



*Supplement of*

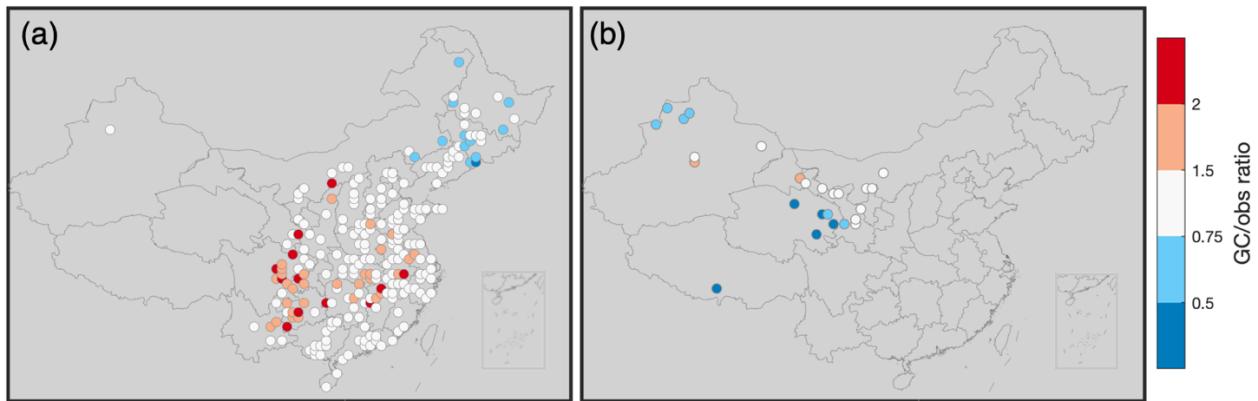
## **Foreign emissions exacerbate PM<sub>2.5</sub> pollution in China through nitrate chemistry**

**Jun-Wei Xu et al.**

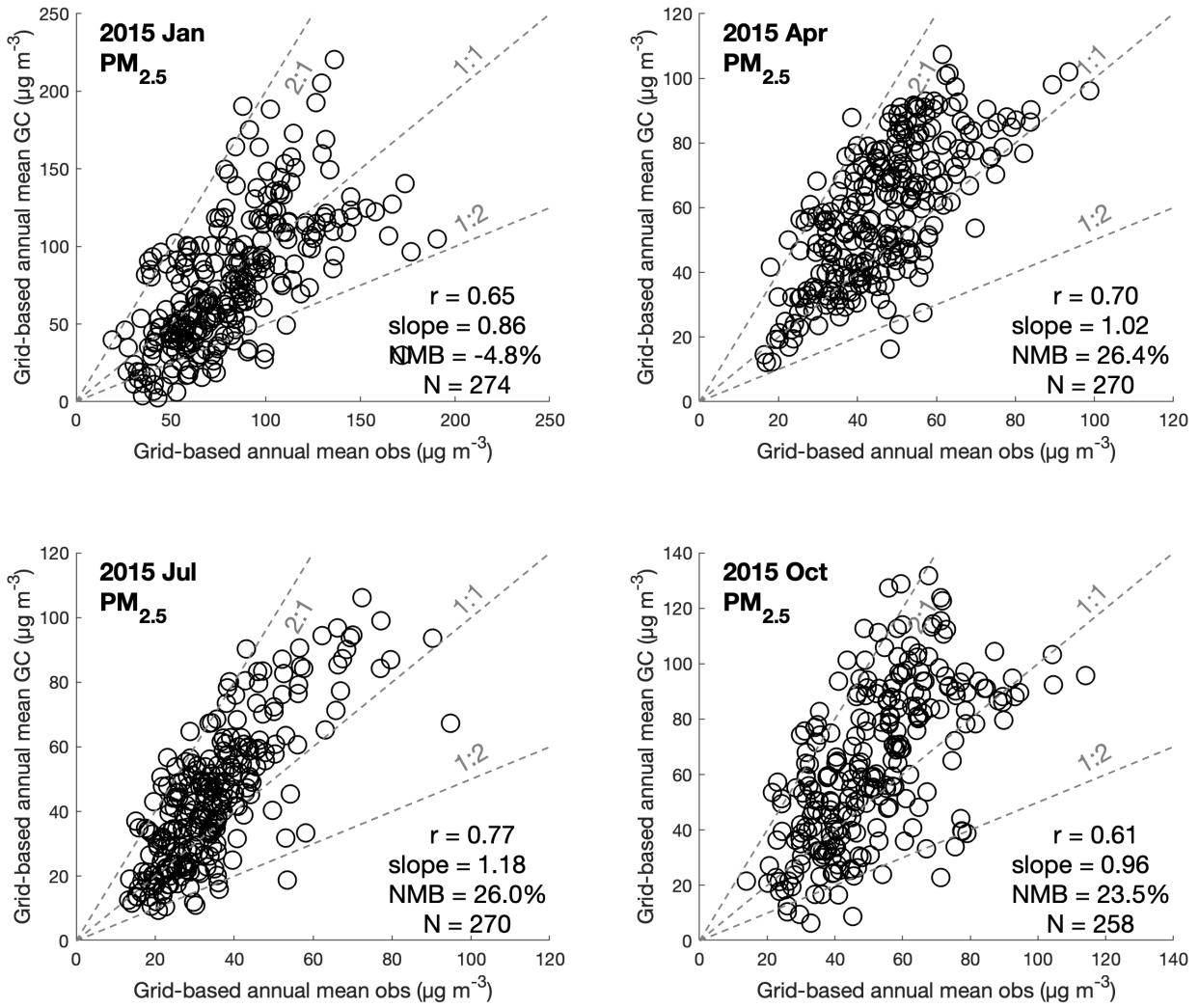
*Correspondence to:* Jintai Lin (linjt@pku.edu.cn)

