

Table S1. China's historical emissions for 2016 and future emission scenario SSP1-RCP2.6-BHE for 2030 and 2050 provided by Tong et al. (2020)

| Unit: Tg        | 2016 | 2030 | 2050 | 2030/2016 | 2050/2016 |
|-----------------|------|------|------|-----------|-----------|
| BC              | 1.3  | 0.49 | 0.28 | 0.38      | 0.22      |
| SO <sub>2</sub> | 13   | 5.9  | 2.3  | 0.44      | 0.17      |
| NO <sub>x</sub> | 23   | 10   | 6.1  | 0.46      | 0.27      |
| OC              | 2.3  | 1.2  | 0.66 | 0.50      | 0.29      |
| VOC             | 28   | 20   | 13   | 0.70      | 0.45      |
| PM2.5           | 8.1  | 3.9  | 2.0  | 0.48      | 0.24      |
| NH <sub>3</sub> | 10   | 8.5  | 6.1  | 0.85      | 0.61      |
| CO              | 142  | 92   | 69   | 0.65      | 0.49      |

Table S2. Information of observation sites for sulfate, nitrate, and black carbon concentrations used for model evaluation

| Site            | Species             | Year           | Lon (°)       | Lat (°)     | References  |
|-----------------|---------------------|----------------|---------------|-------------|---|
| Beijing         | Sulfate,<br>Nitrate | 2006/2008/2010 | 116.33-116.37 | 39.95-39.99 | Zhang et al. (2013), Hu et al. (2016), Sun et al. (2010), Huang et al. (2010) |
| Beijing         | Sulfate,<br>Nitrate | 2016           | 116.32        | 39.95       | Liu et al. (2018)   |
| Beijing         | BC                  | 2008-2016      | 116.37        | 39.97       | Xia et al. (2020)   |
| Zhengzhou       | Sulfate,<br>Nitrate | 2010           | 113.52        | 34.8        | Geng et al. (2013)  |
| Tianjin         | Sulfate<br>Nitrate  | 2008           | 117.17        | 39.01       | Gu et al. (2013)  |
| Nanjing         | Sulfate             | 2015           | 118.71-118.75 | 32.01-32.21 | Ge et al. (2017), Wang et al. (2016), Zhang et al. (2017)                     |
| Hangzhou        | Sulfate             | 2016           | 120.21        | 30.21       | Li et al. (2018)  |
| Jiaxing         | Sulfate             | 2010           | 120.8         | 30.8        | Huang et al. (2013)   |
| Shanghai        | Sulfate,<br>Nitrate | 2011-2014      | 121.53        | 31.23       | Tao et al. (2017)   |
| Shanghai        | BC                  | 2008-2016      | 121.59        | 31.18       | Wei et al. (2020)   |
| Fukue<br>Island | BC                  | 2010-2016      | 128.68        | 32.75       | Kanaya et al. (2019)  |

