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

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Abstract. Air quality network data in China and South Korea show very high year-round mass concentrations of coarse particulate matter (PM), as inferred by difference between PM_{10} and $PM_{2.5}$. Coarse PM concentrations in 2015 averaged 52 μg m^{-3} in the North China Plain (NCP) and 23 μg m^{-3} in the Seoul Metropolitan Area (SMA), contributing nearly half of PM_{10}. Strong daily correlations between coarse PM and carbon monoxide imply a dominant source from anthropogenic fugitive dust. Coarse PM concentrations in the NCP and the SMA decreased by 21% from 2015 to 2019 and further dropped abruptly in 2020 due to COVID-19 reductions in construction and vehicle traffic. Anthropogenic coarse PM is generally not included in air quality models but scavenges nitric acid to suppress the formation of fine particulate nitrate, a major contributor to $PM_{2.5}$ pollution. GEOS-Chem model simulation of surface and aircraft observations from the KORUS-AQ campaign over the SMA in May-June 2016 shows that consideration of anthropogenic coarse PM largely resolves the previous model overestimate of fine particulate nitrate. The effect is smaller in the NCP which has a larger excess of ammonia. Model sensitivity simulations for 2015-2019 show that decreasing anthropogenic coarse PM directly increases $PM_{2.5}$ nitrate in summer, offsetting 80% the effect of nitrogen oxide and ammonia emission controls, while in winter the presence of coarse PM increases the sensitivity of $PM_{2.5}$ nitrate to ammonia and sulfur dioxide emissions. Decreasing coarse PM helps to explain the lack of decrease in wintertime $PM_{2.5}$ nitrate observed in the NCP and the SMA over the 2015-2021 period despite decreases in nitrogen oxide and ammonia emissions. Continuing decrease of fugitive dust pollution means that more stringent nitrogen oxide and ammonia emission controls will be required to successfully decrease $PM_{2.5}$ nitrate.

1. Introduction

Coarse particulate matter (coarse PM; particulate matter between 2.5 μm and 10 μm aerodynamic diameter) is a severe air pollution problem in East Asia, contributing a particle mass comparable to fine particulate matter ($PM_{2.5}$) and thus about half of PM_{10} (Chen et al., 2019; Lee et al., 2015; Qiu et al., 2014; Wang et al., 2018a). It is mainly fugitive mineral dust, with contributions from both natural desert dust and anthropogenic sources including on-road traffic, construction, and agriculture (Wu et al., 2016; Zhao et al., 2017; Liu et al., 2021; Katra, 2020). Atmospheric chemistry models used in air quality applications generally do not include anthropogenic fugitive dust, due to the lack of available emission inventories except for a few urban areas (Li et al., 2021a; Li et al., 2021b; Li et al., 2021c). Aside from its direct interest as an air pollutant, coarse PM can suppress $PM_{2.5}$ by heterogeneously taking up acids (HNO_3, SO_2, and H_2SO_4) that would otherwise lead to $PM_{2.5}$ formation. This uptake has been observed for natural dust events (Wang et al., 2017; Heim et al., 2020; Wang et al., 2018b; Park et al., 2004; Stone et al., 2011), but the more ubiquitous effect from anthropogenic dust has received little study (Kakavas and Pandis, 2021; Hodzic et al., 2006). With increasingly stringent control measures to decrease fugitive dust air pollution in East Asia (Chinese State Council, 2019; Noh et al., 2018; Wu et al., 2016; Xing et al., 2018), it is important to better understand the impact on $PM_{2.5}$ air quality.

A specific issue is the effect of anthropogenic dust on $PM_{2.5}$ nitrate. Nitrate is a major component of $PM_{2.5}$ in urban regions of East Asia including the North China Plain (NCP) (Li et al., 2019; Zhai et al., 2021a) and the Seoul Metropolitan Area (SMA) (Jeong et al., 2022; Kim et al., 2020), and it can dominate haze pollution events in both regions (Fu et al., 2020; Li et al., 2018; Xu et al., 2019; Kim et al., 2017; Kim et al., 2020). $PM_{2.5}$ nitrate over North China in winter has not decreased in recent years despite reductions in emissions of the precursor nitrogen oxides ($NO_x \equiv NO + NO_2$) (Zhai et al., 2021a; Fu et al., 2020) from fossil fuel combustion. This has been attributed to limitation by ammonia (NH_3) emissions, since $PM_{2.5}$ nitrate is mainly present as ammonium nitrate (Zhai et al., 2021a). Decreasing coarse PM emissions is another possible explanation as it would allow more HNO_3 to be available for $PM_{2.5}$ nitrate formation, and it could also shift $PM_{2.5}$ nitrate formation to be more NH_3-limited. Better understanding this sensitivity of $PM_{2.5}$ nitrate to coarse PM is of crucial importance because of recent efforts by the Chinese government to decrease NH_3 emissions (Liao et al., 2022), which are mainly from agriculture with additional urban contributions from vehicle, industrial, and waste disposal sources (Mgelwa et al., 2022).