The copyright of individual parts of the supplement might differ from the article licence.

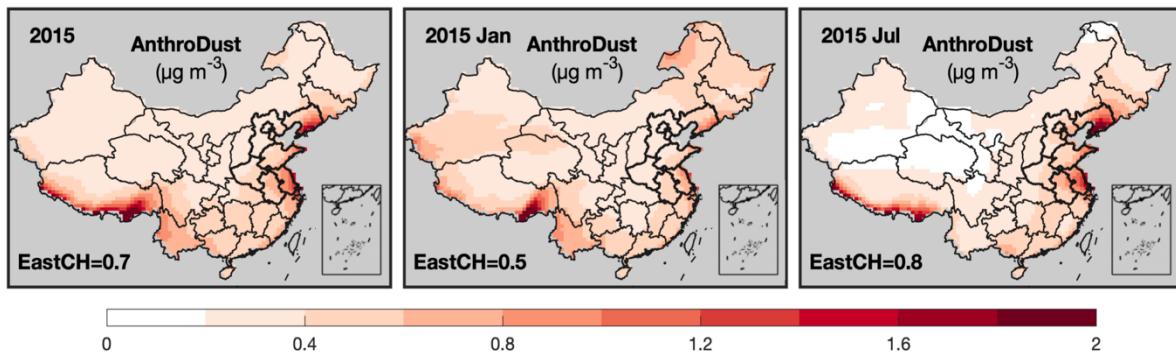
Supplementary information



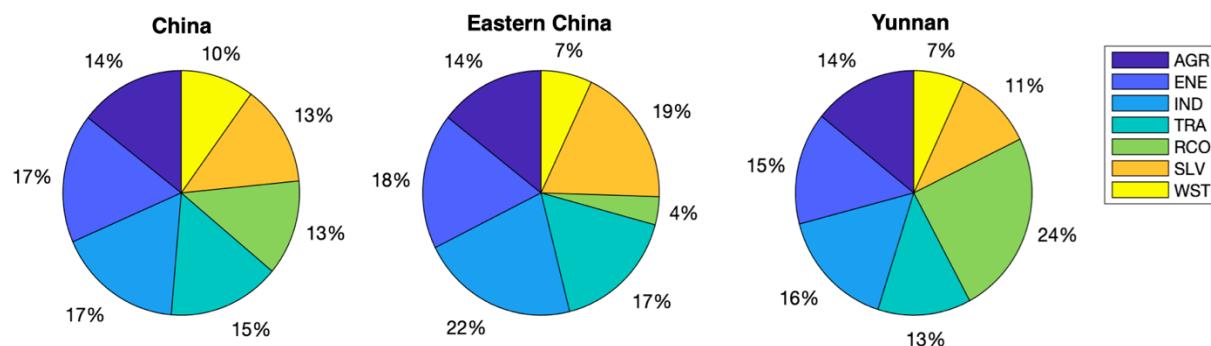
**Figure S1.** The spatial distribution of simulated and observed 2015 mean  $\text{PM}_{2.5}$  concentration ratios at (a) anthropogenic pollutant dominated sites and (b) natural pollutant dominated sites. The simulated concentration at each measurement site represents the  $0.5^\circ \times 0.625^\circ$  grid cell covering that site.