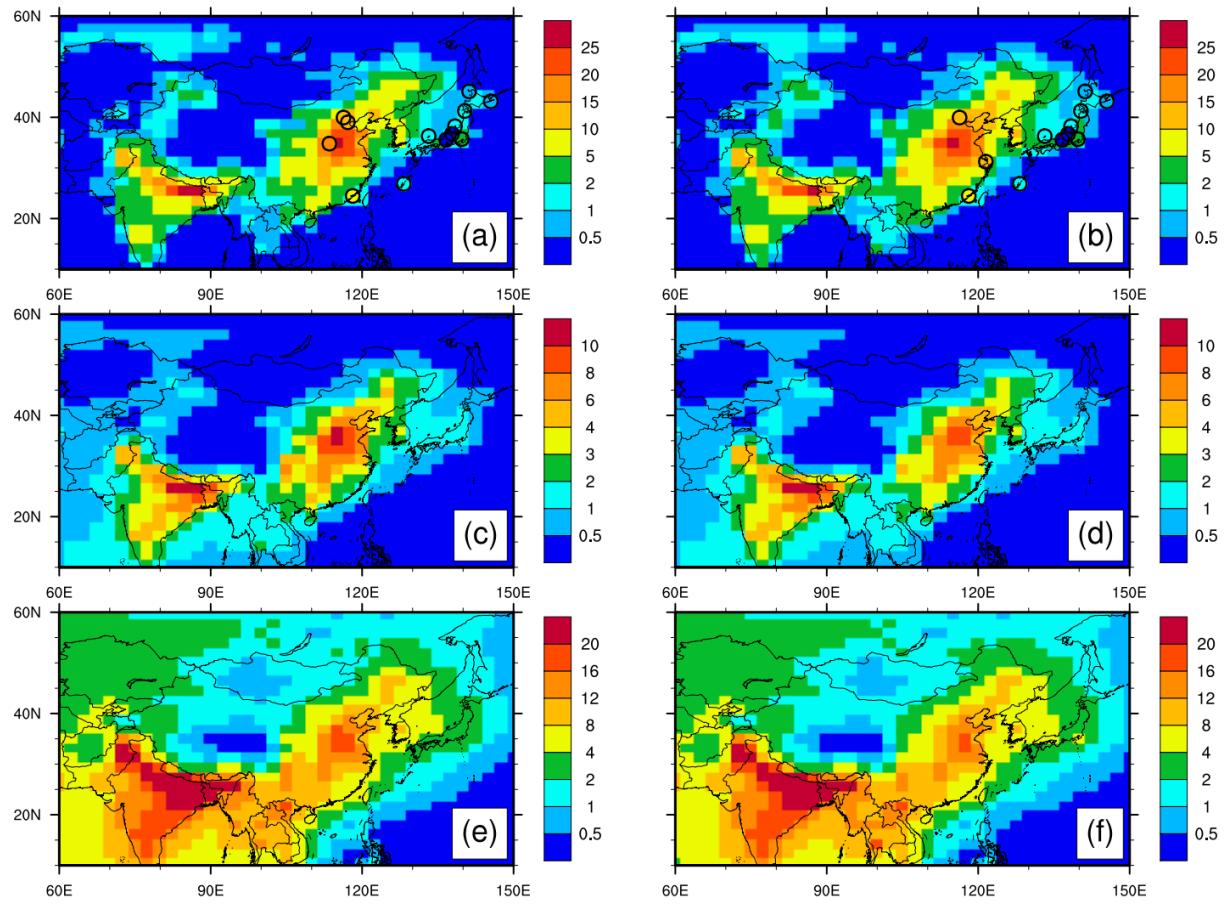


Fig. S1 Simulated annual mean surface concentrations (unit:  $\mu\text{g m}^{-3}$ ) of PM<sub>2.5</sub> nitrate (a-b), ammonium (c-d), and organic aerosols (e-f) between 2008 (left) and 2016 (right). Measurements of nitrate concentrations are shown in colored dots for comparison.