In this work, we show that coarse PM over the NCP and the SMA is mainly anthropogenic and decreased by 21% during the 2015-2019 period. We find that accounting for this anthropogenic coarse PM in the GEOS-Chem atmospheric chemistry model greatly improves the ability of the model to simulate $PM_{2.5}$ nitrate during the KORUS-AQ aircraft campaign over Korea where previous GEOS-Chem simulations found a large overestimate (Travis et al., 2022; Zhai et al., 2021b). From there we examine the implications for the effects of emission controls on long-term trends of $PM_{2.5}$ nitrate in China and South Korea.

2. Coarse PM in China and South Korea

Figure 1 shows the annual mean concentrations of coarse PM in 2015, 2019, and 2020 measured at air quality networks in China and South Korea as the $PM_{10} - PM_{2.5}$ difference. Data for China are from the Ministry of Ecology and Environment (MEE) network (http://www.quotsoft.net/air/) and data for South Korea are from the AirKorea network (https://www.airkorea.or.kr). We remove spurious data when $PM_{2.5}$ is higher than PM_{10}, which account for 1.7% and 0.2% of the dataset respectively in China and South Korea.

We see from Fig. 1 that coarse PM concentrations in China and South Korea are highest in the NCP and the SMA, respectively, indicating a dominant urban anthropogenic origin. Coarse PM in year 2015 averaged 52 $\mu g\ m^{-3}$ in the NCP and 23 $\mu g\ m^{-3}$ in the SMA, contributing nearly half of total PM_{10} (120 $\mu g\ m^{-3}$ in the NCP and 50 $\mu g\ m^{-3}$ in the SMA). National air quality standards for annual mean PM_{10} are 70 $\mu g\ m^{-3}$ in China (urban) and 50 $\mu g\ m^{-3}$ in South Korea, well above the World Health Organization (WHO) recommended annual standard of 15 $\mu g\ m^{-3}$. Coarse PM decreased by 21% in both the NCP and the SMA from 2015 to 2019, reflecting emission controls on fugitive dust (Chinese State Council, 2013, 2018; Noh et al., 2018; Wu et al., 2016), and further decreased strongly in 2020 because of COVID-19 restrictions on traffic and construction. The COVID-19 impact is evident in China by comparing concentrations before and after the sharp January 24, 2020 lockdown (Fig. 2).

Figure 3 shows further evidence of the dominant anthropogenic contribution to coarse PM as the daily correlation with carbon monoxide (CO) in 2015. CO is emitted by incomplete combustion and is a tracer of urban influence. We find strong correlations between coarse PM and CO with consistent slopes except in spring, which features high coarse PM outliers attributable to desert dust events (Heim et al., 2020; Shao and Dong, 2006). Similar correlations to 2015 are found in other years (Fig. S1). The desert dust events drive the seasonal maximum of coarse PM in Fig. 1h.

3. Effect of anthropogenic coarse PM on fine particulate nitrate during KORUS-AQ

We simulated the effect of anthropogenic coarse PM on $PM_{2.5}$ nitrate using the GEOS-Chem model and evaluated the model with observations from the KORUS-AQ aircraft campaign over South Korea in May-June 2016 (Crawford et al., 2021). KORUS-AQ offers a unique data set of detailed aerosol and gas-phase composition over East Asia. Previous GEOS-Chem simulations showed a large overestimate of fine particulate nitrate and a large underestimate of coarse PM (Travis et al., 2022; Zhai et al., 2021b). Particulate nitrate concentrations were measured during KORUS-AQ at the Korea Institute of Science and Technology (KIST) surface site and on the aircraft by Aerosol Mass Spectrometers (AMS) with size cut of 1 μm diameter (PM_1 nitrate) (Kim et al., 2017; Kim et al., 2018). The AMS only detects non-refractory nitrate, taken here to be ammonium nitrate (Fig. S2). Total particulate nitrate with size cut of 4 μm diameter (PM_4 nitrate) was also sampled on the aircraft by the Soluble Acidic Gases and Aerosol (SAGA) instrument (Dibb et al., 2003; McNaughton et al., 2007). Additional measurements on the aircraft included HNO_3 concentrations with a Chemical Ionization Time of Flight Mass Spectrometer (CIT-ToF-CIMS), and aerosol size distributions including coarse PM with a DMT CPSPD Probe. We focus on the observations over the SMA and exclude observations from two process-directed flights (RF7 and RF8) and the Daesan power plant plume following Park et al. (2021).

