Supplement of

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

Fanlei Meng et al.

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S1: Method 1: 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:

$$SOR = \frac{nSO_4^{2-}}{(nSO_4^{2-} + nSO_2)} \quad (1)$$

$$NOR = \frac{nNO_3^-}{(nNO_3^- + nNO_2)} \quad (2)$$

To identify whether acidic species are fully neutralized by NH$_3$ in PM$_{2.5}$, we selected two indicators: the slope of the linear regression between equivalent concentrations of [NH$_4^+$] and [SO$_4^{2-}$] and the slope of the linear regression between equivalent concentrations of [NH$_4^+$] and [SO$_4^{2-}$ + NO$_3^-$] (Sun et al., 2006). In the atmosphere, NH$_3$ is first taken up by sulfuric acid to form ammonium sulfate salts and then any excess NH$_3$ 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$_3$ is not completely neutralized with sulfuric acid and nitric acid, suggesting a NH$_3$-limited environment. Slopes of NS and NSN greater than 1 indicate that they reacted completely, suggesting a NH$_3$-rich environment (Xu et al., 2017).

S2: Method 2: 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
balance method (TEOM from Rupprecht & Patashnick Co., Inc., USA) or the β absorption method (BAM 1020 from Met One Instrument Inc., USA; Tianhong Co., China or Xianhe Co., China). Concentrations of SO\(_2\) were measured using either UV-spectrophotometry (TEI Model 49i from Thermo Fisher Scientific Inc., USA) or Ultraviolet Fluorescence (TEI Model 43i from Thermo Fisher Scientific Inc., USA) methods, and concentrations of NO\(_2\) using the chemiluminescence (TEI Model 42i from Thermo Fisher Scientific Inc., USA) method. The detection limits (DL) of these techniques are sufficient to measure accurately the high or relatively high concentration of PM\(_{2.5}\), SO\(_2\), and NO\(_2\) at all monitoring sites.

Data from all monitoring sites were automatically released to an open website after validation using HJ630-2011 specifications (http://kjs.mep.gov.cn/hjbhbz/bzwb/other/qt/201109/W020120130585014685198.pdf). The instruments for PM\(_{2.5}\) measurements were tested using the reference method by at least three samples based on HJ 618 specifications. The instruments used for SO\(_2\) and NO\(_2\) measurement at each site were tested for zero and scale noises, error of indication, zero and span drifts, etc.

