Foreign emissions exacerbate $PM_{2.5}$ pollution in China through nitrate chemistry

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Abstract

Fine particulate matter ($PM_{2.5}$) pollution is a severe problem in China. Research on the sources of Chinese $PM_{2.5}$ pollution has focused on the contributions of China's domestic emissions. However, the impact of foreign anthropogenic emissions has typically been simplified or neglected, partly due to the perception that the short lifetime of $PM_{2.5}$ (a few days) does not allow long-distance transport. Here we explore the role of foreign anthropogenic emissions in Chinese $PM_{2.5}$ pollution in 2015 using the GEOS-Chem chemical transport model. We validate the model simulations with a comprehensive set of observations of $PM_{2.5}$ and its composition, including sulfate, nitrate, ammonium, black carbon and primary organic aerosols, over China and its surrounding regions. We find that 8% of $PM_{2.5}$ (5 μg m^{-3}) and 19% of nitrate (2.6 μg m^{-3}) over eastern China in 2015 was contributed by foreign anthropogenic emissions. The contributions were the highest in January (6.9 μg m^{-3} $PM_{2.5}$, with 68% nitrate) and the lowest in July (2.7 μg m^{-3} $PM_{2.5}$, with 11% nitrate). Yet, only 30% of such foreign contributions in January was through direct atmospheric transport. The majority (70%) was instead through chemical interactions between foreign-transported aerosol precursors and China's domestic emissions of pollutants. Specifically, the transport of non-methane volatile organic compounds (NMVOCs) from foreign countries enhanced the atmospheric oxidizing capacity and facilitated the oxidation of Chinese nitrogen oxides (NO_x) to form nitric acid (HNO_3) over eastern China. The abundance of Chinese ammonia (NH_3) further partitioned nearly all HNO_3 gas to particulate nitrate, leading to the considerable foreign contributions of nitrate and $PM_{2.5}$ to eastern China. Over southwestern China, foreign anthropogenic emissions contributed 4.9 μg m^{-3} $PM_{2.5}$ concentrations (18% of total $PM_{2.5}$ mass) to Yunnan province, with 37% as organics and 27% as sulfate. Our findings suggest that foreign anthropogenic emissions play an important role in Chinese $PM_{2.5}$ pollution, because of direct aerosol transport and, more importantly, chemical interactions between transported pollutants and China's local emissions. Thus, foreign emission reductions will be very beneficial for improving Chinese air quality.

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1. Introduction

China has been severely affected by fine particulate matter ($PM_{2.5}$, particulate matter smaller than 2.5 μm in aerodynamic diameter) pollution over the past decades from processes of industrialization and urbanization (Geng et al., 2021; West et al., 2016). Over 1 million premature deaths associated with $PM_{2.5}$ pollution occur in China every year (Cohen et al., 2017; Yue et al., 2020; Zhang et al., 2017). In response, the Chinese government imposed stringent emission controls on primary particles and precursor gases in the 5-year Clean Air Action in 2013 (China State Council, 2013), leading to a nationwide emission reduction of 59% for sulfur dioxide (SO_2) and 21% for nitrogen oxides ($NO_x \equiv NO+NO_2$) from 2013 to 2017 (Zhang et al., 2019; Zheng et al., 2018). Correspondingly, annual mean $PM_{2.5}$ concentrations in China decreased by 30~50% from 2013 to 2017 (Ding et al., 2019; Geng et al., 2021; Li et al., 2019), avoiding 64 thousand (6.8%) premature deaths. Despite these remarkable achievements, population-weighted mean $PM_{2.5}$ concentration in China was still as high as 42.1 μg m^{-3} in 2017 (with 2.1 million associated premature deaths; Geng et al., 2021a), far exceeding the newly-revised threshold of 5 μg m^{-3} in the World Health Organization (WHO) Air Quality Guidelines (WHO, 2021). In 2020, the Chinese government further launched the "Beautiful China" strategy, which requires an annual mean $PM_{2.5}$ concentration of $\leq$ 35 μg m^{-3} in all cities by 2035. Yet, nearly 30% of cities in China exceeded that standard based on the 2021 national observation data (Ministry of Ecology and Environment, MEE, 2021). Thus, further improvement on air quality is pressing. However, air quality management has been progressively challenging with the widespread of end-of-pipe control technologies in dominant sources (industrial and power sectors; Xing et al., 2020) and the exhausting benefits of such technologies (Geng et al., 2021). Hence, more comprehensive and in-depth understanding of Chinese $PM_{2.5}$ pollution sources is urgently needed to help prioritize increasingly limited resources for accurate and effective mitigation action.

Recent research on the sources of $PM_{2.5}$ pollution in China has focused mostly on China's domestic anthropogenic emissions (An et al., 2019; Cheng et al., 2021b; Meng et al., 2019; Tang et al., 2022; Tong et al., 2018; Xing et al., 2020). A number of studies have explored how China could further reduce its own emissions of air pollutants through a wide range of energy transformation scenarios to achieve co-benefits of air quality improvement and climate mitigation (Cheng et al., 2021; Peng et al., 2018; Tong et al., 2018, 2020; Xing et al., 2020). Studies have also investigated factors hindering the effectiveness of emission reductions on air quality improvement in China, such as excess ammonia emissions (Bai et al., 2019; Gu et al., 2021; Yan et al., 2021a) and enhanced atmospheric oxidizing capacity associated with NO_x emission reductions in recent years (Huang et al., 2021; Le et al., 2020; Ren et al., 2021; Zang et al., 2022). A few works have studied the inter-provincial transport of pollution across China and found that the contribution of inter-provincial transport to $PM_{2.5}$ concentrations in the most severely polluted regions (such as Beijing) might have exceeded that of local emissions (Li et al.,

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2015). In addition, the range of inter-provincial transport of pollution was not confined within city clusters, such as Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta, but also extended over a long distance across city clusters (Wang et al., 2022). Yan et al. (2021b) further found that transboundary transport from Asian regions (18.5–19.2%, including Chinese regions outside the Wuhan City Cluster) contributed much more to ozone concentrations in the Wuhan City Cluster in Central China than the transport within the city cluster (2.5–3.1%), highlighting the importance of transboundary transport of pollutants to China.