We use GEOS-Chem version 13.0.2 (https://zenodo.org/record/4681204) in a nested-grid simulation over East Asia (100 - 150° E, 20 - 50° N) with a horizontal resolution of 0.5°× 0.625°. The model simulates detailed oxidant-aerosol chemistry relevant to $PM_{2.5}$ nitrate formation (Zhai et al., 2021a) and is driven by meteorological data from the NASA Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2). Formation of semi-volatile ammonium nitrate aerosol is governed by ISORROPIA version 2.2 thermodynamics (Fountoukis and Nenes, 2007). Dry deposition of gases and particles follows a standard resistance-in-series scheme (Wesely, 1989). Wet deposition of gases and particles includes contributions from rainout, washout, and scavenging in convective updrafts (Liu et al., 2001; Luo et al., 2019). The model includes reactive uptake of HNO_3 on dust limited by dust alkalinity and mass transfer (Fairlie et al., 2010), assuming 7.1 % Ca^{2+} and 1.1% Mg^{2+} as carbonates per mass in emitted dust (Shah et al., 2020a; Tang and Han, 2017; Zhang et al., 2014). The relative humidity (RH)-dependent reactive uptake coefficient (γ) of HNO_3 is based on laboratory studies (Liu et al., 2008; Huynh and McNeill, 2020) and observations during natural dust events in Beijing (Tian et al., 2021; Wang et al., 2017), and increases from 0.06 to 0.21 as RH increases from 40% to 80%. Monthly anthropogenic emissions for China are from the Multi-resolution Emission Inventory for China (MEIC) (Zheng et al., 2018; Zheng et al., 2021a; Zheng et al., 2021b), and emissions for other Asian countries including South Korea are from the KORUSv5 inventory (Woo et al., 2020). Fine anthropogenic mineral dust emissions from combustion and industrial sources (ash) are derived from the MEIC and KORUSv5 inventories as the residual of anthropogenic primary $PM_{2.5}$ emissions after excluding primary organic aerosol, black carbon, and primary sulfate (Philip et al., 2017).

We compare the results from the standard model as described above to a simulation where we add anthropogenic coarse PM by using 24-hour average observed coarse PM concentrations from the air quality networks (Fig. 1) as boundary conditions at the lowest model level. For this purpose, we linearly interpolate the daily mean coarse PM data from the network to the GEOS-Chem model horizontal grid and apply them to the coarse dust GEOS-Chem model component with an effective diameter of 4.8 μm. This concentration boundary condition in the lowest model level serves as an implicit source and defines the vertical concentration profile. The resulting vertical profiles of coarse PM in GEOS-Chem over South Korea are consistent with KORUS-AQ aircraft observations (Fig. S3). Anthropogenic coarse PM is assumed to be mainly fugitive dust with the same alkalinity properties as natural dust (Zhang et al., 2014; Tang and Han, 2017).

Figure 4 compares GEOS-Chem to the KORUS-AQ observations including median diurnal PM_1 nitrate at the KIST site and median aircraft vertical profiles over the SMA. The model is sampled along the aircraft flight tracks at the times of the observations, all in daytime. PM_{1-4} nitrate is derived as the difference between SAGA PM_4 nitrate and AMS PM_1 nitrate. Here we take ammonium nitrate in the model for comparison to PM_1 observations, and size-resolved dust nitrate for comparison to PM_{1-4} observations. In this way, any dust-associated refractory PM_1 nitrate is included in the PM_{1-4} profiles, for both observations and the GEOS-Chem model. Such classification does not allow for supermicron ammonium nitrate, but KORUS-AQ observations found ammonium nitrate to be mainly submicron (Kim et al., 2018). GEOS-Chem results are shown both for the standard model (not including anthropogenic coarse PM) and with the addition of anthropogenic coarse PM. In both simulations, we adjusted the diurnal variation of NH_3 emission to match the NH_3 observations made at the Olympic Park site, 7 km southeast of KIST (Fig. S4).

The standard GEOS-Chem simulation without anthropogenic fugitive dust overestimates daytime PM_1 nitrate (aircraft and surface) by about a factor of two while underestimating PM_{1-4} nitrate by about a factor of two (Fig. 4a, b, and c). Coarse PM in the standard simulation (from natural dust and sea salt) is near zero, in contrast to observations (Fig. 4d). Adding anthropogenic coarse PM to the model corrects this bias and further corrects the PM_1 and PM_{1-4} nitrate biases by providing an added sink for HNO_3. We find that anthropogenic coarse PM takes up HNO_3 three times faster than dry deposition and that this uptake is limited by mass-transfer rather than alkalinity (only 60-70% of the coarse dust alkalinity in surface air is neutralized on average). The shift from PM_1 to PM_{1-4} nitrate is consistent with the uptake of HNO_3 by coarse PM, with some of this uptake in the model taking place on dust coarser than 4 μm and so not observed by PM_{1-4} nitrate. Half of the model overestimate of HNO_3 is corrected (Fig. 4e), with the remainder possibly due to an underestimate of HNO_3 deposition velocity (Travis et al., 2022). The model overestimates nighttime nitrate in surface air at the KIST site, even with anthropogenic coarse PM. This nighttime nitrate in the model is driven by heterogeneous NO_2 and N_2O_5 chemistry under stratified conditions, which could be subject to large local errors (Travis et al., 2022).