Along with the acid gases, surface NH\(_3\) concentrations over China for the 2008–2016 period (the current availability) was extracted from the study of Liu et al. (2019), which were estimated using IASI (the Infrared Atmospheric Sounding Interferometer) NH\(_3\) retrievals and NH\(_3\) vertical profiles (Fig. S9). Although the satellite-derived surface NH\(_3\) concentrations are described in detail by Liu et al. (2019), a brief summary is given here for the reader’s convenience. The NH\(_3\) total columns were derived from the IASI-A instrument (aboard the MetOp-A platform) morning overpass observations (i.e.,
09:30 local time at the Equator during overpass), which have a circular footprint of 12 km diameter at nadir and an ellipsoid shaped footprint of up to 20 km × 39 km at the maximum diameter (Van Damme et al., 2018). The IASI NH₃ datasets are the ANNI-NH₃-v2.2R-I retrieval product, which was developed by converting hyperspectral range index data to NH₃ columns using an Artificial Neural Network for IASI (ANNI) algorithm (Whitburn et al., 2016). The NH₃ vertical profiles were simulated from the Goddard Earth Observing System-Chemistry (GEOS-Chem) atmospheric transport model considering H₂SO₄-HNO₃-NH₃ aerosol thermodynamics mechanisms (Whitburn et al., 2016; Van Damme et al., 2017), and were used to convert the satellite NH₃ columns to surface NH₃ concentrations. The satellite NH₃ predictions are reliable (average $R^2 = 0.919$ and $p<0.001$) by validating against the in-situ surface observations on a monthly basis (Liu et al., 2019)
Figure S1. Spatial distribution of the 1498 monitoring sites (blue dots) and the 218 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$^{-3}$) 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$_{2.5}$ 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) $\text{SO}_4^{2-}$, (b) $\text{NO}_3^-$ and (c) $\text{NH}_4^+$ in January 2010. (d) scatter plot of simulated and observed concentrations of $\text{SO}_4^{2-}$, $\text{NO}_3^-$ and $\text{NH}_4^+$. 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.
Figure S6. Scatter plots of CMAQ simulations versus surface observations for PM$_{2.5}$, NO$_2$, and SO$_2$ concentrations before the COVID-lockdown (black dots) and during the COVID-lockdown period (red dots).
Figure S7. The left column shows simulated (shaded) and observed (dot) monthly-mean 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.
Figure S8. Daily and monthly concentration of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) 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$_3$. (b) Annual average ground-based measured NH$_3$ concentrations 2008-2016 across all of China and in northern and southern regions. Data for map (a) are from NH$_3$ satellite retrievals combining vertical profiles from Goddard Earth Observing System-Chemistry (GEOS-Chem).
Figure S10. Simulated PM$_{2.5}$ concentrations (in $\mu g \text{ m}^{-3}$) without (basic) and with 50% ammonia (NH$_3$) emissions reductions in January for the years 2010, 2014, 2017 and 2020 in four megacity clusters (BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SCB: Sichuan Basin, PRD: Pearl River Delta). Inset maps indicate the location of each region. ** denotes significant difference without and with 50% ammonia emission reductions ($P < 0.05$). $n$ is the number of calculated samples by grid extraction. Error bars are 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.)
Figure S11. The spatial distributions of simulated SIA concentrations (in μg m⁻³) 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.)
**Figure S12.** The spatial distributions of simulated PM$_{2.5}$ concentrations (in $\mu$g m$^{-3}$) without (a) and with (b) 50% ammonia emissions reduction for the years 2010, 2014, 2017 and 2020. The % decreases in PM$_{2.5}$ 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$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Spring, Summer, Fall, and Winter during 2000-2019. Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three-periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S14. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Spring in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S15. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Summer in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S16. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Fall in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S17. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Winter in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S18. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in Urban and Rural sites during 2000-2019. Bars with ** denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three-periods using Kruskal-Wallis and Dunn’s test. The $n$ represents independent sites; more detail on this is presented in Section 2.2.
**Figure S19.** Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in urban sites in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three-periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
Figure S20. Comparisons of observed concentrations of (a) PM$_{2.5}$, (b) SO$_4^{2-}$, (c) NO$_3^-$, and (d) NH$_4^+$ between non-hazy and hazy days in rural sites in Period I (2000–2012), Period II (2013–2016), and Period III (2017–2019). Bars with different letters denote 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 represent the 75th and 25th percentiles; the line within the box represents the median 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 the three periods using Kruskal-Wallis and Dunn’s test. The n represents independent sites; more detail on this is presented in Section 2.2.
**Figure S21.** Overlay of observed (colored circles) and simulated (color map) monthly concentrations of PM$_{2.5}$ in January 2014 and 2017.
Table S1. Summary of number of measurement 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$_4^{2-}$ + NO$_3^-$], SOR is sulfur oxidation ratio, and NOR is nitrogen oxidation ratio.

| Compounds | No. of measurement sites | $N_{fs}$ | $5n + 10$ |
|-----------|--------------------------|----------|-----------|
| OC        | 84                       | 531290   | 430       |
| EC        | 101                      | 396171   | 515       |
| SO$_4^{2-}$ | 151                  | 2385388  | 765       |
| NO$_3^-$  | 175                      | 4542962  | 885       |
| Cl$^-$    | 105                      | 886197   | 535       |
| F         | 26                       | 1587     | 140       |
| NH$_4^+$  | 166                      | 4026300  | 840       |
| Na$^+$    | 82                       | 343318   | 420       |
| 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: $^a$ $N_{fs}$ is Rosenberg's fail safe-numbers, calculated to assess the robustness of findings on PM$_{2.5}$. $^b$ $n$ is the number of sites.
Table S2. Anthropogenic emissions of SO\textsubscript{2} in January of 2010, 2014, 2017 and 2020, and the percentage decreases in SO\textsubscript{2} emissions between successive pairs of years.