However, the influence of transboundary transport to China air quality has been hardly investigated for $PM_{2.5}$. This is likely due to the perception that the relatively short lifetime of $PM_{2.5}$ (a few days) does not permit long-distance transport (Wang et al., 2019). Only a few have investigated the influence of pollutant emissions from neighboring countries (Jiang et al., 2013; Koplitz et al., 2017) on China, yet have typically focused on one particular sector, such as biomass burning emissions from South Asia (Jiang et al., 2013) or coal emissions from Southeast Asia (Koplitz et al., 2017). A comprehensive assessment of transboundary $PM_{2.5}$ pollution in China from foreign sources is lacking. In contrast, studies on the transboundary $PM_{2.5}$ pollution from China to neighboring countries have received considerable attention (Choi et al., 2019; Jiang et al., 2013; Kurokawa and Ohara, 2020; Park et al., 2014). This contrast is likely due to another perception that transboundary pollution from foreign countries to China is minor since China's domestic emissions far exceeded those from neighboring countries, such as Korea, Japan, India and the Southeast Asia (Kurokawa and Ohara, 2020; McDuffie et al., 2020). However, the pollutant emission pattern in China and neighboring countries may shift in the future. Emissions in China have decreased considerably (Zheng et al., 2018) and the trend is expected to continue with the launch of ambitious policies on air pollution (the 2035 "Beautiful China") and climate change (the 2060 carbon neutrality). In contrast, emissions from India and Southeast Asian countries have been estimated to increase in the future by various projections, given their fast-economic growth and a lack of clear commitments on either air quality or climate mitigation (IEA, 2021). For example, Koplitz et al. (2017) revealed that the projected increase of coal emissions in Southeast Asian countries will lead to 49780 excess deaths per year associated with $PM_{2.5}$ pollution in 2030, with 9000 (18%) of these excess deaths occurring in China. The transboundary pollution from neighboring countries to China may become increasingly prominent in the future. Thus, an effective air quality management action for the achievement of the "Beautiful China" target requires a clear understanding of the current contribution of foreign anthropogenic emissions to Chinese $PM_{2.5}$ pollution.

A comprehensive assessment of foreign contributions to $PM_{2.5}$ pollution in China relies on a complex representation of aerosol emissions and chemical reactions across a large spatial domain. The GEOS-Chem global chemical transport model has been widely applied to $PM_{2.5}$ studies over Asia (i.e., China, India, Southeast Asia, Korea and Japan; Choi et al., 2019; Koplitz et al., 2017; Miao et al., 2020; Venkataraman et al., 2018; Wang et al., 2004; Zhang et al., 2015),

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thereby applicable to such research. Although the model has been extensively validated for total $PM_{2.5}$ mass concentrations over China using observational data, compositional $PM_{2.5}$ across China and total $PM_{2.5}$ for other Asian countries are far less evaluated due to scarce observations (Cheng et al., 2021a; Koplitz et al., 2017; Miao et al., 2020). This limits the credibility of the model's representation of aerosol emission and chemical reactions across a large domain. Thus, a more comprehensive evaluation of the GEOS-Chem simulation is needed to support model estimates of the influence of transboundary pollution on air quality in China.

In this study, we use the GEOS-Chem model to quantify the contributions of foreign anthropogenic emissions to total and compositional $PM_{2.5}$ mass concentrations over China in 2015. We first evaluate our model simulations with comprehensive observations of total and compositional $PM_{2.5}$ concentrations across China and other Asian countries. Then, we quantify the contributions of foreign anthropogenic emissions to China $PM_{2.5}$ and compositional concentrations in 2015. Finally, we reveal the physical and chemical pathways leading to such contributions.

2. GEOS-Chem simulations

We conducted a series of simulations using the GEOS-Chem chemical transport model (v13.2.1; http://www.geos-chem.org) to 1) represent 2015 $PM_{2.5}$ and composition concentrations over Asia, 2) quantify the contributions of foreign anthropogenic emissions to total and compositional $PM_{2.5}$ concentrations over China, and 3) understand the role and the mechanisms of direct transport and chemical interactions in transboundary pollution in China. Simulation configurations are summarized in Table 1 and are elaborated as the following.

2.1 The GEOS-Chem simulation of ground-level $PM_{2.5}$

We used the flex-grid capability of the GEOS-Chem classic model v13.2.1 to simulate aerosol concentrations over Asia and the adjacent area (11° S–60° N, 30°–150° E; Figure 2a) at a horizontal resolution of 0.5° ×0.625° and at 47 vertical levels between the surface and ~ 0.01 hPa. The lowest vertical layer has a thickness of about 130 m. We regard the pollutant concentrations in this layer as "ground-level". Detailed descriptions of the flex-grid setup can be found at http://wiki.seas.harvard.edu/geos-chem/index.php/FlexGrid. Our flex-grid domain extended the traditionally-defined nested Asia domain (11° S–55° N, 60°–150° E) in the model (Figure 2a) to better represent the transport of anthropogenic pollutants from Central Asia to China that has not been studied yet. Our simulations were driven by assimilated meteorological data from MERRA-2 provided by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center. Convective transport in the model was computed from the convective mass fluxes in the meteorological archive as described by Wu et al. (2007). A non-local scheme was used to represent vertical mixing within the planetary boundary layer (PBL), as

it accounts for different states of mixing based on the static instability (Lin and McElroy, 2010). Boundary conditions were archived from global simulations at a resolution of 2°× 2.5°. We spun up every simulation for 1 month to remove the effects of initial conditions.

GEOS-Chem simulates $PM_{2.5}$ concentrations as the sum of sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic aerosol (OA ≡ primary OA + secondary OA), black carbon (BC), fine dust and fine sea salt component concentrations. The sulfate−nitrate−ammonium (SNA) aerosol system was simulated following Fountoukis and Nenes (2007) and Park et al. (2004), including heterogeneous chemistry with dinitrogen pentoxide (N_2O_5) uptake by aerosol, and hydroperoxyl radical (HO_2) uptake by aerosol. Gas−aerosol partitioning of SNA was simulated by the ISORROPIA II thermodynamic equilibrium scheme (Pye et al., 2009). We used a simple scheme to represent secondary organic aerosol formation (Heald et al., 2012) and used a spatially resolved ratio to calculate organic mass from organic aerosol concentrations (Philip et al., 2014). Natural dust simulation followed the Mineral Dust Entrainment and Deposition (DEAD) scheme (Fairlie et al., 2007). Sea salt aerosol simulation was described in Jaeglé et al. (2011). Dry deposition of gases and particles followed a standard resistance-in-series scheme, with updates from Jaeglé et al. (2018). Wet deposition was described in Liu et al. (2001), Wang et al. (2011) and Wang et al. (2014), with updates from Luo et al., (2020) that included a faster below-cloud scavenging of HNO_3. We calculated the simulated $PM_{2.5}$ and composition concentrations at 35% relative humidity (RH) for consistency with ground-based measurements.