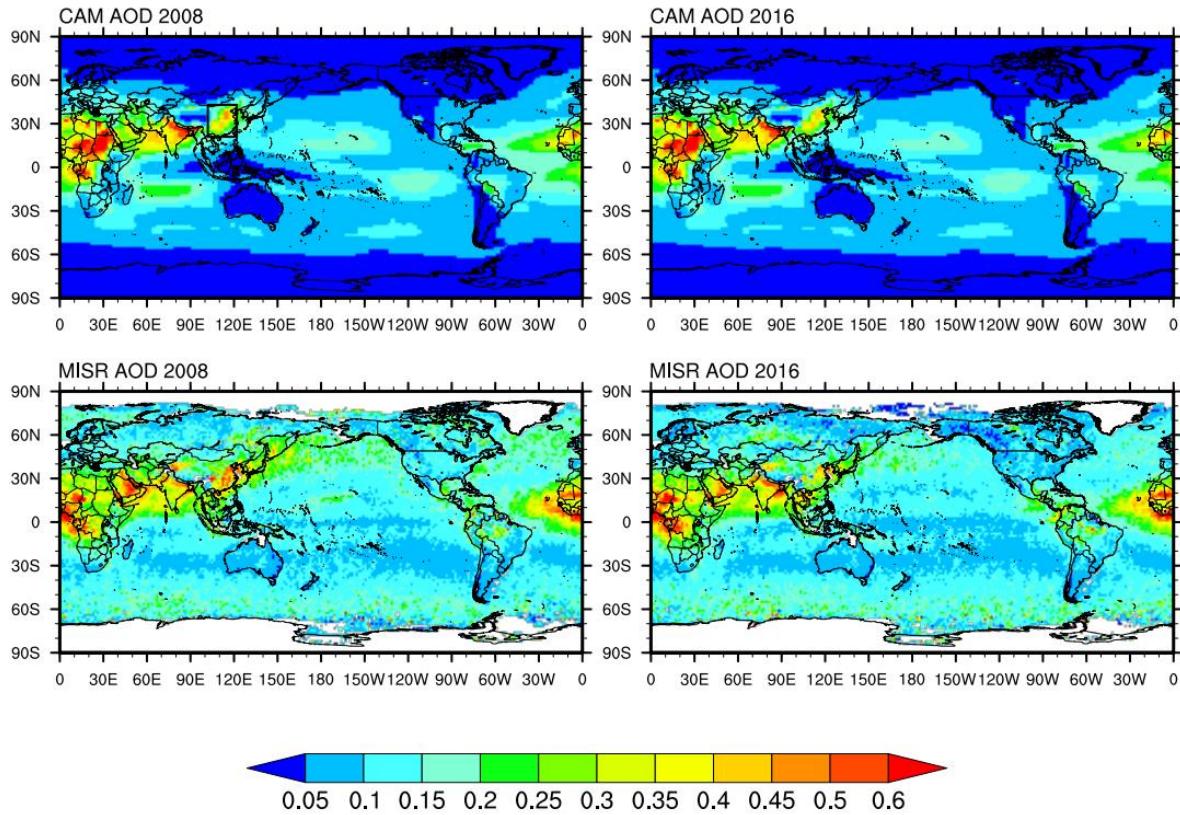


Fig. S2 Comparison of modeled AOD 550 nm with MISR observations in 2008 and 2016. Note that the CAM AOD in 2016 is derived from the simulation using the emission year of 2016 for China.

