm
	Sichuan	http://www.mynky.cn/Html/FuWuSanNong/NongJiBaiKe/202006/1Hb7Hk3Ac0Sf19181709.html

Note: These websites were accessed on September 20, 2021. Most of these websites were published by the national or provincial governments in 2016.

Table S6. Parameterizations used in the WRF-Chem model

Options	WRF-Chem
Microphysics option	Purdue Lin Scheme
Longwave radiation option	RRTMG Scheme
Shortwave radiation option	RRTMG Scheme
Surface layer option	Revised MM5 Monin-Obukhov Scheme
Land surface option	Unified Noah land-surface model
Urban canopy model	Single-layer UCM Scheme
Boundary layer option	YSU Scheme
Cumulus option	Grell 3D ensemble Scheme
Photolysis scheme	Fast-J
Dust scheme	Shao_2004
Chemistry option	CBMZ
Aerosol option	MOSAIC

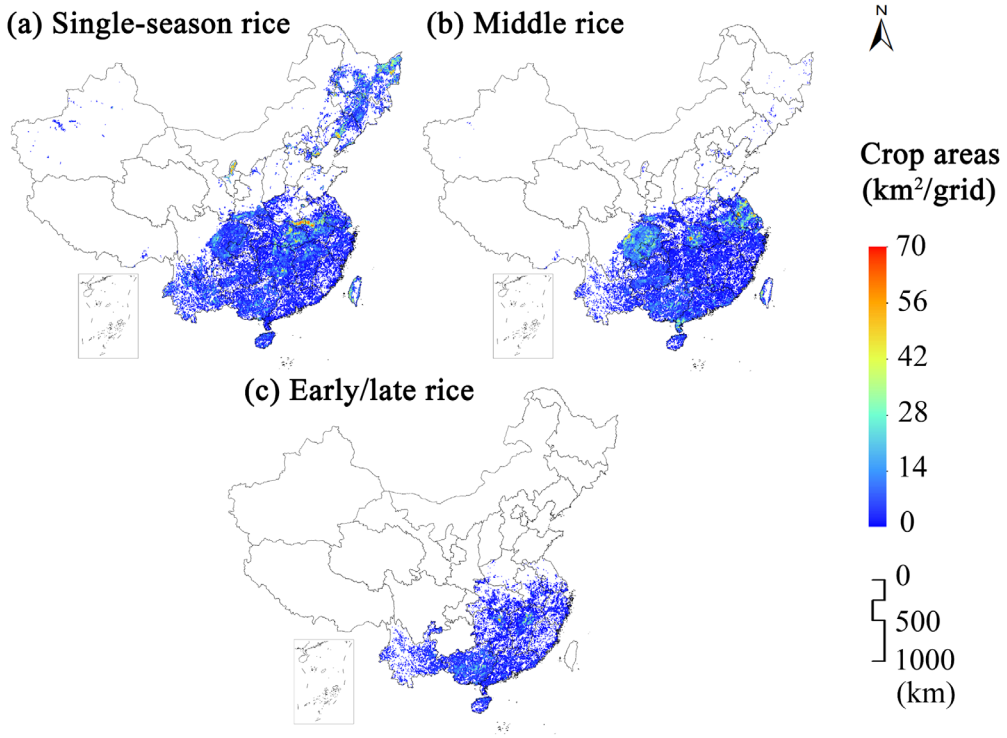

Fig. S1. Spatial distribution of single-season rice, middle rice and early/late rice.