2.2 Emissions for baseline simulation

We conducted a baseline simulation ("Base" run in Table 1) for 2015 January, April, July, October and treated the mean of the four months as annual mean. Our simulations were all at a resolution of 0.5° ×0.625°, unless otherwise specified. The baseline simulation used emissions as described below.

Anthropogenic emissions for China were taken from the Multi-resolution Emission Inventory (MEIC) for 2015 (Zheng et al., 2018), and for the rest of the world were taken from the Community Emissions Data System (CEDS) version 2 for 2015 (https://data.pnnl.gov/dataset/CEDS-4-21-21). Other emissions were default in GEOS-Chem. Fine anthropogenic fugitive dust emissions from combustion and industrial sources for countries except China (FR_AFCID) were taken from Philip et al. (2017), and from the MEIC inventory for China (CH_AFCID). Aircraft emissions were from the Aviation Emissions Inventory Code (AEIC) inventory (Stettler et al., 2011). Natural emissions include lightning NO_x from Murray et al. (2012), soil NO_x, biogenic non-methane volatile organic carbons (NMVOCs) and sea salt from off-line emissions developed by Weng et al. (2020), biomass burning emissions from the Global Fire Emissions Database version 4 (GFED4; Randerson et al., 2015), volcano emissions from Fisher et al. (2011), marine dimethyl sulfide (DMS) emissions from Breider et al. (2017)

and dust emissions using the Mineral Dust Entrainment and Deposition (DEAD) scheme (Zender et al., 2003).

2.3 Sensitivity simulations for the contributions of foreign anthropogenic emissions to China $PM_{2.5}$ and composition concentrations

We quantified contributions of foreign anthropogenic emissions to China total and compositional $PM_{2.5}$ concentrations by taking the difference of the baseline simulation ("Base" run in Table 1) and a sensitivity simulation that excluded foreign anthropogenic emissions from the baseline simulation ("CHAnth" run in Table 1). Such a foreign contribution is referred to as "FR_total". We also conducted simulations to quantify the sectoral contributions of foreign anthropogenic emissions to China's $PM_{2.5}$ concentrations. Sectoral contributions were calculated by taking the difference of a simulation that included one sector of foreign anthropogenic emissions (agriculture, industry, energy, traffic, residential combustion, solvent use, waste burning) at a time one and a simulation without foreign anthropogenic emissions ("CHAnth" in Table 1).

We further conducted sensitivity simulations to attribute the transboundary pollution to 1) direct transport of foreign $PM_{2.5}$ to China and 2) chemical interactions between transported foreign pollutants and Chinese emissions. We quantified the contribution of direct transport in transboundary pollution (referred to as "FR_transport") by taking the difference of a sensitivity simulation that excluded China anthropogenic emissions ("FRAnth" run in Table 1) and another sensitivity simulation that excluded both China and foreign anthropogenic emissions ("NoAnth" run in Table 1). Transboundary pollution through chemical interactions with China's local emissions (referred to as "FR_chemistry") were calculated as the differences between total foreign anthropogenic contributions (FR_total) and direct transport contributions (FR_transport).

We conducted sensitivity simulations to understand main pollutants driving the chemical interactions of transboundary pollution with Chinese emissions. Specifically, we quantified the contributions of foreign anthropogenic emissions of NMVOCs and NO_x to O_3, NO_3 and N_2O_5, HNO_3 and NO_3^- concentrations in China. Such contributions include both the direct transport of foreign pollutants and chemical interactions between foreign-transported and China domestic emissions of pollutants. We quantified the total contributions by foreign anthropogenic emissions of NMVOCs as the difference between a simulation with full emissions and a simulation that excluded foreign anthropogenic emissions of NMVOCs ("Base" – "No_FRAnthNMVOCs" runs in Table 1). To quantify the direct transport share of the total contributions, we excluded China's domestic emissions to avoid interactions with foreign-transported pollutants, and calculated it as the difference between simulations that included and excluded foreign anthropogenic emissions ("FRAnthNMVOCs" – "NoAnth" runs in Table 1). The contributions of chemical interactions between foreign NMVOCs and China's domestic emissions were quantified as the difference

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between the total contributions and the direct transport share of the total contributions. Similarly, the total contributions by foreign anthropogenic emissions of NO_x were calculated as "Base" – "No_FRAnthNO_x" runs in Table 1, with the corresponding direct transport share of the total contributions calculated as "FRAnthNO_x" – "NoAnth" runs in Table 1, and the chemistry share calculated as the difference between the total and the direct transport share.

To reduce computational costs, we conducted NMVOCs-related sensitivity simulations at a resolution of 2°× 2.5° (Table 1). The differences of $PM_{2.5}$ over eastern China between 2°× 2.5° and 0.5° ×0.625° resolutions is about 5% (3 μg m^{-3}). Compositional differences are within 20%, with the largest difference in nitrate (2.6 μg m^{-3}; 18%) and the lowest in black carbon (0.1 μg m^{-3}; 4%). These differences are within the reasonable range associated with spatial resolutions.

3. Ground-level observations of $PM_{2.5}$ and composition in China and other Asian countries

We evaluated the modeled $PM_{2.5}$ and composition concentrations in the base year (2015) using ground-level $PM_{2.5}$ observations from the network of China National Environmental Monitoring Center (CNEMC), the literature search and the World Health Organization (WHO) database.