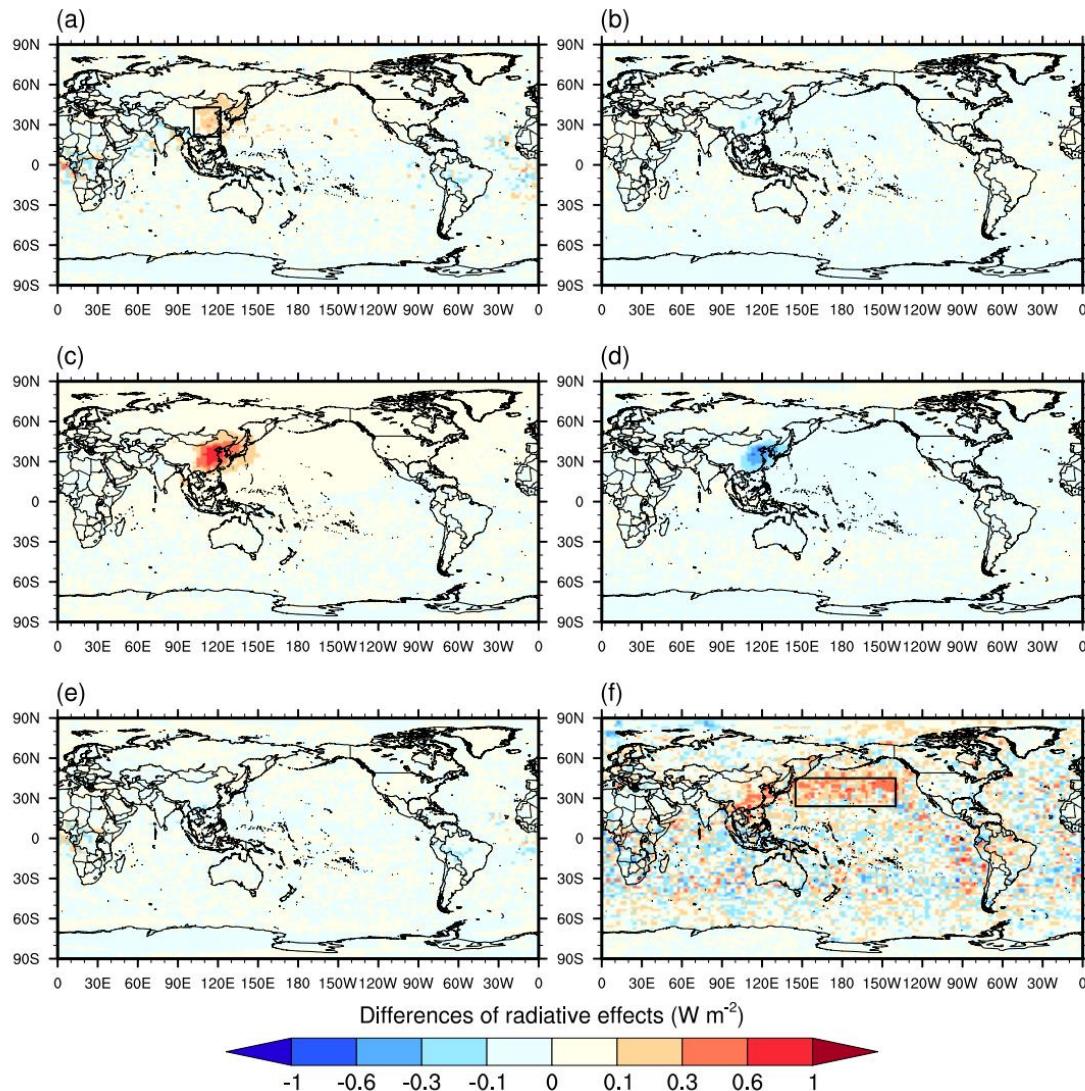


Fig. S3. DRF from total aerosols (a), BC (b), sulfate (c), nitrate + ammonium (d), and OA + dust (e), and aerosol-induced clouds effects (f), due to the reduction in  $\text{SO}_2$  emissions in China.

