



Supplement of

Coarse particulate matter air quality in East Asia: implications for fine particulate nitrate

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9 Figure S1. Daily correlations of coarse PM and CO over the North China Plain (NCP) and Seoul Metropolitan Area

10 (SMA). Same as Fig. 3 in the main text but for the years 2016, 2017, 2018, and 2019.



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- the DC-8 aircraft (right). 2*[SO42-] + [NO3-] and [NH4+] are in charge balance, suggesting that AMS measured nitrate is
- mainly inorganic nitrate that is associated with ammonium.

KORUS-AQ median vertical profiles of coarse PM in SMA



- 20 21 Figure S3. Median vertical profiles of coarse PM over SMA during KORUS-AQ. Data filtering criteria is the same as Fig. 4 in the main text. Aircraft observations of coarse PM are derived from the size distribution measurements by insitu Aerosol Particles from the DMT CPSPD Probe.



Diurnal variations of PM1 nitrate and NH3 at surface sites in SMA and diurnal scaling factors of NH3 emission

Figure S4. Effects of anthropogenic coarse PM and diurnal variations of NH₃ emission on PM₁ nitrate and NH₃ over

26 Seoul Metropolitan Area (SMA) during KORUS-AQ. Same as Fig. 4a in the main text but with the diurnal profiles of

27 NH₃ at the Olympic Park site in SMA, diurnal scaling factors of NH₃ emission, and GEOS-Chem model results with

28 default and adjusted NH₃ emissions added. NH₃ was measured by ion chromatography at the Olympic Park site in

29 SMA, ~7 km to the southeast of KIST. The default diurnal scaling factors of NH₃ emission is provided by the Multi-

30 resolution Emission Inventory for China (MEIC) agriculture emission sector.



Observed PM_{2.5} nitrate (median, 25-75th percentiles) GEOS-Chem (without dust uptake of HNO₃ from Zhai et al. (2021)) + dust (natural + anthropogenic) uptake of HNO₃

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33 Figure S5. Effects of dust uptake of HNO₃ on winter and summer mean particulate nitrate over the North China Plain.

34 Observed PM_{2.5} nitrate at sites in NCP and the GEOS-Chem model results without considering dust uptake of HNO₃

are from our previous study (Zhai et al., 2021). GEOS-Chem model results are sampled at NCP observation sites.

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38 39 Table S1 Concentrations of multi-year winter and summer mean^a fine particulate nitrate observed in Beijing and Seoul, Unit: $\mu g m^{-3}$. Data is visualized in Fig. 5 in the main text.

	Winter					Summer				
	IAP ^b	Tsinghua ^c	PKU ^d	NIER ^e	KIST ^f	IAP	Tsinghua	PKU	NIER	KIST
2015	14.6		14.6	9.2		5.0	8.4	12.3	0.4	
2016				7.9	6.6				1.3	1.4
2017		16.5		9.4	8.9		7.7		0.8	
2018	14.7	11.5		6.7	5.2	8.6	9.0		0.7	0.5
2019	11.4	12.8		10.3	10.5	6.1	7.6		0.7	
2020	13.8	15.8				7.2	8.5			0.3
2021		19.2				3.0	6.8			

40 ^aWinter mean are December-January-February averages and summer mean are June-July-August averages unless 41 otherwise noted.

42 ^b Winter mean fine particulate nitrate at the Institute of Atmospheric Physics (IAP) site are January-February-March

43 44 averages (Lei et al., 2021). Winter data for year 2015 and year 2018 are Q-ACSM NR-PM₁. Winter data for year 2019 and 2020 are ToF-ACSM NR-PM2.5. Summer data for year 2015 (1 June - 3 July and 15 August - 31 August) are AMS

45 NR-PM₁. Summer data for years 2018, 2019, 2020, and 2021 are ACSM NR-PM_{2.5}.

46 ° PM_{2.5} nitrate at Tsinghua campus from 2017 to 2021 are measured by Monitor for AeRosols and Gases (MARGA,

47 Metrohm Ltd., Switzerland) (Xu et al., 2019). Winter mean are January-February-March averages. PM1 nitrate in 48

summer 2015 is from Li et al. (2018). ^d Data is from ref Liu et al. (2019).

^e Data is measured by an ambient ion monitor (AIM, URG-9000D/USA)(Jo et al., 2020; Jeong et al., 2022).