Ground-level $PM_{2.5}$ observations were obtained from the CNEMC network (http://106.37.208.228:8082/). We used hourly measurements for 2015 January, April, July and October. $PM_{2.5}$ mass concentrations were measured using the micro-oscillating balance method or the β- absorption method (Zhang and Cao, 2015). We further applied quality controls to hourly CNEMC data. Specifically, we removed a day if there were < 14 valid data within the day and removed a month if there were < 25 days of valid data within the month. These in whole removed 12% $PM_{2.5}$ hourly data, and finally retained observations for 1179 sites in 314 cities for model evaluation (Fig. S1). To compare with the GEOS-Chem simulated $PM_{2.5}$ concentrations, we calculated the grid-averaged and monthly-averaged $PM_{2.5}$ concentrations from the CNEMC to match spatially and temporally with the model.

We collected compositional $PM_{2.5}$ observations from publicly available studies, as shown in Table S1. We selected observations that spun at least one-year or seasonal/monthly measurements centered at January, April, July or October to match our model simulations. A total of 56 observation data from 17 cities in 16 provinces for 2014–2016 were collected for major $PM_{2.5}$ chemical composition, including sulfate, nitrate, ammonium, organic aerosol (OA), and black carbon. We sampled the GEOS-Chem simulated concentrations from locations and periods (monthly or annual) of observations for evaluation.

To evaluate the modelled $PM_{2.5}$ concentrations outside China, we collected $PM_{2.5}$ measurement data for 2013–2016 from the WHO ambient air pollution in cities database

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(https://www.who.int/data/gho/data/themes/air-pollution). This database provides annual average $PM_{2.5}$ concentrations for more than 500 cities globally. We retained $PM_{2.5}$ measurements that were directly measured and excluded data inferred from PM_{10} concentrations. We also retained the sites within our simulation domain and the grid cells where anthropogenic emissions exceeded natural emissions (by comparing $PM_{2.5}$ concentrations in "FRAnth" and "NoAnth" runs in Table 1). A total of 83 sites covering 10 Asian countries (Bangladesh, Indonesia, India, Japan, Korea, Malaysia, Myanmar, Philippine, Thailand and Vietnam) were finally selected in our study as shown in Fig. 2a. Because of the differences in measurement approaches between jurisdictions, and the absence of details regarding measurement data (Brauer et al., 2016), we compared annual average $PM_{2.5}$ concentrations in the WHO dataset with those estimated from a hybrid of satellite observations, a chemical transport model and ground-based measurements (van Donkelaar et al., 2021). We removed the sites if the differences were more than 40%. The final sites retained in our study are shown in Fig. 2b.

4. $PM_{2.5}$ and composition concentrations across China and other Asian countries

Figure 1 shows modelled and observed annual mean concentrations of $PM_{2.5}$ and composition across China. The modelled total and compositional $PM_{2.5}$ are for 2015. Observations of total $PM_{2.5}$ are for 2015 and of composition are for 2014–2016. The spatial distribution of modelled $PM_{2.5}$ exhibits a broad high across eastern China, driven primarily by sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), and organics (OA). $PM_{2.5}$ over the Sichuan Basin is also high, contributed mostly by sulfate, ammonium and organics. The enhanced $PM_{2.5}$ concentrations over the west are dominated by mineral dust. Compared to observations, the modelled $PM_{2.5}$ well reproduces the spatial variation of $PM_{2.5}$ across China, with a correlation coefficient (r) of 0.73 and a normalized mean bias (NMB) of 15.7%. The slight overestimation is primarily contributed by sites in the Sichuan Basin (Figure S1a), where local emissions (anthropogenic and vegetation emissions; Wang et al., 2018), meteorological conditions (humid and stagnant; Chen et al., 2014; Liao et al., 2017), combined with the special terrain (plain surrounded by hills; Chen et al., 2014; Wang et al., 2017) make it hard for the model to represent aerosol processes, especially the secondary aerosol formation process (Liao et al., 2017; Tao et al., 2017; Wang et al., 2018). The seasonal evaluation of simulated $PM_{2.5}$ is shown in Fig. S2 and exhibits a good consistency with observations across seasons (r = 0.61~0.77), demonstrating the model's capability in capturing the seasonal pattern of aerosol processes.

We further evaluate our modelled compositional $PM_{2.5}$ with observations from 56 sites across 16 provinces and municipalities in China to better understand the performance of the simulation. The scatterplots of composition comparison in Fig. 1 show a mixture of annual and monthly data depending on the availability of observations data from the literature. We find that the modelled compositional $PM_{2.5}$ reasonably reproduce the vast spatial variation of $PM_{2.5}$ composition from observations. Sulfate and nitrate simulations are particularly improved over previous model

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studies where sulfate was significantly underestimated (NMB~-40%) and nitrate was substantially overestimated (NMB~80%; Gao et al., 2018; Miao et al., 2020). The better representation of nitrate is owing to the faster nitrate removal in the Luo et al. (2020) deposition scheme. Organics are also well reproduced by the model (NMB=4.7%), suggesting the effectiveness of the simple SOA scheme in representing total SOA mass. Ammonium and black carbon show relatively large discrepancies (NMB = 24.9% and -12.8%, respectively), potentially reflecting model biases in chemical reactions or gas-particle partition for ammonium formation (Miao et al., 2020) and emission inventories for BC (Zhang et al., 2019). In addition, outstanding differences in observation approaches (i.e., thermal, optical or incandescence measurements for black carbon; different relative humidity in measurements; Bond et al., 2013; Snider et al., 2016) across literature are another major reasons for the discrepancies.

The evaluation of modelled $PM_{2.5}$ concentrations for other Asian regions has been rarely conducted due to limited observations (Koplitz et al., 2017). Here, we compare our modelled $PM_{2.5}$ concentrations with observations from 10 Asian countries around China to understand the model performance in regions whose pollution could influence China through transboundary transport. Figure 2 shows good agreement between modelled and observed total $PM_{2.5}$ mass across countries, with a correlation coefficient of 0.76 and a NMB of 3.5%, despite large uncertainties in the measurements collected by different countries. There is an underestimate at coastal sites (e.g., in Philippine; Fig. 2a) where sea salt aerosols potentially make larger contributions than our simulations. Specifically, our simulated $PM_{2.5}$ is very consistent with observations in the Southeast Asia (NMB = 2.7%; including Indonesia, Myanmar, Philippines, Thailand and Vietnam), India (3.8%) and other countries in South Asia (NMB=7.9%; including Bangladesh, Bhutan, Maldives, Nepal, Pakistan, Sri Lanka). Simulations over Japan and South Korea show relatively larger bias (NMB = 17.4% for Japan and 40% for South Korea). Zhai et al. (2021) attributed the 43% bias in the GEOS-Chem simulation of surface $PM_{2.5}$ over South Korea in their study to nighttime nitrate formation, although their updates of faster below-cloud scavenging of HNO_3 (Luo et al., 2020) corrected the overall nitrate bias in East Asia.