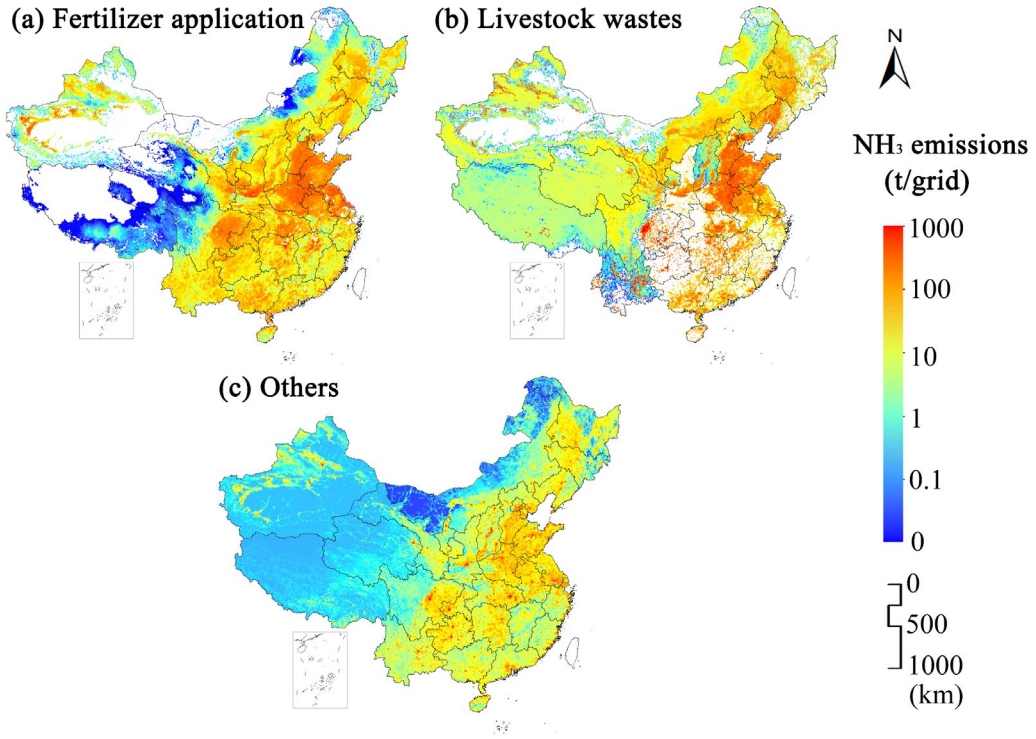

Fig. S2. Geographical distribution of NH3 emission from fertilizer application, livestock wastes, and others in mainland China (2016).

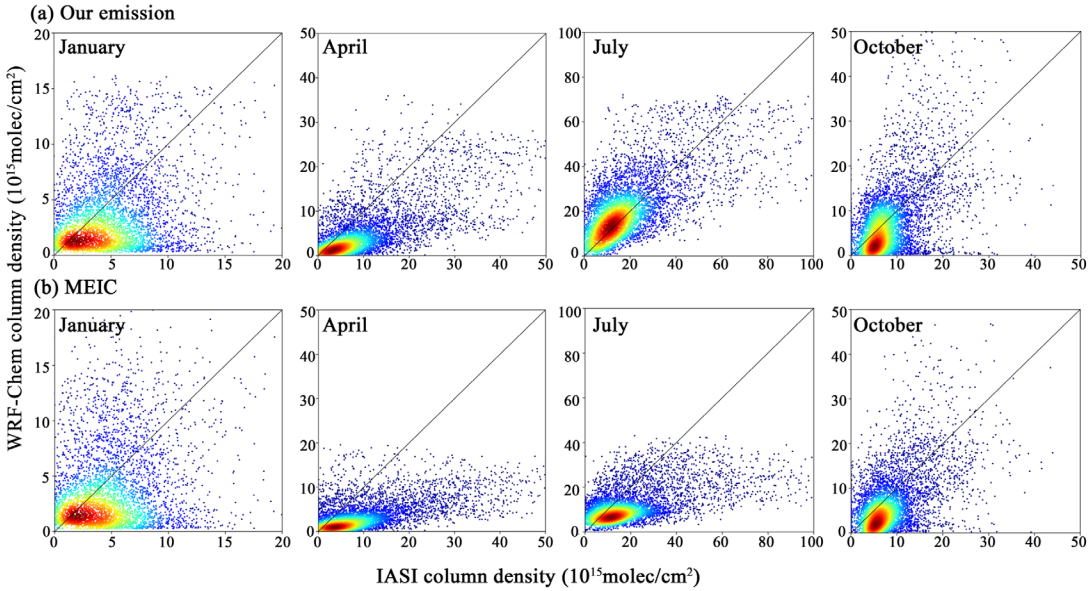

Fig. S3. Comparison between IASI-based VCDs and simulated NH_3 VCDs obtained in this study and MEIC, for January, April, July, and October. The range of the axes on the scatter plots for the different months is not the same.

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