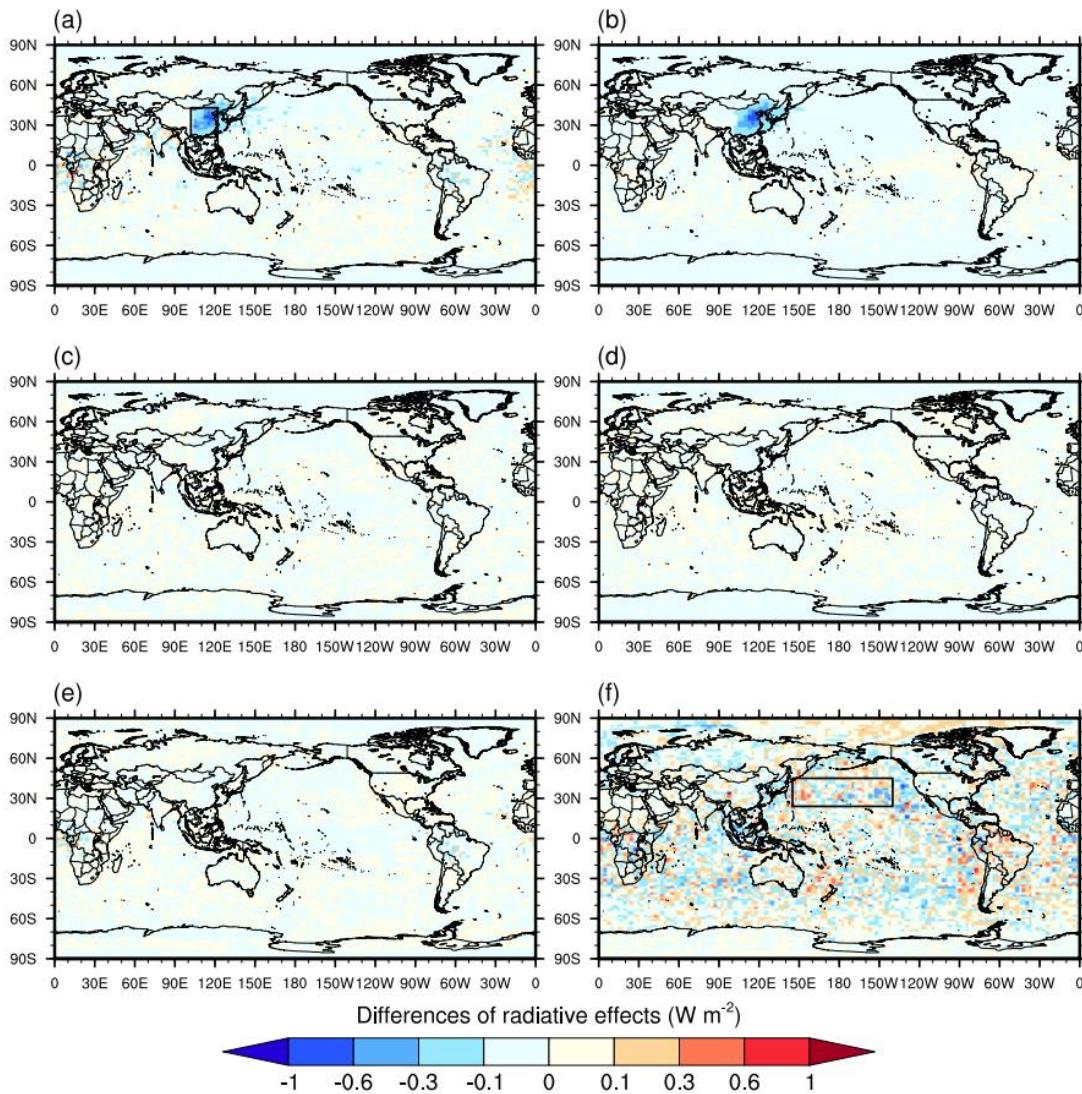


Fig. S4 The same with Fig. S3, but due to the reduction in BC emissions.

## References

- Ge, X., He, Y., Sun, Y., Xu, J., Wang, J., Shen, Y., and Chen, M.: Characteristics and Formation Mechanisms of Fine Particulate Nitrate in Typical Urban Areas in China, *Atmosphere*, 8, 10.3390/atmos8030062, 2017.
- Geng, N., Wang, J., Xu, Y., Zhang, W., Chen, C., and Zhang, R.: PM<sub>2.5</sub> in an industrial district of Zhengzhou, China: Chemical composition and source apportionment, *Particuology*, 11, 99-109, <https://doi.org/10.1016/j.partic.2012.08.004>, 2013.

