



## Supplement of

# Trends in secondary inorganic aerosol pollution in China and its responses to emission controls of precursors in wintertime

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#### 1 S1: Method1: Calculation of other parameters from meta-analysis

Sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) are indicators of
secondary pollutant transformation in the atmosphere (Sun et al., 2006; Xu et al., 2017).
Higher SOR and NOR values imply greater oxidation of gaseous species to sulfate- and
nitrate-containing secondary particles (Sun et al., 2006), respectively. Their formulae
are as follows, where *n* refers to the molar concentrations:

7 
$$SOR = nSO_4^{2-} / (nSO_4^{2-} + nSO_2)$$
 (1)

8 
$$NOR = nNO_3^-/(nNO_3^- + nNO_2)$$
 (2)

To identify whether acidic species are fully neutralized by NH<sub>3</sub> in PM<sub>2.5</sub>, we selected 9 two indicators: the slope of the linear regression between equivalent concentrations of 10  $[NH_4^+]$  and  $[SO_4^{2-}]$  and the slope of the linear regression between equivalent 11 concentrations of  $[NH_4^+]$  and  $[SO_4^{2^-} + NO_3^-]$  (Sun et al., 2006). In the atmosphere, NH<sub>3</sub> 12 is first taken up by sulfuric acid to form ammonium sulfate salts and then any excess 13 14 NH<sub>3</sub> may then react with nitric and hydrochloric acids to form ammonium nitrate and ammonium chloride (Ianniello et al., 2010). When the slope of NS is less than 1, NH<sub>3</sub> 15 is not completely neutralized with sulfuric acid and nitric acid, suggesting a NH<sub>3</sub>-16 17 limited environment. Slopes of NS and NSN greater than 1 indicate that they reacted completely, suggesting a NH<sub>3</sub>-rich environment (Xu et al., 2017). 18

### 19 S2: Method2: Additional information about the monitoring data

The concentrations of  $PM_{2.5}$ ,  $SO_2$ , and  $NO_2$  were measured at state-controlled air sampling sites located within each city or county. To avoid direct influence of potential air pollution sources, most of the monitoring sites were situated in background locations in the urban areas. Concentrations of  $PM_{2.5}$  were measured using the micro oscillating

24	balance method (TEOM from Rupprecht & Patashnick Co., Inc.,USA) or the $\beta$
25	absorption method (BAM 1020 from Met One Instrument Inc., USA; Tianhong Co.,
26	China or Xianhe Co., China). Concentrations of SO <sub>2</sub> were measured using either UV-
27	spectrophotometry (TEI Model 49i from Thermo Fisher Scientific Inc., USA) or
28	Ultraviolet Fluorescence (TEI Model 43i from Thermo Fisher Scientific Inc., USA)
29	methods, and concentrations of $NO_2$ using the chemiluminescence (TEI Model 42i from
30	Thermo Fisher Scientific Inc., USA) method. The detection limits (DL) of these
31	techniques are sufficient to measure accurately the high or relatively high concentration
32	of PM <sub>2.5</sub> , SO <sub>2</sub> , and NO <sub>2</sub> at all monitoring sites.
33	Data from all monitoring sites were automatically released to an open website after
34	validation using HJ630-2011 specifications
35	(http://kjs.mep.gov.cn/hjbhbz/bzwb/other/qt/201109/W020120130585014685198.pdf).
36	The instruments for $PM_{2.5}$ measurements were tested using the reference method by at
37	least three samples based on HJ 618 specifications. The instruments used for $SO_2$ and
38	NO2 measurement at each site were tested for zero and scale noises, error of indication,
39	zero and span drifts, etc.
40	Along with the acid gases, surface NH <sub>3</sub> concentrations over China for the 2008–
41	2016 period (the current availability) was extracted from the study of Liu et al. (2019),
42	which were estimated using IASI (the Infrared Atmospheric Sounding Interferometer)
43	NH <sub>3</sub> retrievals and NH <sub>3</sub> vertical profiles (Fig. S9). Although the satellite-derived surface
44	NH <sub>3</sub> concentrations are described in detail by Liu et al. (2019), a brief summary is given
45	here for the reader's convenience. The NH3 total columns were derived from the IASI-
46	A instrument (aboard the MetOp-A platform) morning overpass observations (i.e.,