We also examined the effect of anthropogenic coarse PM on $PM_{2.5}$ nitrate concentrations in the NCP. $PM_{2.5}$ nitrate observations in NCP are mostly filter-collected bulk $PM_{2.5}$ nitrate, which could be biased low in summer due to volatilization (Chow et al., 2005). Previous evaluation of GEOS-Chem with 2013 and 2015 $PM_{2.5}$ nitrate observations across China in summer and winter found no significant bias in 2015 or winter 2013 but an overestimate in summer 2013 (Zhai et al., 2021a). That simulation did not include HNO_3 uptake by dust (natural or anthropogenic). We find here that including HNO_3 uptake by fine ($PM_{2.5}$) dust has little effect on total $PM_{2.5}$ nitrate but partitions 10% of ammonium nitrate mass to fine dust nitrate in winter and 23% in summer (Fig. S5). Adding anthropogenic coarse PM in GEOS-Chem decreases modeled ammonium nitrate in the NCP by 10-20% in winter and by 25-30% in summer, a relatively more modest effect than over the SMA because of larger excess of NH_3. The comparison with $PM_{2.5}$ nitrate observations here indicates that fine dust associated nitrate should be considered when comparing modeled particle nitrate to bulk $PM_{2.5}$ nitrate data.

4. Implications for long-term trends of $PM_{2.5}$ nitrate and responses to emission controls

There are to our knowledge no continuous long-term records of $PM_{2.5}$ nitrate concentrations in China or South Korea. Figure 5 shows a multi-year compilation of winter and summer mean PM_1 and $PM_{2.5}$ nitrate observations from individual field campaigns in Beijing and Seoul over 2015-2021 (Table S1). We find no significant trends in winter, consistent with previous studies in the NCP that examined shorter periods (Fu et al., 2020). In summer, observations tend to show a decrease over the period but with large interannual variations driven by meteorology (Li et al., 2018; Zhai et al., 2021a).

Changes in anthropogenic emissions of NO_x, SO_2, NH_3, $PM_{2.5}$, and coarse PM could all affect $PM_{2.5}$ nitrate, and we used GEOS-Chem to investigate these effects for the 2015-2019 period. The Multi-resolution Emission Inventory for China (MEIC) reports that NO_x emissions in the NCP decreased by 11% from 2015 to 2019, SO_2 emissions decreased by 54%, and primary $PM_{2.5}$ from combustion decreased by

35% (Zheng et al., 2021a). This primary $PM_{2.5}$ includes a 40% contribution from mineral ash that we treat as anthropogenic fine dust and decreased by 27% from 2015 to 2019. The MEIC also reports a 15% decrease of NH_3 emissions over China from 2015 to 2019 (19% for the NCP), while the PKU-NH_3 emission inventory reports a 6% decrease over China from 2015 to 2018 (Liao et al., 2022). Observations of surface NO_2 and SO_2 over the SMA imply a 22% decrease of NO_x emissions and a 40% decrease of SO_2 emissions from 2015 to 2019 (Bae et al., 2021; Colombi et al., 2022). Coarse PM decreased by 33% over the NCP and by 31% over SMA during the same period (considering winter and summer data only).

Figure 6 shows the resulting emission-driven changes of $PM_{2.5}$ nitrate over the NCP and SMA from 2015 to 2019 as simulated by GEOS-Chem in sensitivity simulations applying either 2015 or 2019 emissions to the same meteorological year (2019), and with or without anthropogenic coarse PM. The sum of changes driven by individual emission changes amounts to the total emission-driven net change. Sensitivities to NH_3 and primary $PM_{2.5}$ emissions in the SMA are solely driven by emission trends in China since we assume no emission trends for these species in South Korea.

The model reproduces the lack of trend in winter and the decreasing trend in summer seen in the observations for both the NCP and SMA. The lack of trend in winter reflects offsetting influences from decreasing NO_x, NH_3, and primary $PM_{2.5}$ emissions on the one hand, and decreasing SO_2 and coarse PM emissions on the other hand. Decreasing SO_2 increases the availability of NH_3 for nitrate formation (Fu et al., 2020; Zhai et al., 2021a). Decreasing primary $PM_{2.5}$ reduces the aerosol volume available for heterogeneous conversion of NO_x to HNO_3 (Shah et al., 2020b). Decreasing coarse PM has relatively little direct effect on $PM_{2.5}$ nitrate in winter in the NCP because abundant atmospheric NH_3 combined with low temperatures drives HNO_3 near-quantitively to ammonium-nitrate particles, and subsequent mass transfer of HNO_3 from ammonium nitrate to coarse PM is very slow because of the weak HNO_3 partial pressure (Wexler and Seinfeld, 1992). The decrease of coarse PM still quantitatively offsets the benefit from NO_x emission controls, which has been the main vehicle for controlling $PM_{2.5}$ nitrate. Consideration of coarse PM in the model further increases the sensitivity of $PM_{2.5}$ nitrate to NH_3 and SO_2 emissions respectively by 30% and 46%. This is because coarse PM provides an additional sink for the small fraction of HNO_3 that remains in the gas phase, which increases the sensitivity of the atmospheric lifetime of total nitrate (ammonium nitrate + HNO_3) to changes in NH_3 or SO_2 emissions (Zhai et al., 2021a).