5. Contributions of foreign anthropogenic emissions to total and compositional $PM_{2.5}$ in China

Figure 3 shows the contributions of foreign anthropogenic emissions to total and compositional $PM_{2.5}$ concentrations over China in 2015. On the national level, foreign anthropogenic emissions contribute about 2.4 $\mu g\ m^{-3}$ $PM_{2.5}$ to China in 2015, accounting for 6.2% of the national average $PM_{2.5}$ concentration. The foreign influence exhibits prominent spatial heterogeneity, with the largest contribution of 5.0 $\mu g\ m^{-3}$ $PM_{2.5}$ (8%) to eastern China (outlined in Fig. 3; including Anhui, Hebei, Henan, Jiangsu, Liaoning, Shandong, Beijing and Tianjin). Considering the WHO newly-revised guideline for $PM_{2.5}$ annual exposure level ($\leq$ 5 $\mu g\ m^{-3}$), foreign anthropogenic emissions alone would make $PM_{2.5}$ concentrations over eastern

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China reach the WHO standard, threating the health of nearly 500 million residents there. Transboundary pollution is also outstanding along the southwestern border, contributing 4.9 μg m^{-3} $PM_{2.5}$ (18%) to Yunnan province, mostly driven by anthropogenic emissions from South Asia (i.e., India).

The transboundary pollution over eastern and the southwestern China is contributed by different chemical components of $PM_{2.5}$, as shown in Fig. 3. Eastern China is mainly driven by nitrate and ammonium, explaining 70% of transboundary $PM_{2.5}$. Particularly, 18% of nitrate and 12% of ammonium concentrations over eastern China in 2015 are driven by transboundary pollution. However, such enhancement over eastern China is not observed for organic matter, partly due to the simplified SOA formation scheme that is not able to fully represent the chemical formation of SOA, which is a common issue in chemical transport models (Pennington et al., 2021; Shrivastava et al., 2017). Leibensperger et al. (2011) and Koplitz et al. (2017) found similar nitrate enhancement yet to a much lesser extent (< 0.2 μg m^{-3}). They attributed the nitrate enhancement to the increase in ozone that speeded up the rate at which Chinese local NO_x emissions were converted to nitrate. We will discuss the mechanism in more detail in the next section. The sulfate contribution is very small (< 0.5 μg m^{-3}). Leibensperger et al. (2011) proposed a reason that H_2O_2 (the key oxidant for sulfate formation) is abundant most of the year over eastern China, thereby insensitive to additional transboundary source. When H_2O_2-limited conditions prevail in winter, cloud cover is infrequent, limiting the in-cloud oxidation of SO_2. Thus, the influence of transboundary pollution to sulfate over eastern China is weak. The transboundary $PM_{2.5}$ over the southwestern China is primarily contributed by organics (1.8 μg m^{-3}; 37% of transboundary $PM_{2.5}$), which is consistent with previous studies that found massive biomass burning emissions in South Asia contributed considerable organics to the southern China (Jiang et al., 2013). Sulfate contributes 27% of transboundary $PM_{2.5}$ over the southwestern China, where the inflow of hot and humid atmosphere from the South Asia facilitates the in-cloud formation of sulfate (Jiang et al., 2013). In addition, anthropogenic fugitive dust emissions from foreign countries make an influence to China $PM_{2.5}$, accounting for 14% of the $PM_{2.5}$ increase in both eastern and southwestern regions, as shown in Figure S3.

We further investigate the seasonal variation of transboundary pollution in China to understand potential sources of foreign contributions. Figure 4 presents the seasonal enhancement of $PM_{2.5}$, nitrate and ammonium over China in 2015 driven by foreign anthropogenic emissions. Transboundary pollution of $PM_{2.5}$ over eastern China is the largest in January (6.9 μg m^{-3}) and gradually decreases to the smallest in July (2.7 μg m^{-3}). Affected regions also change prominently with seasons. In January, the boundary of transboundary $PM_{2.5}$ larger than 9 μg m^{-3} extends to the south of eastern China domain outlined in Fig. 4, whereas in July, that boundary shrinks to a much smaller region along the east coast. Source attribution (Fig. S4) reveals that all of the seven major anthropogenic emission source sectors in foreign countries contributed similarly (10-17% for each sector) to China $PM_{2.5}$ in January, yet their

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relative importance exhibits spatial heterogeneity. Over eastern China, industry and solvent use in foreign countries are the largest sources (likely because of their considerable amount of NMVOCs emissions that increase the atmospheric oxidizing capacity over eastern China, as will be discussed in Section 6), whereas over Yunnan province, residential combustion in foreign countries makes the largest contribution.

These seasonal characteristics of transboundary $PM_{2.5}$ are similar to those of nitrate and ammonium. In January, 68% transboundary $PM_{2.5}$ over eastern China is contributed by nitrate (4.7 μg m^{-3}) and 19% by ammonium (1.3 μg m^{-3}), yet these fractions decrease to 11% for both nitrate and ammonium in July. The transboundary nitrate for the majority of eastern China exceeds 5 μg m^{-3} in January, yet decreases to less than 1 μg m^{-3} and even negative in July. These prominent seasonal variations of transboundary $PM_{2.5}$, nitrate and ammonium reflect different processes controlling the transboundary pollution in winter and summer in China.

6. Physical and chemical mechanisms of foreign anthropogenic contributions to $PM_{2.5}$ over eastern China

We investigate the relative importance of direct transport and chemical interactions in driving the considerable transboundary pollution over eastern China in January and July, as shown in Figure 5. In January, the transboundary $PM_{2.5}$, nitrate and ammonium are predominantly (71–97%) driven by chemical interactions, suggesting that the transboundary pollution in winter over eastern China is not through direct transport of nitrate and ammonium, but through chemical interactions between directly transported precursors from foreign countries and local emissions in China. In July, however, nearly all transboundary $PM_{2.5}$ over eastern China is driven by direct transport, with 30% of the direct transport contributed by anthropogenic fugitive dust from foreign countries (Fig. S3). The transboundary nitrate is still primarily driven by chemical interactions in July (89%), yet the magnitude is too small to substantially affect total $PM_{2.5}$.