47	09:30 local time at the Equator during overpass), which have a circular footprint of 12
48	km diameter at nadir and an ellipsoid shaped footprint of up to 20 km $\times$ 39 km at the
49	maximum diameter (Van Damme et al., 2018). The IASI NH <sub>3</sub> datasets are the ANNI-
50	NH <sub>3</sub> -v2.2R-I retrieval product, which was developed by converting hyperspectral range
51	index data to NH <sub>3</sub> columns using an Artificial Neural Network for IASI (ANNI)
52	algorithm (Whitburn et al., 2016). The NH <sub>3</sub> vertical profiles were simulated from the
53	Goddard Earth Observing System-Chemistry (GEOS-Chem) atmospheric transport
54	model considering H <sub>2</sub> SO <sub>4</sub> -HNO <sub>3</sub> -NH <sub>3</sub> aerosol thermodynamics mechanisms (Whitburn
55	et al., 2016; Van Damme et al., 2017), and were used to convert the satellite NH <sub>3</sub>
56	columns to surface NH3 concentrations. The satellite NH3 predictions are reliable
57	(average $R^2 = 0.919$ and p<0.001) by validating against the in-situ surface observations
58	on a monthly basis (Liu et al., 2019)
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Figure S1. Spatial distribution of the 1498 monitoring sites (blue dots) and the 218

<sup>74</sup> meta-analysis sites (red dots).



Figure S2. Timeline of policies to improve air quality in China. The green text indicates
the start of control policies from the Chinese government for the pollutant highlighted;
the \* symbol and red text indicates the pollutant emission reduction target in the given
5-year plan. The different background colors denote the three different pollutant history
periods (Periods I, II and III) described in the main text.



**Figure S3.** (a) Simulated and observed monthly mean  $PM_{2.5}$  concentrations (µg m<sup>-3</sup>) for January 2010. The observations are from the ChinaHighAirPollutants (CHAP, https://weijing-rs.github.io/product.html) database. (b) Scatter plots of simulated versus observed monthly means PM<sub>2.5</sub> concentration in the BTH, YRD, PRD, and SCB regions. 



**Figure S4**. Overlay of observed (colored circles) and simulated (color map) monthly mean concentrations of (a)  $SO_4^{2^-}$ , (b)  $NO_3^-$  and (c)  $NH_4^+$  in January 2010. (d) scatter plot of simulated and observed concentrations of  $SO_4^{2^-}$ ,  $NO_3^-$  and  $NH_4^+$ . The dotted lines correspond to the 1:2 and 2:1 lines. The observations are collected from the literature (See Table S5).



Figure S5. Time series of the observed (red dots) and simulated (black line) (a) hourly concentrations of  $PM_{2.5}$  and (b) daily concentrations of  $NO_2$  and  $SO_2$  in January 2010 in Beijing; (c) daily concentrations of  $PM_{2.5}$  during 14-30 January 2010 at monitoring sites in Shangdianzi, Chengdu, Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP-CAS) and Tianjin. The normalized mean bias (NMB) normalized mean error (NME), and correlation coefficient (*R*) are given in the plots.

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Figure S6. Scatter plots of CMAQ simulations versus surface observations for PM<sub>2.5</sub>,
NO<sub>2</sub>, and SO<sub>2</sub> concentrations before the COVID-lockdown (black dots) and during the
COVID-lockdown period (red dots).



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**Figure S7.** The left column shows simulated (shaded) and observed (dot) monthlymean temperature at 2 m above the ground (T2) (a), wind speed (WS) (b), and (c) relative humidity (RH) for January 2010. The right column shows scatterplots of simulated versus observed T2, WS, RH at 400 monitoring sites in China. The value of correlation coefficients (*R*) is presented on each scatterplot.

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- $NH_4^+$  in Quzhou in China during 2002-2019



Figure S9. (a) Spatial patterns of trends between 2008 and 2016 in annual mean
concentrations of NH<sub>3</sub>. (b) Annual average ground-based measured NH<sub>3</sub> concentrations
2008-2016 across all of China and in northern and southern regions. Data for map (a)
are from NH<sub>3</sub> satellite retrievals combining vertical profiles from Goddard Earth
Observing System-Chemistry (GEOS-Chem).