|            | 2010  | 2014  | 2017  | 2020  | 2014-2010 | 2017-2014 | 2020-2017 |
|------------|-------|-------|-------|-------|-----------|-----------|-----------|
| Beijing    | 20410 | 10899 | 4051  | 2998  | -47       | -63       | -26       |
| Tianjin    | 29000 | 22111 | 9042  | 7233  | -24       | -59       | -20       |
| Hebei      | 18349 | 146125| 70877 | 59536 | -20       | -51       | -16       |
| Shanxi     | 193721| 151346| 111566| 89253 | -22       | -26       | -20       |
| Inner Mongolia | 158304| 121932| 66986 | 56938 | -23       | -45       | -15       |
| 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       |

Note: SO\textsubscript{2} emissions were provided by the Multi-resolution Emission Inventory (MEIC) (http://meicmodel.org) for the years 2010, 2014 and 2017. The SO\textsubscript{2} emissions of 2020...
are based on 2017 MEIC as a case of special control following Huang et al. (2021) approach.
Table S3. Anthropogenic emissions of NO\(_x\) in January of 2010, 2014, 2017 and 2020, and the percentage decreases in NO\(_x\) emissions between successive pairs of years.

| Region        | 2010       | 2014       | 2017       | 2020       | 2014-2010 | 2017-2014 | 2020-2017 |
|---------------|------------|------------|------------|------------|-----------|-----------|-----------|
|               | Ton        | Ton        | Ton        | Ton        | %         | %         | %         |
| 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      | 65958      | 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       |

Note: NO\(_x\) emissions were provided by the Multi-resolution Emission Inventory (MEIC) ([http://meicmodel.org](http://meicmodel.org)) for the years 2010, 2014 and 2017. The NO\(_x\) 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 NH₃ emissions reductions with their corresponding estimated percentage emissions reductions (reduction efficiency).

| 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%               |

Note: The NH₃ emissions control options and corresponding emissions reduction efficiency are from Liu et al. (2019). The feasible control options can reduce China’s NH₃ emissions by 30-50% based on the PKU-NH₃ emission model.
Table S5. Monthly mean concentration of $\text{SO}_4^{2-}$, $\text{NO}_3^-$, and $\text{NH}_4^+$ in January in 2010.

| ID | City     | Lat | Lon | $\text{SO}_4^{2-}$ ($\mu\text{g m}^{-3}$) | $\text{NO}_3^-$ ($\mu\text{g m}^{-3}$) | $\text{NH}_4^+$ ($\mu\text{g m}^{-3}$) | Reference          |
|----|----------|-----|-----|------------------------------------------|----------------------------------------|------------------------------------------|--------------------|
| 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 S6 Simulated SIA concentrations (in μg m$^{-3}$) with (basic) and 50% ammonia (NH$_3$) emissions reductions in January for years 2010, 2014, 2017, and 2020 in four megacity clusters.

|        | 2010 (Period I) | 2014 (Period II) | 2017 (Period III) | 2020 (Special control) |
|--------|-----------------|------------------|-------------------|------------------------|
|        | Base 50%NH$_3$  | Base 50%NH$_3$   | Base 50%NH$_3$   | Base 50%NH$_3$         |
| 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).
Table S7. The effectiveness of potential end-of pipe controls on SO$_2$ and NO$_x$ emissions reductions for different production sectors (unit: %).

| Sector                  | SO$_2$ | NO$_x$ |
|-------------------------|--------|--------|
| Electric                | 30     | 31     |
| Industry - building materials | 45     | 59     |
| Industry - boiler       | 24     | 7      |
| Industry - steel        | —      | 3      |
| Building                | 2      | —      |

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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