We further explore the changes in key chemical species for nitrate formation to understand the chemical mechanism driving the considerable nitrate enhancement over eastern China. Figure 6 shows the contributions of transboundary pollution to concentrations of precursor gases (NO_x), oxidants (O_3, N_2O_5 and NO_3) and oxidized products (total inorganic nitrate including gas-phase HNO_3 and particulate NO_3^-) in nitrate chemistry over eastern China in January and July. Fig. 6 (top-left) shows that, in January, chemical interactions between foreign-emitted NMVOCs and China's local aerosols promote the O_3 production over eastern China by 2.6 ppb, leading to a drop of NO_x (-1.8 μg N m^{-3}) and an increase of N_2O_5+NO_3 and HNO_3+NO_3^- (0.88 μg N m^{-3}) concentrations over eastern China. The presence of excess ammonia over eastern China due to the reduction of SO_2 (Liu et al., 2018, 2019) further partitions nearly all HNO_3 gas to into particulate nitrate (nitrate ammonium, NH_4NO_3), leading to about 1 μg N m^{-3} (or 2.6 μg m^{-3}) nitrate increase there. These results reveal that, in January, when the O_3 production is in a

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NMVOCs-limited regime, the additional NMVOCs from transboundary sources promote the O_3 production and the subsequent nitrate formation over eastern China, which otherwise would be limited by the lack of O_3 (Jin and Holloway, 2015; Li et al., 2018; Wang et al., 2017).

In addition to NMVOCs, NO_x is another important precursor of O_3. We therefore conducted sensitivity simulations to understand the role that foreign anthropogenic emissions of NO_x play in the nitrate enhancement in eastern China. As shown in Fig. 6 (top-bottom) for January, the promotion of nitrate formation is very small with the additional influence of NO_x emitted from foreign sources. This is due to the high local emissions of NO_x over eastern China in January that suppresses the O_3 formation under a NO_x-saturated regime.

In July, although foreign NMVOCs contribute about 4 ppb O_3 to eastern China through the chemistry process, they hardly lead to much difference in nitrate concentrations (Fig. 6 bottom). This is because that a lack of excess aerosols (compared to winter) limits the transformation of N_2O_5 and NO_3 to HNO_3 on aerosol surface. In addition, the oxidation of NO_x by OH is sufficiently fast in summer and the abundance of OH over eastern China in summer makes the NO_x oxidation process insensitive to the added O_3 from foreign countries. Therefore, a lack of excess aerosols and the abundance of OH in summer makes the transboundary transport of ozone precursors minor in contributions to nitrate concentrations over eastern China.

7. Conclusions

An effective air quality improvement action requires an accurate understanding of $PM_{2.5}$ sources. This work complements our understanding of $PM_{2.5}$ sources in China by investigating the influence of foreign transboundary transport through the GEOS-Chem simulation. Our extensive and comprehensive evaluation of the GEOS-Chem model for $PM_{2.5}$ total and compositional mass concentrations in China and 10 additional Asian countries showed a reasonable consistency with observations. Based on model simulations, we found that foreign anthropogenic emissions played an important role in Chinese $PM_{2.5}$ pollution, because of direct aerosol transport and, more importantly, chemical interactions between transboundary pollutants and China's local emissions. Over eastern China, the transport of NMVOCs from foreign anthropogenic emissions increased O_3 concentrations by 2.6 ppb through chemical interactions with China's local aerosols. The additional O_3, combined with high local emissions of NO_x and ammonia led to a nitrate enhancement of 2.6 $\mu g\ m^{-3}$ in January. Over the southwestern China, transboundary transport contributed 18% $PM_{2.5}$ to Yunnan province in 2015, mostly driven by the direct transport of aerosols from anthropogenic emissions in South Asia. There are a few sources of uncertainty in this study, for example the wet deposition of nitrate and the simplified secondary organic aerosol formation scheme, but they do not manifest themselves as systematic biases.

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In light of the physical and chemical mechanisms of transboundary pollution in China, further improvements of air quality for the "Beautiful China" target requires different emission reduction strategies for different regions. Over eastern China, reductions of both foreign and domestic anthropogenic emissions can reduce transboundary $PM_{2.5}$ pollution to China, since a considerable amount of transboundary transported $PM_{2.5}$ is formed through the chemical interactions between pollutants from both sources. To reduce the transboundary influence driven by chemical interactions, source sectors with considerable emissions of NMVOCs in foreign countries, such as industry and solvent use, are of particular importance. Over the southwestern China, foreign emission reductions will be necessary for improving air quality there since the transboundary pollution was primarily through the direct transport of aerosols from foreign countries. Given the declining trend of Chinese anthropogenic emissions in the present and the future, the likely rising emissions from adjacent countries in the future could become an increasingly important problem for Chinese air quality protection if no action is taken to avoid emissions increases in adjacent countries. Our study points to the need to carefully consider the potential influence of transboundary pollution when making the long-term air quality improvement strategies in China. Future studies could further investigate the impact of transboundary pollution transport to China under future emission scenarios, or to extend this study to other regions in the world where transboundary pollution could potentially increase domestic $PM_{2.5}$ pollution through nitrate chemistry. Investigation into the bi-directional $PM_{2.5}$ contributions between countries would also be helpful for developing practical policies for regional cooperation on emission reductions.

Data availability

Data presented in this paper are available upon request to the corresponding author.