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Figure S10. Simulated PM<sub>2.5</sub> concentrations (in µg m<sup>-3</sup>) without (basic) and with 50% 151 ammonia (NH<sub>3</sub>) emissions reductions in January for the years 2010, 2014, 2017 and 152 2020 in four megacity clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River 153 154 Delta, SCB: Sichuan Basin, PRD: Pearl River Delta). Inset maps indicate the location of each region. \*\* denotes significant difference without and with 50% ammonia 155 emission reductions (P < 0.05). *n* is the number of calculated samples by grid extraction. 156 Error bars are standard errors of means. (Period I (2000–2012), Period II (2013–2016), 157 and Period III (2017-2019); Special control is the restrictions in economic activities 158 159 and associated emissions during the COVID-19 lockdown period in 2020.) 160



Figure S11. The spatial distributions of simulated SIA concentrations (in  $\mu$ g m<sup>-3</sup>) without (a) and with (b) 50% ammonia emissions reduction for the years 2010, 2014, 2017 and 2020. The % decreases in SIA concentrations in each year for the simulations with the emissions reductions are shown in row (c). (Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions in economic activities and associated emissions during the COVID-19 lockdown period in 2020.)

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Figure S12. The spatial distributions of simulated PM<sub>2.5</sub> concentrations (in μg m<sup>-3</sup>)
without (a) and with (b) 50% ammonia emissions reduction for the years 2010, 2014,
2017 and 2020. The % decreases in PM<sub>2.5</sub> concentrations in each year for the
simulations with the emissions reductions are shown in row (c). (Period I (2000–2012),
Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions
in economic activities and associated emissions during the COVID-19 lockdown period
in 2020.)



Figure S13. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 191 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Spring, Summer, Fall, and Winter 192 during 2000-2019. Bars with different letters denote significant differences among the 193 three periods (P <0.05) (upper and lowercase letters for non-hazy and hazy days, 194 respectively). The upper and lower boundaries of the boxes represent the 75th and 25th 195 percentiles; the line within the box represents the median value; the whiskers above and 196 below the boxes represent the 90th and 10th percentiles; the point within the box 197 represents the mean value. Comparison of the pollutants among the three-periods using 198 Kruskal-Wallis and Dunn's test. The n represents independent sites; more detail on this 199 is presented in Section 2.2. 200



Figure S14. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 202 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Spring in Period I (2000–2012), 203 Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 204 significant differences among the three periods (P < 0.05) (upper and lowercase letters 205 for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 206 represent the 75th and 25th percentiles; the line within the box represents the median 207 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; 208 the point within the box represents the mean value. Comparison of the pollutants among 209 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 210 sites; more detail on this is presented in Section 2.2. 211



213 Figure S15. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Summer in Period I (2000–2012), 214 Period II(2013–2016), and Period III(2017–2019). Bars with different letters denote 215 216 significant differences among the three periods (P < 0.05) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 217 represent the 75th and 25th percentiles; the line within the box represents the median 218 219 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among 220 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 221 sites; more detail on this is presented in Section 2.2. 222



Figure S16. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 224 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Fall in Period I (2000–2012), Period 225 II (2013-2016), and Period III (2017-2019). Bars with different letters denote 226 227 significant differences among the three periods (P < 0.05) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 228 represent the 75th and 25th percentiles; the line within the box represents the median 229 230 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among 231 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 232 sites; more detail on this is presented in Section 2.2. 233



Figure S17. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 235 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Winter in Period I (2000–2012), 236 Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 237 significant differences among the three periods (P < 0.05) (upper and lowercase letters 238 for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 239 represent the 75th and 25th percentiles; the line within the box represents the median 240 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; 241 the point within the box represents the mean value. Comparison of the pollutants among 242 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 243 sites; more detail on this is presented in Section 2.2. 244

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Figure S18. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 247 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in Urban and Rural sites during 2000-248 2019. Bars with \*\* denote significant differences among the three periods (P < 0.05) 249 250 (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes represent the 75th and 25th percentiles; the line within 251 the box represents the median value; the whiskers above and below the boxes represent 252 253 the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among the three-periods using Kruskal-Wallis and Dunn's 254 test. The *n* represents independent sites; more detail on this is presented in Section 2.2. 255