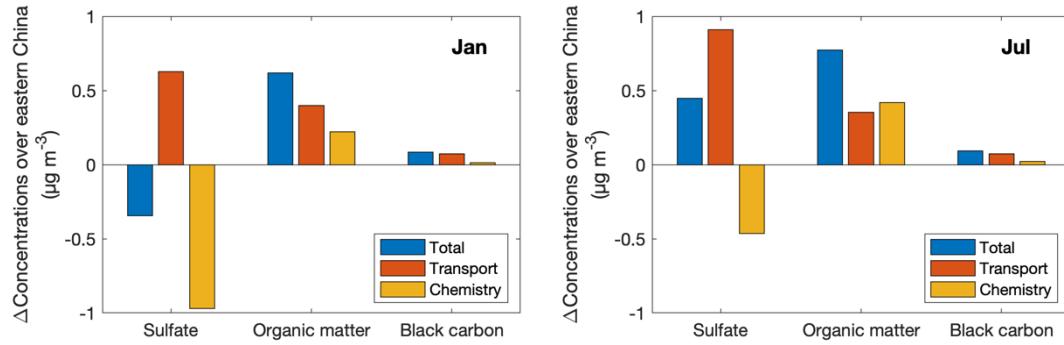
**Figure S2.** Comparison of seasonal PM<sub>2.5</sub> concentrations from simulations and observations for 2015. The simulated concentrations are collocated and coincident with observations from the CNEMC network. Four months (Jan, Apr, Jul and Oct) are selected as the representative of the seasons. Statistics are correlation coefficient (r), slope of the fitting line, normalized mean bias (NMB) and the number of observations (N).



**Figure S3.** Annual mean contributions of foreign anthropogenic dust emissions to China in 2015 as simulated by the GEOS-Chem model. Thick black lines outline the eastern China discussed in this work. Text in the bottom left corner of each panel refers to mean concentrations ( $\mu\text{g m}^{-3}$ ) over the eastern China contributed by foreign anthropogenic emissions.



**Figure S4.** Simulated sectoral contributions of foreign anthropogenic emissions to  $\text{PM}_{2.5}$  concentrations over the entire China, eastern China and Yunnan province in January 2015. Sectors include agriculture (AGR), energy (ENE), industry (IND), transportation (TRA), residential combustion (RCO), solvent use (SLV), and waste burning (WST).



**Figure S5.** Contributions of foreign anthropogenic emissions to sulfate, organic matter and black carbon concentrations in January and July over the eastern China. Total concentrations contributions by foreign anthropogenic emissions are split into contributions from direct transport and chemical interactions according to the legend.

**Table S1.** Observations of PM<sub>2.5</sub> composition concentrations from the literature. Concentrations are all in the unit of  $\mu\text{g m}^{-3}$ . NaN indicates a lack of record from the literature.

Province or municipality	City	Latitude (°)	Longitude (°)	Start Time	Duration (months)	Sulfate	Nitrate	Ammonium	Organic aerosol	Black carbon	References
Beijing	Beijing	39.59	116.21	2014.10	1	11.3	28.56	10.76	13.92	1.91	Xu (2019)
Beijing	Beijing	39.59	116.21	2014.1	1	9.96	11.43	5.13	22.04	6.27	Li (2019)
Beijing	Beijing	40.02	116.28	2014.1	1	22.61	24.41	4.5	44.84	16.03	Li (2019)
Beijing	Beijing	39.90	116.40	2014.1	2	54.21	65.66	10.82	31.04	7.86	Li (2019)
Beijing	Beijing	39.90	116.40	2014.1	1	9.6	12.1	6.7	47.585	9.475	Gao (2018)
Beijing	Beijing	39.90	116.40	2014.4	1	10.7	10.7	11.4	22.105	6.217	Gao (2018)
Beijing	Beijing	39.90	116.40	2014.7	1	25.6	25.6	14.1	14.872	5.329	Gao (2018)
Beijing	Beijing	39.90	116.40	2014.10	1	21.1	45.5	13.9	32.245	6.032	Gao (2018)
Chongqing	Chongqing	30.39	108.37	2014.1	1	NaN	NaN	NaN	13.263	2.41	Huang (2018)
Chongqing	Chongqing	30.39	108.37	2014.4	1	NaN	NaN	NaN	10.934	2.73	Huang (2018)
Gansu	Xigu	36.1	103.62	2014.4	1	5.2	2.2	1.4	11.8	4.7	Wang (2016)
Gansu	Xigu	36.1	103.62	2014.10	1	9	11.3	6.5	17.2	8.5	Wang (2016)
Guangdong	Guangzhou	23.07	113.15	2014.1	1	9.8	5.5	4.8	11.6	5	Tao (2017)
Guangdong	Guangzhou	23.07	113.15	2014.4	1	7	2.1	3.2	7.4	2.2	Tao (2017)
Guangdong	Guangzhou	23.07	113.21	2014.9	1	11.4	1	4.4	7.9	3.6	Tao (2017)
Guangdong	Guangzhou	23.07	113.15	2014.10	1	12.6	2.4	5.1	10.3	3.1	Tao (2017)
Hainan	Haikou	19.32	110.10	2015.1	9	NaN	NaN	NaN	5.6	2.5	Tao (2017)
Hebei	Baoding	38.87	115.47	2014.1	1	25.3	25.3	17.1	124.924	16.973	Gao (2018)
Hebei	Baoding	38.87	115.47	2014.1	1	10.6	10.6	9.3	17.691	3.394	Gao (2018)
Hebei	Baoding	38.87	115.47	2014.7	1	13.1	13.1	8.3	23.156	0.915	Gao (2018)
Hebei	Baoding	38.87	115.47	2014.10	1	32.4	32.4	16.5	38.942	8.772	Gao (2018)
Henan	Zhengzhou	34.8	113.5167	2014.12	2	NaN	NaN	NaN	25	5	Wang (2017)