In summer, we find that the decrease in coarse PM over the 2015-2019 period directly cancels half of the benefit from decreasing NO_x, SO_2, NH_3, and primary $PM_{2.5}$ emissions in the NCP, with less effect in the SMA. Over the NCP, the decrease of coarse PM offsets 80% of the benefits from NO_x and NH_3 emission controls. Unlike in winter, decreasing SO_2 suppresses nitrate formation by decreasing the aerosol liquid water content (Stelson and Seinfeld, 1982). The effect of decreasing coarse PM emissions in summer is larger than in winter because warmer temperatures allow more HNO_3 to remain in the gas phase under

NH_3-HNO_3-H_2SO_4 thermodynamics and thus be scavenged by coarse PM.

5. Conclusions

Coarse PM (PM_{10} - $PM_{2.5}$) in urban areas of China and South Korea is very high year-round and is mainly of anthropogenic origin as fugitive dust except for natural desert dust events in spring. Annual mean coarse PM concentrations decreased by 21% from 2015 to 2019 in both the North China Plain (NCP) and the Seoul Metropolitan Area (SMA), with steeper decreases in 2020 because of COVID-19 restrictions on traffic and construction. Considering only winter and summer when the influence of natural dust is small, we find that anthropogenic fugitive dust emissions decreased by about 30% from 2015 to 2019 in both the NCP and the SMA.

Anthropogenic coarse PM is of direct air quality concern because it accounts for about half of total PM_{10} in the NCP and the SMA, but it also takes up HNO_3 effectively and can thus suppress formation of fine particulate nitrate which is a major component of $PM_{2.5}$ pollution. Comparison of GEOS-Chem model simulations to surface and aircraft observations from the KORUS-AQ campaign over the SMA in May-June 2016 shows that accounting for anthropogenic coarse PM largely corrects previous model overestimates of fine particulate nitrate.

Decrease in anthropogenic coarse PM emissions to improve PM_{10} air quality could have the unintended consequence of increasing $PM_{2.5}$ nitrate, offsetting the gains from decreases in NO_x and NH_3 emissions. Compilation of 2015-2021 observations of fine particulate nitrate in Beijing and Seoul suggests little trend in winter and a decrease in summer, consistent with GEOS-Chem. Decreasing coarse PM in the model in winter offsets the benefit of decreasing NO_x emissions, and coarse PM further increases the sensitivity of $PM_{2.5}$ nitrate to changes in NH_3 and SO_2 emissions by affecting the lifetime of total inorganic nitrate (ammonium nitrate + HNO_3). In summer, decreasing coarse PM in the NCP offsets 80% of the $PM_{2.5}$ nitrate benefit of decreasing NO_x and NH_3 emissions. As coarse PM continues to decrease in response to fugitive dust pollution control, there is a greater need to reduce NH_3 and NO_x emissions in order to decrease fine particulate nitrate air pollution in East Asia.

Data availability. $PM_{2.5}$, PM_{10}, and CO data over China are from http://www.quotsoft.net/air/, over South Korea are from https://www.airkorea.or.kr/web/last_amb_hour_data?pMENU_NO=123. Surface and aircraft data during KORUS-AQ are from https://doi.org/10.5067/Suborbital/KORUSAQ/DATA01. Multi-year compilation of winter and summer mean PM_1 and $PM_{2.5}$ nitrate are provided in Table S1.

Supplement. The supplement related to this article is uploaded at submission.

Author Contributions. S.Z. and D.J.J. designed the research. S.Z. performed the research. D.C.P., N.K.C., V.S., L.H.Y., and H.L. helped with data analysis and results interpretation. Q.Z. provided the MEIC emission inventory. S.W., H.K., Y.S., J.S.C, J.S.P., J.E.D., T.L., J.S.H, and B.E.A provided observation data. J.H.W. and Y.K. provided the KORUSv5 emission inventory. G.L., F.Y., and K.L. helped with model simulations. S.Z. and D.J.J wrote the paper with input from all other authors.

Competing Interests. The authors declare no competing interests.