Figure S19. Comparisons of observed concentrations of (a)  $PM_{2.5}$ , (b)  $SO_4^{2-}$ , (c)  $NO_3^{-}$ , 257 and (d) NH<sub>4</sub><sup>+</sup> between non-hazy and hazy days in urban sites in Period I (2000–2012), 258 Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 259 260 significant differences among the three periods (P < 0.05) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 261 represent the 75th and 25th percentiles; the line within the box represents the median 262 263 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among 264 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 265 sites; more detail on this is presented in Section 2.2. 266



268 and (d)  $NH_4^+$  between non-hazy and hazy days in rural sites in Period I (2000–2012), 269 Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 270 271 significant differences among the three periods (P < 0.05) (upper and lowercase letters for non-hazy and hazy days, respectively). The upper and lower boundaries of the boxes 272 represent the 75th and 25th percentiles; the line within the box represents the median 273 274 value; the whiskers above and below the boxes represent the 90th and 10th percentiles; the point within the box represents the mean value. Comparison of the pollutants among 275 the three-periods using Kruskal-Wallis and Dunn's test. The n represents independent 276 sites; more detail on this is presented in Section 2.2. 277

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Figure S21. Overlay of observed (colored circles) and simulated (color map) monthly

concentrations of PM<sub>2.5</sub> in January 2014 and 2017.

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**Table S1.** Summary of number of measurment sites from different databases assembled from peer-reviewed publications and used for analyses in the present study of  $PM_{2.5}$  component concentrations, NS, NSN, SOR, and NOR. The details of information in Supporting Material Dataset. NS is the slope of the regression equation between  $[NH_4^+]$  and  $[SO_4^{2-}]$ , NSN is the slope of the regression equation between  $[NH_4^+]$  and  $[SO_4^{2-}]$ , SOR is sulfur oxidation ratio, and NOR is nitrogen oxidation ratio.

	No. of	N <sub>a</sub> a	
	measurement sites	$IVf_S$	$5n + 10^{b}$
OC	84	531290	430
EC	101	396171	515
$SO_4^{2-}$	151	2385388	765
NO <sub>3</sub> -	175	4542962	885
Cl-	105	886197	535
F⁻	26	1587	140
$\mathrm{NH_4^+}$	166	4026300	840
$Na^+$	82	343318	420
$\mathbf{K}^+$	91	523882	465
$Ca^{2+}$	75	88883	385
$Mg^{2+}$	68		350
NS	145	—	735
NSN	144		730
SOR	38		200
NOR	33		175

Note: <sup>a</sup>  $N_{f_s}$  is Rosenberg's fail safe-numbers, calculated to assess the robustness of

findings on  $PM_{2.5}$ . <sup>b</sup> *n* is the number of sites.

	2010	2014	2017	2020	2014-2010	2017-2014	2020-2017
	Tonne	Tonne	Tonne	Tonne	%	%	%
Beijing	20410	10899	4051	2998	-47	-63	-26
Tianjin	29000	22111	9042	7233	-24	-59	-20
Hebei	183194	146125	70877	59536	-20	-51	-16
Shanxi	193721	151346	111566	89253	-22	-26	-20
Inner	158304	121932	66986	56938	-23	-45	-15
Mongolia	0.000		110.10	00551	21	47	•
Liaoning	97766	77459	41043	29551	-21	-47	-28
Jilin	44399	34995	23713	18259	-21	-32	-23
Heilongjiang	44491	43441	28536	20832	-2	-34	-27
Shanghai	43016	28112	14390	8346	-35	-49	-42
Jiangsu	113216	69162	27388	20267	-39	-60	-26
Zhejiang	52789	35704	17846	12671	-32	-50	-29
Anhui	43583	30433	15703	12248	-30	-48	-22
Fujian	37907	19804	13537	9476	-48	-32	-30
Jiangxi	40179	28746	14362	11346	-28	-50	-21
Shandong	244765	178189	84499	63374	-27	-53	-25
Henan	125492	72270	34617	27002	-42	-52	-22
Hubei	182208	112715	69204	53287	-38	-39	-23
Hunan	92142	90407	68003	51002	-2	-25	-25
Guangdong	75644	46140	35595	23849	-39	-23	-33
Guangxi	68551	43141	22565	16247	-37	-48	-28
Hainan	4008	4790	3933	2950	20	-18	-25
Chongqing	120968	67877	35101	23868	-44	-48	-32
Sichuan	113414	75375	37241	27186	-34	-51	-27
Guizhou	191009	181314	111426	83569	-5	-39	-25
Yunnan	66724	54142	33106	24830	-19	-39	-25
Tibet	60	66	97	82	10	47	-15
Shaanxi	105817	76442	40069	32856	-28	-48	-18
Gansu	38708	23976	19749	16590	-38	-18	-16
Qinghai	4778	5594	4310	3362	17	-23	-22
Ningxia	28415	24767	20062	15247	-13	-19	-24
Xinjiang	44162	45561	24929	21190	3	-45	-15
China	2608842	1923034	1103546	845445	-26	-43	-23