### Reference:

Cao, J.-J., Shen, Z.-X., Chow, J. C., Watson, J. G., Lee, S.-C., Tie, X.-X., Ho, K.-F., Wang, G.-H. and Han, Y.-M.: Winter and Summer PM 2.5 Chemical Compositions in Fourteen Chinese

Cities, J. Air Waste Manage. Assoc., 62(10), 1214–1226, doi:10.1080/10962247.2012.701193, 2012.

Dai, Q., Bi, X., Liu, B., Li, L., Ding, J., Song, W., Bi, S., Schulze, B. C., Song, C., Wu, J., Zhang, Y., Feng, Y. and Hopke, P. K.: Chemical nature of PM<sub>2.5</sub> and PM<sub>10</sub> in Xi'an, China: Insights into primary emissions and secondary particle formation, Environ. Pollut., 240, 155–166, doi:<https://doi.org/10.1016/j.envpol.2018.04.111>, 2018.

Gao, J., Wang, K., Wang, Y., Liu, S., Zhu, C., Hao, J., Liu, H., Hua, S. and Tian, H.: Temporal-spatial characteristics and source apportionment of PM<sub>2.5</sub> as well as its associated chemical species in the Beijing-Tianjin-Hebei region of China, Environ. Pollut., 233, 714–724, doi:<https://doi.org/10.1016/j.envpol.2017.10.123>, 2018.

Huang, T., Chen, J., Zhao, W., Cheng, J. and Cheng, S.: Seasonal Variations and Correlation Analysis of Water-Soluble Inorganic Ions in PM<sub>2.5</sub> in Wuhan, 2013, Atmos. , 7(4), doi:10.3390/atmos7040049, 2016.

Huang, Y., Liu, Y., Zhang, L., Peng, C. and Yang, F.: Characteristics of Carbonaceous Aerosol in PM<sub>2.5</sub> at Wanzhou in the Southwest of China, Atmos. , 9(2), doi:10.3390/atmos9020037, 2018.

Jiang, N., Duan, S., Yu, X., Zhang, R. and Wang, K.: Comparative major components and health risks of toxic elements and polycyclic aromatic hydrocarbons of PM<sub>2.5</sub> in winter and summer in Zhengzhou: Based on three-year data, Atmos. Res., 213, 173–184, doi:<https://doi.org/10.1016/j.atmosres.2018.06.008>, 2018.

Kong, S. F., Li, L., Li, X. X., Yin, Y., Chen, K., Liu, D. T., Yuan, L., Zhang, Y. J., Shan, Y. P. and Ji, Y. Q.: The impacts of firework burning at the Chinese Spring Festival on air quality: insights of tracers, source evolution and aging processes, Atmos. Chem. Phys., 15(4), 2167–2184, doi:10.5194/acp-15-2167-2015, 2015.

Li, X., Jiang, L., Bai, Y., Yang, Y., Liu, S., Chen, X., Xu, J., Liu, Y., Wang, Y., Guo, X., Wang, Y. and Wang, G.: Wintertime aerosol chemistry in Beijing during haze period: Significant contribution from secondary formation and biomass burning emission, Atmos. Res., 218, 25–33, doi:<https://doi.org/10.1016/j.atmosres.2018.10.010>, 2019.