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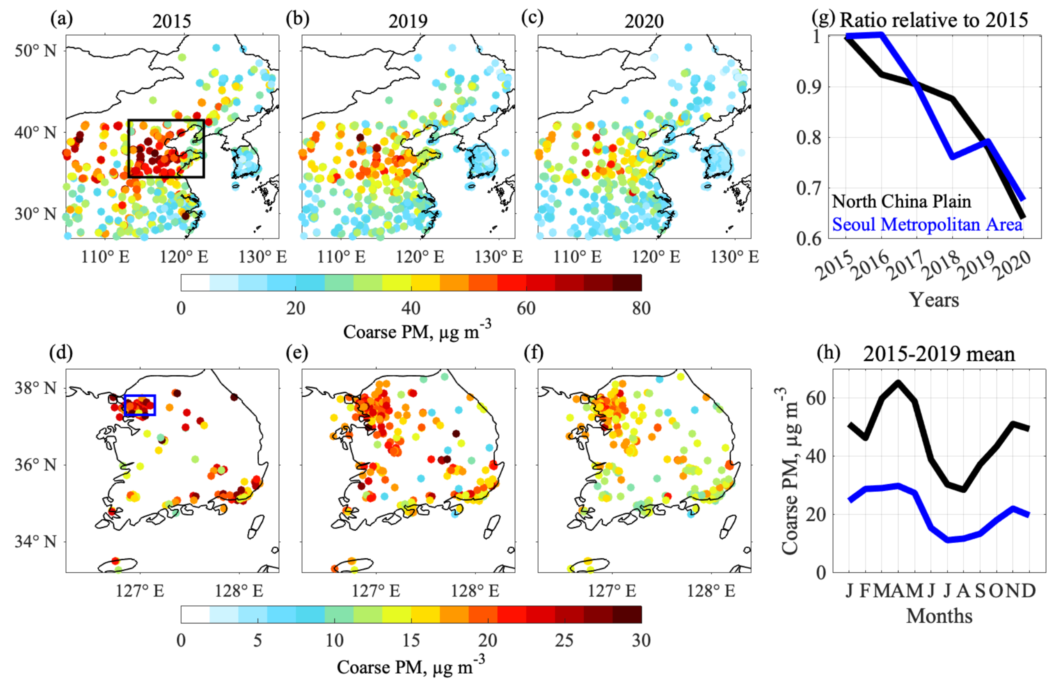


Figure 1. Distributions and trends of coarse PM concentrations over China and South Korea during 2015-2020. Here and elsewhere, coarse particulate matter (PM) is defined as particles between 2.5 and 10 µm aerodynamic diameter and its concentration is determined by subtracting $PM_{2.5}$ from PM_{10} in the air quality network data. Panels (a)-(c) show the annual mean concentrations in 2015, 2019, and 2020 over China and panels (d)-(f) show the same for South Korea. The rectangles in (a) and (d) delineate the North China Plain or NCP (113 - 122.5° E, 34.5 - 41.5° N) and the Seoul Metropolitan area or SMA (126.7 - 127.3° E, 37.3 - 37.8° N). Panel (g) shows annual trends relative to 2015 in the NCP (197 sites) and the SMA (33 sites) averaged over sites with at least 70% data coverage each year from 2015 to 2020. Panel (h) shows the mean 2015-2019 seasonality over the NCP and SMA.

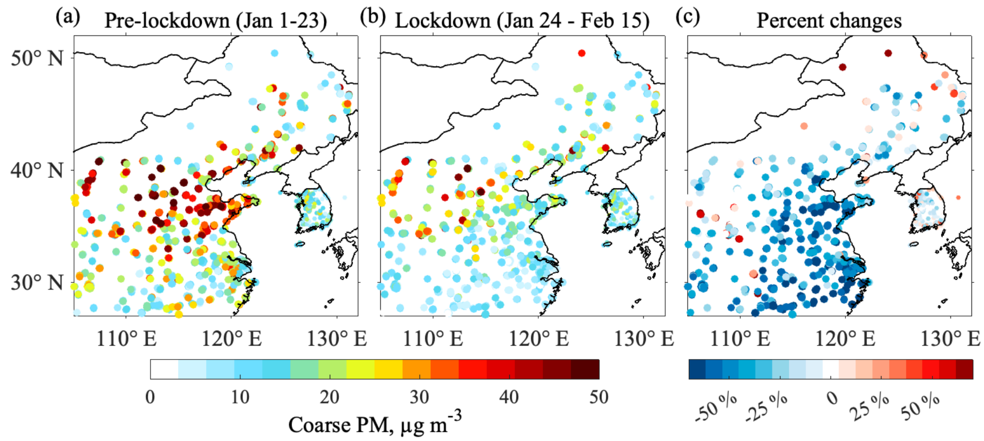


Figure 2. Response of coarse PM to COVID-19 lockdown in China. (a) Coarse PM averaged for the three weeks before the China national lockdown (January 1-23, 2020). (b) Coarse PM averaged during the three-week lockdown (January 24 - February 15, 2020). (c) Percent changes of coarse PM between lockdown and pre-lockdown periods.