Table S2. Anthropogenic emissions of SO2 in January of 2010, 2014, 2017 and 2020,

Note:  $SO_2$  emissions were provided by the Multi-resolution Emission Inventory (MEIC)

(http://meicmodel.org) for the years 2010, 2014 and 2017. The SO<sub>2</sub> emissions of 2020

are based on 2017 MEIC as a case of special control following Huang et al. (2021) approach.

	2010	2014	2017	2020	2014-2010	2017-2014	2020-2017
	Tonne	Tonne	Tonne	Tonne	%	%	%
Beijing	32325	27223	24931	13712	-16	-8	-45
Tianjin	33978	37380	30435	18870	10	-19	-38
Hebei	177625	167812	148367	81602	-6	-12	-45
Shanxi	106872	95243	82741	49645	-11	-13	-40
Inner Mongolia	129645	120068	111328	79043	-7	-7	-29
Liaoning	113719	112970	104711	62826	-1	-7	-40
Jilin	61173	58140	60342	36808	-5	4	-39
Heilongjiang	77226	81565	74725	47077	6	-8	-37
Shanghai	45395	32961	31539	16400	-27	-4	-48
Jiangsu	153102	142730	131740	65870	-7	-8	-50
Zhejiang	95531	75644	71440	35720	-21	-6	-50
Anhui	86796	87662	78304	34454	1	-11	-56
Fujian	47505	41396	46573	22821	-13	13	-51
Jiangxi	39804	39120	34918	16411	-2	-11	-53
Shandong	222442	201757	177591	88796	-9	-12	-50
Henan	137270	126230	105735	45466	-8	-16	-57
Hubei	76893	69558	59338	26702	-10	-15	-55
Hunan	67695	61721	56416	27644	-9	-9	-51
Guangdong	109844	87421	86116	43058	-20	-1	-50
Guangxi	47006	42915	35959	17980	-9	-16	-50
Hainan	6813	7437	7689	4306	9	3	-44
Chongqing	37763	36995	32855	15442	-2	-11	-53
Sichuan	82543	80131	69170	34585	-3	-14	-50
Guizhou	50554	43218	33805	20621	-15	-22	-39
Yunnan	52995	42479	36285	17779	-20	-15	-51
Tibet	2428	2337	3625	2357	-4	55	-35
Shaanxi	58296	56807	48598	26729	-3	-14	-45
Gansu	37634	31398	28059	14871	-17	-11	-47
Qinghai	7872	10535	8907	4810	34	-15	-46
Ningxia	23645	27323	27936	17879	16	2	-36
Xinjiang	42625	62771	48156	31301	47	-23	-35
China	2265015	2110946	1898332	1021583	-7	-10	-46

Table S3. Anthropogenic emissions of NO<sub>x</sub> in January of 2010, 2014, 2017 and 2020,

Note: NO<sub>x</sub> emissions were provided by the Multi-resolution Emission Inventory (MEIC)

(http://meicmodel.org) for the years 2010, 2014 and 2017. The NO<sub>x</sub> emissions of 2020

are based on 2017 MEIC as a case of special control following Huang et al. (2021) approach.

Table S4. Control options for NH<sub>3</sub> emissions reductions with their corresponding

Abatement option	Application processes	Reduction efficiency
Avoiding over-fertilization	Synthetic fertilizer application	>20%
Deep application of fertilizers	Synthetic fertilizer application	~50%
Low crude protein feed	Whole manure management chain	10-40%
Using deep litter in floor and regular washing	Manure in house	20-50%
Covering solid and slurry manure	Manure storage	>60%
Incorporation or plough after spreading	Field application of manure	40-80%
All	NH3 emissions for all China	30-50%

estimated percentage emissions reductions (reduction efficiency).