Author contributions

J.L. led the study. J.X. and J.L. designed the study. J.X. performed the model simulations and conducted the data analysis. J.A. collected observation data of $PM_{2.5}$ composition from the literature. H.K. processed $PM_{2.5}$ observations from the CNEMC website. J.X. wrote the manuscript with inputs from J.L. All authors commented on the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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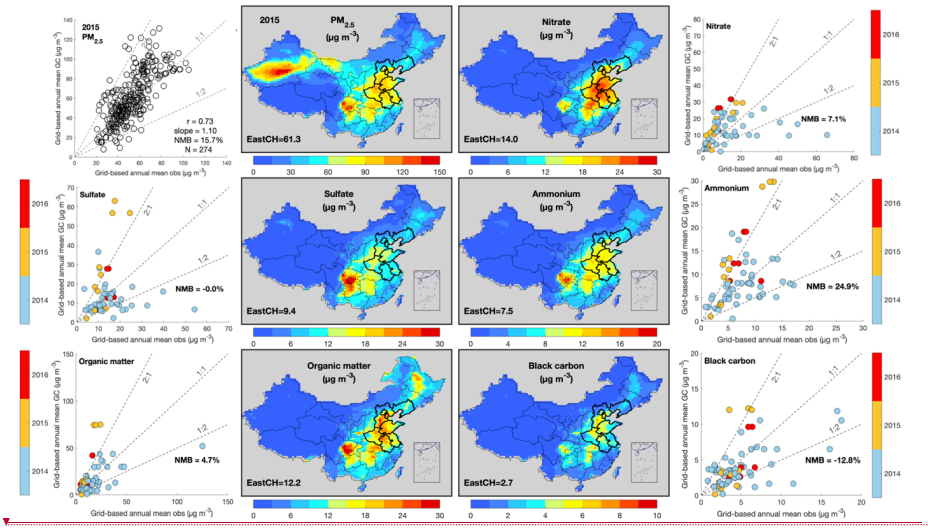

Figure 1. Total and compositional $PM_{2.5}$ concentrations in China. Spatial distributions of $PM_{2.5}$ and composition concentrations are annual mean concentrations simulated by the GEOS-Chem model for 2015 January, April, July and October with a resolution of 0.5° x 0.625°. We regard the mean of the four months as annual mean. Thick black lines outline eastern China discussed in this work. Text in the bottom left corner of each map refers to mean concentrations (μg m^{-3}) over eastern China. The scatterplot of $PM_{2.5}$ compares the simulated annual mean concentrations with collocated and coincident observations for 2015 from the CNEMC network. Scatterplots of composition compares both annual and monthly concentrations of the observed and the coincident simulated concentrations according to the availability of observations from the literature for 2014-2016.

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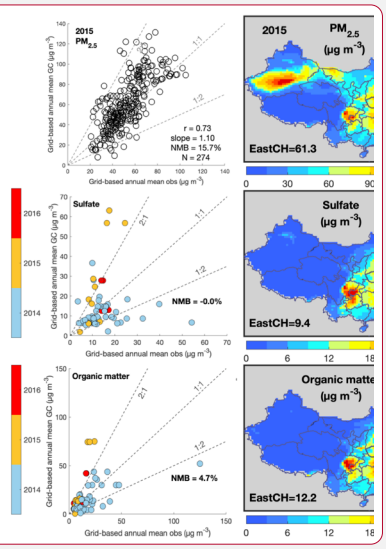

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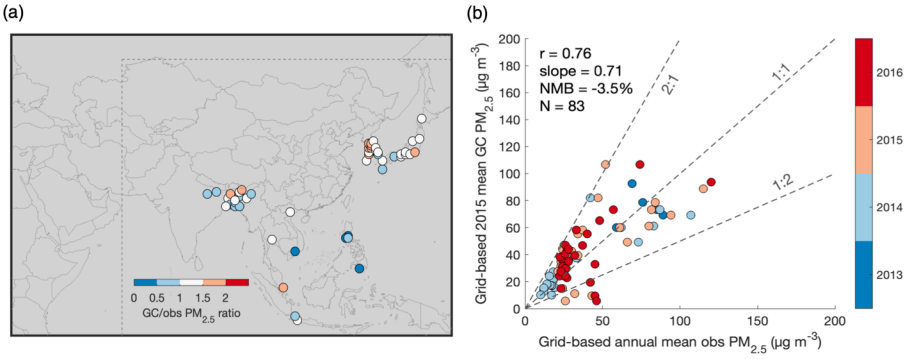

Figure 2. Annual mean $PM_{2.5}$ concentrations at anthropogenic emission dominated sites in the simulation domain outside China. (a) The spatial distribution of simulated and observed $PM_{2.5}$ concentration ratios. The simulated concentration at each measurement site represents the 0.5°x 0.625° grid cell covering that site. Dashed lines represent the default nested Asia domain (11° S–55° N, 60°–150° E) in the model. The flex-grid domain in our study is shown as the entire domain of the map. (b) Scatterplot comparing simulated concentrations for 2015 with collocated observations from the WHO for 2013–2016.

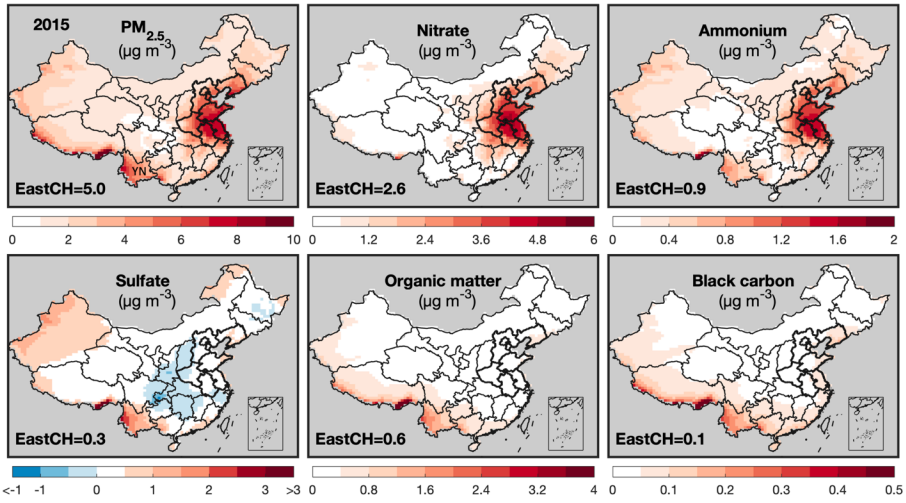

Figure 3. Simulated annual mean contributions of foreign anthropogenic emissions to China's total and compositional $PM_{2.5}$ concentrations in 2015. Thick black lines outline eastern China discussed in this work. Text in the bottom left corner of each panel refers to mean concentrations (μg m^{-3}) over eastern China contributed by foreign anthropogenic emissions. The foreign impact

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on China's anthropogenic dust concentrations are shown in Figure S3. YN in the $PM_{2.5}$ subplot refers to the location of Yunnan province.