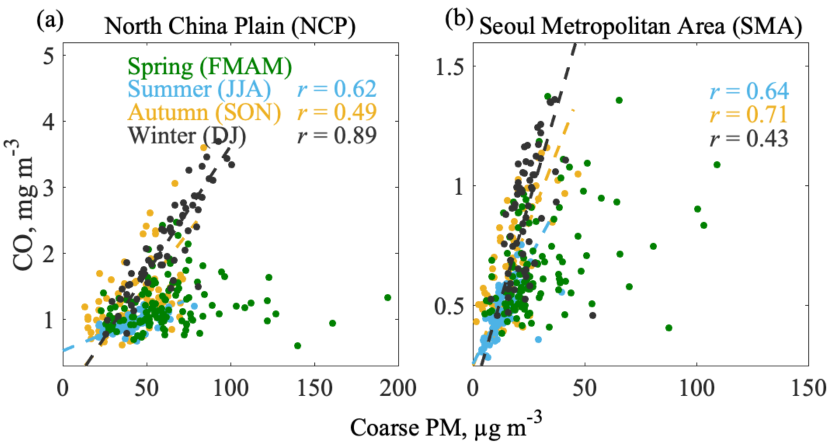


Figure 3. Daily correlations of coarse PM and CO concentrations over the North China Plain (NCP) and Seoul Metropolitan Area (SMA) in 2015. Coarse PM and CO concentrations are 24-h averages of air quality network observations spatially averaged over the two regions. Also shown are the correlation coefficients and reduced-major-axis regression lines except in spring when the correlation is not significant (p-value > 0.05). We include February in spring to cover the season of natural dust events (Tang and Han, 2017).

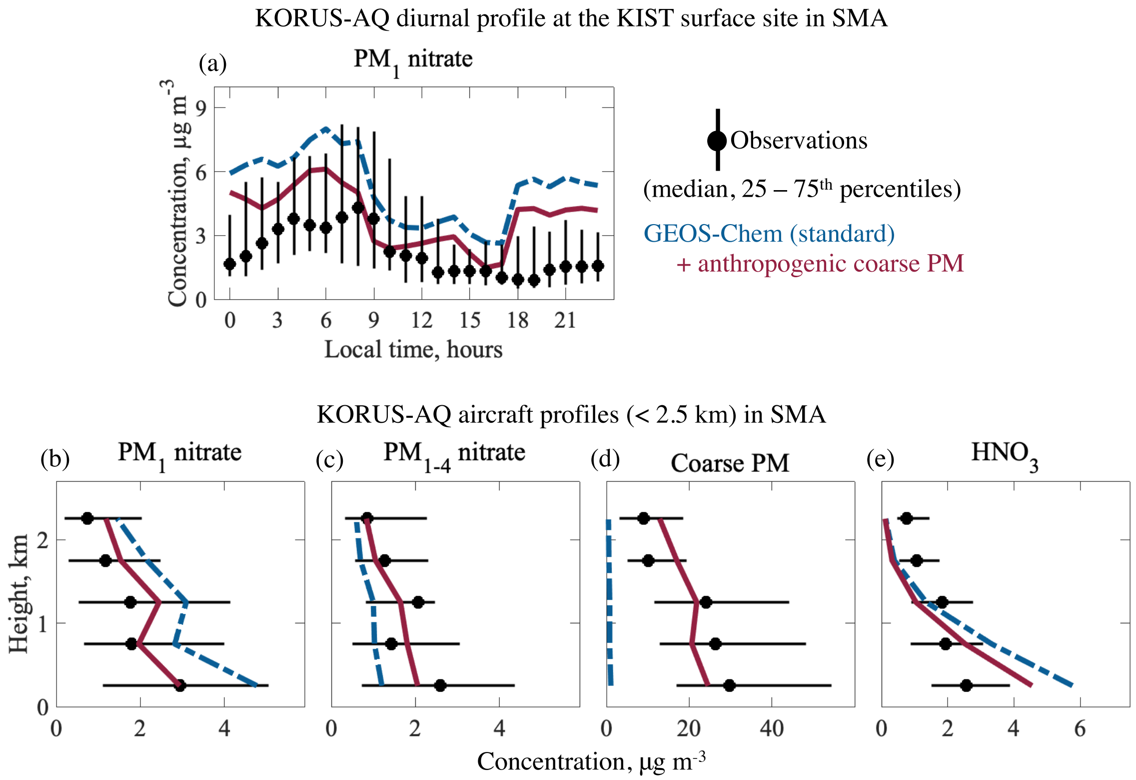


Figure 4. Effect of anthropogenic coarse PM on nitrate concentrations over the Seoul Metropolitan Area (SMA) during the KORUS-AQ campaign (May-June 2016). GEOS-Chem model results without (standard) and with anthropogenic coarse PM are compared to surface and aircraft observations. (a) Median diurnal variation (error bars are 25th and 75th percentiles) of non-refractory PM_1 nitrate (taken to be ammonium nitrate) at the Korea Institute of Science and Technology (KIST) site. (b)-(e) Median vertical profiles of non-refractory PM_1 nitrate, PM_{1-4} nitrate, coarse PM ($PM_{2.5-10}$), and HNO_3 concentrations for the ensemble of flights over the SMA. Horizontal bars for the observations indicate 25th-75th percentiles.