^f PM₁ nitrate at the Korea Institute of Science and Technology (KIST) site is HR-ToF-AMS NR-PM₁ (Kim et al.,

2017). Winter mean concentration are averaged from 5 December 2015 to 21 January 2016 for year 2016 (Kim et al.,

2017), from 1 January 2017 to 10 February 2017 for year 2017, from 17 January 2018 to 22 February 2018 for year

2018, and from 22 February 2019 to 2 April 2019 for year 2019 (Kim et al., 2020). Summer mean concentration are

49 50 51 52 53 54 55 56 averaged from 27 July to 31 August for year 2016, from 6 August to 31 August for year 2018, from 13 August to 31 August for year 2020.

58 References

- Jeong, J. I., Seo, J., and Park, R. J.: Compromised Improvement of Poor Visibility Due to PM Chemical
 Composition Changes in South Korea, Remote Sens., 14, 5310, <u>https://doi.org/10.3390/rs14215310</u>,
 2022.
- Jo, Y.-J., Lee, H.-J., Jo, H.-Y., Woo, J.-H., Kim, Y., Lee, T., Heo, G., Park, S.-M., Jung, D., Park, J., and
 Kim, C.-H.: Changes in inorganic aerosol compositions over the Yellow Sea area from impact of
 Chinese emissions mitigation, Atmos. Res., 240, 104948,
 https://doi.org/10.1016/j.atmosres.2020.104948, 2020.
- Kim, H., Zhang, Q., and Sun, Y.: Measurement report: Characterization of severe spring haze episodes and
 influences of long-range transport in the Seoul metropolitan area in March 2019, Atmos. Chem. Phys.,
 20, 11527-11550, <u>https://doi.org/10.5194/acp-20-11527-2020</u>, 2020.
- Kim, H., Zhang, Q., Bae, G. N., Kim, J. Y., and Lee, S. B.: Sources and atmospheric processing of winter aerosols in Seoul, Korea: insights from real-time measurements using a high-resolution aerosol mass spectrometer, Atmos. Chem. Phys., 17, 2009-2033, https://doi.org/10.5194/acp-17-2009-2017, 2017.
- Lei, L., Zhou, W., Chen, C., He, Y., Li, Z., Sun, J., Tang, X., Fu, P., Wang, Z., and Sun, Y.: Long-term characterization of aerosol chemistry in cold season from 2013 to 2020 in Beijing, China, Environ. Pollut., 268, 115952, https://doi.org/10.1016/j.envpol.2020.115952, 2021.
- Li, H., Zhang, Q., Zheng, B., Chen, C., Wu, N., Guo, H., Zhang, Y., Zheng, Y., Li, X., and He, K.: Nitrate-driven urban haze pollution during summertime over the North China Plain, Atmos. Chem. Phys., 18, 5293-5306, https://doi.org/10.5194/acp-18-5293-2018, 2018.
- Liu, M., Huang, X., Song, Y., Tang, J., Cao, J., Zhang, X., Zhang, Q., Wang, S., Xu, T., Kang, L., Cai, X.,
 Zhang, H., Yang, F., Wang, H., Yu, J. Z., Lau, A. K. H., He, L., Huang, X., Duan, L., Ding, A., Xue,
 L., Gao, J., Liu, B., and Zhu, T.: Ammonia emission control in China would mitigate haze pollution
 and nitrogen deposition, but worsen acid rain, Proc. Natl. Acad. Sci. USA, 116, 7760-7765,
 https://doi.org/10.1073/pnas.1814880116, 2019.
- Xu, Q., Wang, S., Jiang, J., Bhattarai, N., Li, X., Chang, X., Qiu, X., Zheng, M., Hua, Y., and Hao, J.:
 Nitrate dominates the chemical composition of PM_{2.5} during haze event in Beijing, China, Sci. Total
 Environ., 689, 1293-1303, https://doi.org/10.1016/j.scitotenv.2019.06.294, 2019.
- Zhai, S., Jacob, D. J., Wang, X., Liu, Z., Wen, T., Shah, V., Li, K., Moch, J. M., Bates, K. H., Song, S.,
 Shen, L., Zhang, Y., Luo, G., Yu, F., Sun, Y., Wang, L., Qi, M., Tao, J., Gui, K., Xu, H., Zhang, Q.,
 Zhao, T., Wang, Y., Lee, H. C., Choi, H., and Liao, H.: Control of particulate nitrate air pollution in
 China, Nat. Geosci., 14, 389-395, https://doi.org/10.1038/s41561-021-00726-z, 2021.
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