- Gu, J. X., Wu, L. P., Huo, G. Y., Bai, Z. P., Du, S. Y., Liu, A. X., and Xie, Y. Y.: Pollution Character and Source of Water-Soluble Inorganic Ions in PM<sub>2.5</sub> over Tianjin (in Chinese), Environmental Monitoring in China, 29, 30-34, 2013.
- Hu, W., Hu, M., Hu, W., Jimenez, J. L., Yuan, B., Chen, W., Wang, M., Wu, Y., Chen, C., Wang, Z., Peng, J., Zeng, L., and Shao, M.: Chemical composition, sources, and aging process of submicron aerosols in Beijing: Contrast between summer and winter, *J. Geophys. Res.-Atmos.*, 121, 1955-1977, 10.1002/2015JD024020, 2016.
- Huang, X.-F., Xue, L., Tian, X.-D., Shao, W.-W., Sun, T.-L., Gong, Z.-H., Ju, W.-W., Jiang, B., Hu, M., and He, L.-Y.: Highly time-resolved carbonaceous aerosol characterization in Yangtze River Delta of China: Composition, mixing state and secondary formation, *Atmos. Environ.*, 64, 200-207, <https://doi.org/10.1016/j.atmosenv.2012.09.059>, 2013.
- Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L. W., Liu, X. G., Zhang, Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, *Atmos. Chem. Phys.*, 10, 8933-8945, 10.5194/acp-10-8933-2010, 2010.
- Kanaya, Y., Yamaji, K., Miyakawa, T., Taketani, F., Zhu, C., Choi, Y., Komazaki, Y., Ikeda, K., Kondo, Y., and Klimont, Z.: Rapid reduction of black carbon emissions from China: evidence from 2009-2019 observations on Fukue Island, Japan, *Atmos. Chem. Phys. Discuss.*, 2019, 1-28, 10.5194/acp-2019-1054, 2019.
- Li, K., Chen, L., White, S. J., Zheng, X., Lv, B., Lin, C., Bao, Z., Wu, X., Gao, X., Ying, F., Shen, J., Azzi, M., and Cen, K.: Chemical characteristics and sources of PM<sub>1</sub> during the 2016 summer in Hangzhou, *Environ. Pollut.*, 232, 42-54, <https://doi.org/10.1016/j.envpol.2017.09.016>, 2018.
- Liu, M., Huang, X., Song, Y., Xu, T., Wang, S., Wu, Z., Hu, M., Zhang, L., Zhang, Q., Pan, Y., Liu, X., and Zhu, T.: Rapid SO<sub>2</sub> emission reductions significantly increase tropospheric ammonia concentrations over the North China Plain, *Atmos. Chem. Phys.*, 18, 17933-17943, 10.5194/acp-18-17933-2018, 2018.
- Sun, J., Zhang, Q., Canagaratna, M. R., Zhang, Y., Ng, N. L., Sun, Y., Jayne, J. T., Zhang, X., Zhang, X., and Worsnop, D. R.: Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer, *Atmos. Environ.*, 44, 131-140, <https://doi.org/10.1016/j.atmosenv.2009.03.020>, 2010.
- Tao, J., Zhang, L., Cao, J., and Zhang, R.: A review of current knowledge concerning PM<sub>2.5</sub> chemical composition, aerosol optical properties and their relationships across China, *Atmos. Chem. Phys.*, 17, 9485-9518, 10.5194/acp-17-9485-2017, 2017.
- Tong, D., Cheng, J., Liu, Y., Yu, S., Yan, L., Hong, C., Qin, Y., Zhao, H., Zheng, Y., Geng, G., Li, M., Liu, F., Zhang, Y., Zheng, B., Clarke, L., and Zhang, Q.: Dynamic projection of anthropogenic emissions in China: methodology and 2015-2050 emission pathways under a range of socio-economic, climate policy, and pollution control scenarios, *Atmos. Chem. Phys.*, 20, 5729-5757, 10.5194/acp-20-5729-2020, 2020.
- Wang, J., Ge, X., Chen, Y., Shen, Y., Zhang, Q., Sun, Y., Xu, J., Ge, S., Yu, H., and Chen, M.: Highly time-resolved urban aerosol characteristics during springtime in Yangtze River Delta, China: insights from soot particle aerosol mass spectrometry, *Atmos. Chem. Phys.*, 16, 9109-9127, 10.5194/acp-16-9109-2016, 2016.
- Wei, C., Wang, M. H., Fu, Q. Y., Dai, C., Huang, R., and Bao, Q.: Temporal Characteristics and Potential Sources of Black Carbon in Megacity Shanghai, China, *J. Geophys. Res.-Atmos.*, 125, 10.1029/2019jd031827, 2020.
- Xia, Y., Wu, Y., Huang, R.-J., Xia, X., Tang, J., Wang, M., Li, J., Wang, C., Zhou, C., and Zhang, R.: Variation in black carbon concentration and aerosol optical properties in Beijing: Role of emission control and meteorological transport variability, *Chemosphere*, 254, 10.1016/j.chemosphere.2020.126849, 2020.

- Zhang, Y., Sun, J., Zhang, X., Shen, X., Wang, T., and Qin, M.: Seasonal characterization of components and size distributions for submicron aerosols in Beijing, *Science China Earth Sciences*, 56, 890-900, 10.1007/s11430-012-4515-z, 2013.
- Zhang, Y., Tang, L., Croteau, P. L., Favez, O., Sun, Y., Canagaratna, M. R., Wang, Z., Couvidat, F., Albinet, A., Zhang, H., Sciare, J., Prévôt, A. S. H., Jayne, J. T., and Worsnop, D. R.: Field characterization of the PM<sub>2.5</sub> Aerosol Chemical Speciation Monitor: insights into the composition, sources, and processes of fine particles in eastern China, *Atmos. Chem. Phys.*, 17, 14501-14517, 10.5194/acp-17-14501-2017, 2017.