Ma, X., Xiao, Z., He, L., Shi, Z., Cao, Y., Tian, Z., Vu, T. and Liu, J.: Chemical Composition and Source Apportionment of PM<sub>2.5</sub> in Urban Areas of Xiangtan, Central South China, Int. J. Environ. Res. Public Heal. , 16(4), doi:10.3390/ijerph16040539, 2019.

Tao, J., Zhang, L., Cao, J., Zhong, L., Chen, D., Yang, Y., Chen, D., Chen, L., Zhang, Z., Wu, Y., Xia, Y., Ye, S. and Zhang, R.: Source apportionment of PM<sub>2.5</sub> at urban and suburban areas of the Pearl River Delta region, south China - With emphasis on ship emissions, Sci. Total Environ., 574, 1559–1570, doi:<https://doi.org/10.1016/j.scitotenv.2016.08.175>, 2017.

Tao, Y., Ye, X., Ma, Z., Xie, Y., Wang, R., Chen, J., Yang, X. and Jiang, S.: Insights into different nitrate formation mechanisms from seasonal variations of secondary inorganic aerosols in Shanghai, Atmos. Environ., 145, 1–9, doi:<https://doi.org/10.1016/j.atmosenv.2016.09.012>, 2016.

Wang, H., Tian, M., Chen, Y., Shi, G., Liu, Y., Yang, F., Zhang, L., Deng, L., Yu, J., Peng, C. and Cao, X.: Seasonal characteristics, formation mechanisms and source origins of PM<sub>2.5</sub> in two megacities in Sichuan Basin, China, Atmos. Chem. Phys., 18(2), 865–881, doi:10.5194/acp-18-865-2018, 2018.

Wang, Q., Jiang, N., Yin, S., Li, X., Yu, F., Guo, Y. and Zhang, R.: Carbonaceous species in PM<sub>2.5</sub> and PM<sub>10</sub> in urban area of Zhengzhou in China: Seasonal variations and source apportionment, Atmos. Res., 191, 1–11, doi:<https://doi.org/10.1016/j.atmosres.2017.02.003>,

2017.

- Wang, Y., Jia, C., Tao, J., Zhang, L., Liang, X., Ma, J., Gao, H., Huang, T. and Zhang, K.: Chemical characterization and source apportionment of PM<sub>2.5</sub> in a semi-arid and petrochemical-industrialized city, Northwest China, *Sci. Total Environ.*, 573, 1031–1040, doi:<https://doi.org/10.1016/j.scitotenv.2016.08.179>, 2016.
- Xu, J.-S., Xu, M.-X., Snape, C., He, J., Behera, S. N., Xu, H.-H., Ji, D.-S., Wang, C.-J., Yu, H., Xiao, H., Jiang, Y.-J., Qi, B. and Du, R.-G.: Temporal and spatial variation in major ion chemistry and source identification of secondary inorganic aerosols in Northern Zhejiang Province, China, *Chemosphere*, 179, 316–330, doi:<https://doi.org/10.1016/j.chemosphere.2017.03.119>, 2017.
- Xu, W., Liu, X., Liu, L., Dore, A. J., Tang, A., Lu, L., Wu, Q., Zhang, Y., Hao, T., Pan, Y., Chen, J. and Zhang, F.: Impact of emission controls on air quality in Beijing during APEC 2014: Implications from water-soluble ions and carbonaceous aerosol in PM<sub>2.5</sub> and their precursors, *Atmos. Environ.*, 210, 241–252, doi:<https://doi.org/10.1016/j.atmosenv.2019.04.050>, 2019.
- Zhang, Y., Zhang, H., Deng, J., Du, W., Hong, Y., Xu, L., Qiu, Y., Hong, Z., Wu, X., Ma, Q., Yao, J. and Chen, J.: Source regions and transport pathways of PM<sub>2.5</sub> at a regional background site in East China, *Atmos. Environ.*, 167, 202–211, doi:<https://doi.org/10.1016/j.atmosenv.2017.08.031>, 2017.
- Zong, Z., Wang, X., Tian, C., Chen, Y., Fu, S., Qu, L., Ji, L., Li, J. and Zhang, G.: PMF and PSCF based source apportionment of PM<sub>2.5</sub> at a regional background site in North China, *Atmos. Res.*, 203, 207–215, doi:<https://doi.org/10.1016/j.atmosres.2017.12.013>, 2018.