Note: The NH<sub>3</sub> emissions control options and corresponding emissions reduction

efficiency are from Liu et al. (2019). The feasible control options can reduce China's

NH<sub>3</sub> emissions by 30-50% based on the PKU-NH<sub>3</sub> emission model.

						<b>N 11 T</b>	
ID	City	Lat	Lon	<b>SO</b> <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> -	$\mathbf{NH}_{4}^{+}$	Reference
	City	Lat	LOII	$(\mu g m^{-3})$	$(\mu g m^{-3})$	$(\mu g m^{-3})$	Kelefenee
1	Guangzhou	113.4	27.1	17.8	13	6.5	(Tao et al., 2014)
2	Beijing	116.3	39.9	8.5	7.3	4.7	(Zhang et al., 2012)
3	Beijing	116.3	40.0	8.5	7.3	4.5	(Zhang et al., 2012)
4	Beijing	116.4	40.0	14.23	17.09	5.21	(Cao et al., 2014)
5	Beijing	116.7	40.9	6.64	8.84	2.83	(Zhang et al., 2012)
6	Guangzhou	113.5	23.2	17.8	13	3.3	(Tao et al., 2014)
7	Xiamen	118.1	24.6	17.67	13.15	9.17	(Zhang et al., 2012)
8	Beijing	116.4	40.0	15.8	15.9	8.2	(Pan et al., 2012)
9	Baoding	115.5	38.9	37.6	24.0	16.3	(Pan et al., 2012)
10	Tangshan	118.2	39.6	22.7	20.1	20.8	(Pan et al., 2012)
11	Tianjin	117.2	39.1	20.0	17.9	6.6	(Pan et al., 2012)
12	Xinglong	117.6	40.4	31.5	28.0	17.2	(Pan et al., 2012)

**Table S5.** Monthly mean concentration of  $SO_4^{2-}$ ,  $NO_3^{-}$ , and  $NH_4^+$  in January in 2010.

**Table S6** Simulated SIA concentrations ( in  $\mu$ g m<sup>-3</sup>) with (basic) and 50% ammonia (NH<sub>3</sub>) emissions reductions in January for years 2010, 2014, 2017, and 2020 in four megacity clusters.

	2010 (Period I)		2014 (Period II)		20 (Perio	2017 (Period III)		2020 (Special control)	
	Base	50%NH3	Base	50%NH <sub>3</sub>	Base	50%NH <sub>3</sub>	Base	50%NH <sub>3</sub>	
BTH	29.9±1.2	24.0±1.1	29.9±1.2	24.4±1.1	27.8±1.1	23.1±1.0	21.6±0.8	19.6±0.8	
YRD	42.7±0.9	31.6±0.8	41.5±0.9	31.1±0.8	37.8±0.9	28.8±0.8	26.9±0.5	22.6±0.5	
SCB	57.8±1.2	43.5±1.1	52.9±1.0	41.4±1.0	44.5±0.8	35.9±0.8	28.8±0.5	25.2±0.5	
PRD	13.9±0.5	10.0±0.3	11.9±0.4	8.7±0.3	10.3±0.4	7.5±0.3	7.2±0.2	5.9±0.2	

Note: The value is mean ± standard errors of means. (Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019); Special control is the restrictions in economic activities and associated emissions during the COVID-19 lockdown period in 2020. BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta).

Sector	$SO_2$	NO <sub>x</sub>
Electric	30	31
Industry - building materials	45	59
Industry - boiler	24	7
Industry - steel		3
Building	2	

**Table S7.** The effectiveness of potential end-of pipe controls on  $SO_2$  and  $NO_x$  emissionsreductions for different production sectors (unit: %).

Note: The effectiveness of potential end-of pipe controls on  $SO_2$  and  $NO_x$  emissions reductions for different production sectors from Xing et al. (2021). GetData Graph Digitizer (Version 2.25, http://www.getdatagraph-digitizer.com) was used to digitize the % effectiveness of  $SO_2$  and  $NO_x$  from figures.

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