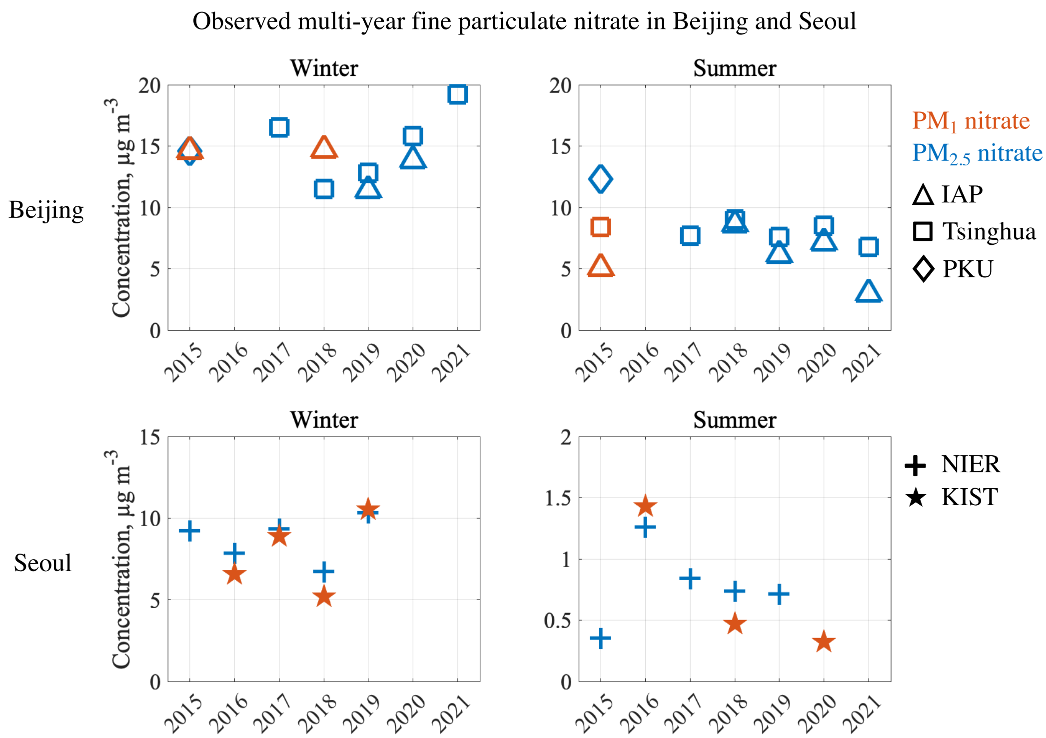
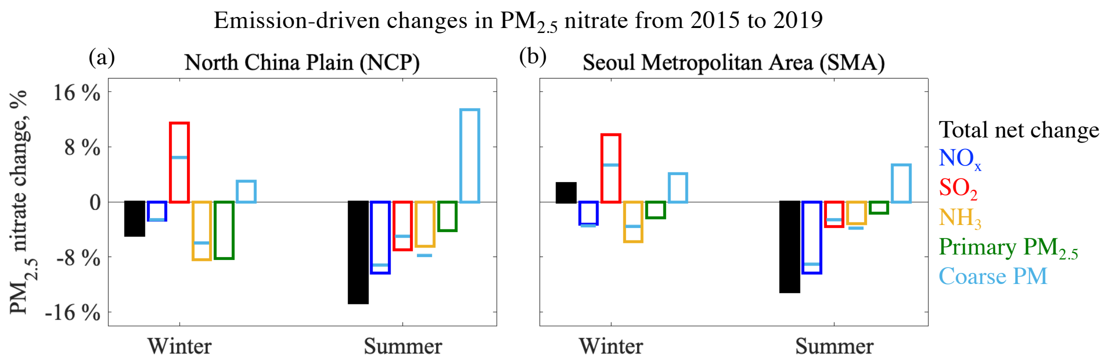


Figure 5. Long-term trend of fine particulate nitrate concentrations in Beijing and Seoul over the 2015-2021 period. Mean PM_1 or $PM_{2.5}$ concentrations in winter and summer are compiled from individual field campaigns in Beijing at the Institute of Atmospheric Physics (IAP), Tsinghua University (Tsinghua), and Peking University (PKU) sites and in Seoul at the National Institute of Environmental Research (NIER) and Korea Institute of Science and Technology (KIST) sites (Table S1). Note the differences in scales between panels.



Emission changes from 2015 to 2019

	NO_x	SO_2	NH_3	Primary $PM_{2.5}$	Coarse PM
NCP	-11%	-54%	-19%	-35%	-33%
SMA	-22%	-40%	0%	0%	-31%

Figure 6. Emission-driven changes in mean $PM_{2.5}$ nitrate from 2015 to 2019 over the NCP and SMA. Results are from GEOS-Chem sensitivity simulations including total and individual emission changes over the period, all for the same meteorological year (2019) and applied both to China and South Korea (so the effects of NH_3 and primary $PM_{2.5}$ over the SMA are due to long-range transport from China). Values are seasonal means for winter and summer. The blue lines superimposed on the NO_x, SO_2, and NH_3 sensitivity bars show the effects from simulations not accounting for the effect of HNO_3 uptake by anthropogenic coarse PM.