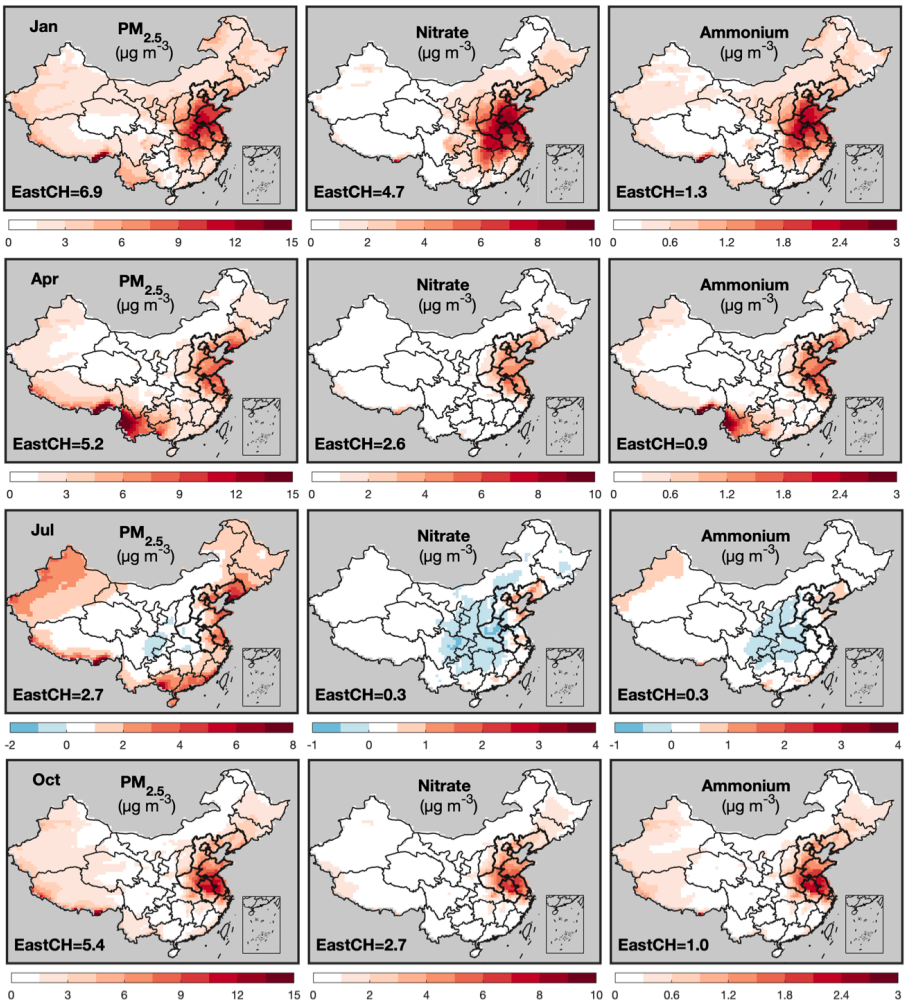

Figure 4. Same as Fig. 3, but for January, April, July and October as denoted by text in the top left corner of each row.

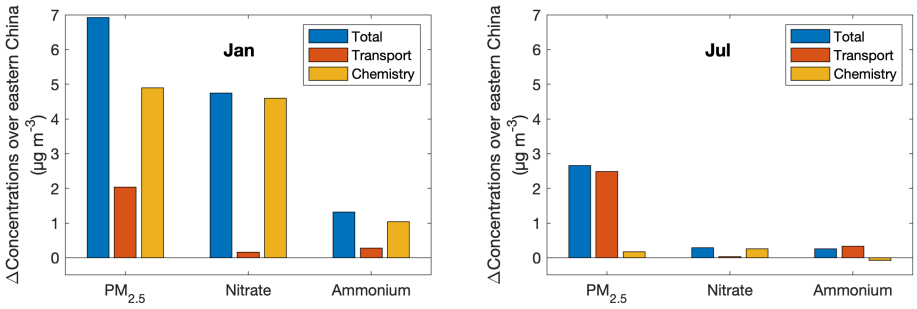

Figure 5. Foreign anthropogenic contributions to $PM_{2.5}$, nitrate and ammonium concentrations in January and July over eastern China. Total concentration contributions of foreign anthropogenic emissions are split into contributions from direct transport and chemical interactions according to the legend. The contributions to other $PM_{2.5}$ components are shown in Figure S5.

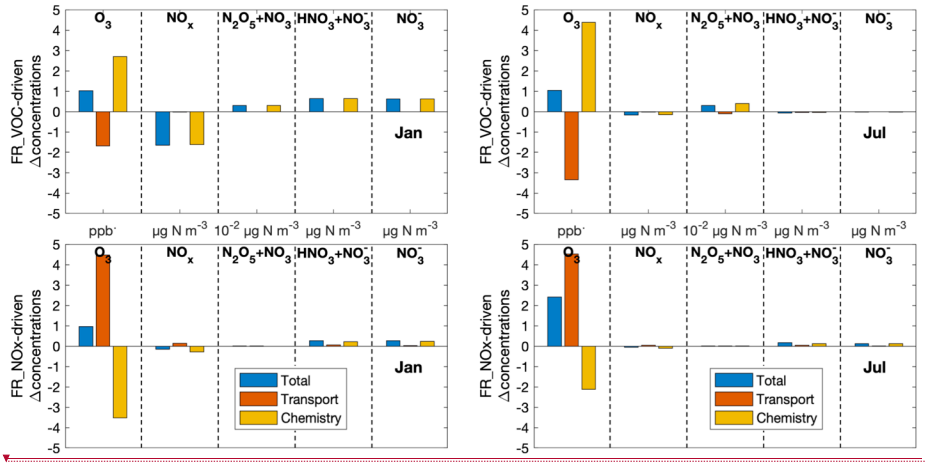

Figure 6. Contributions of foreign anthropogenic emissions of NMVOCs (top) and NO_x (bottom) to atmospheric oxidizing capacity and oxidation products over eastern China as simulated by the GEOS-Chem model for 2015 January (left) and July (right). Total foreign anthropogenic contributions are split into contributions from direct transport and chemical interactions according to the legend. O_3 concentrations are 24-hour average concentrations. Nitrogen-related species are presented in the unit of μg N m^{-3} for the convenience of nitrogen budget calculation.

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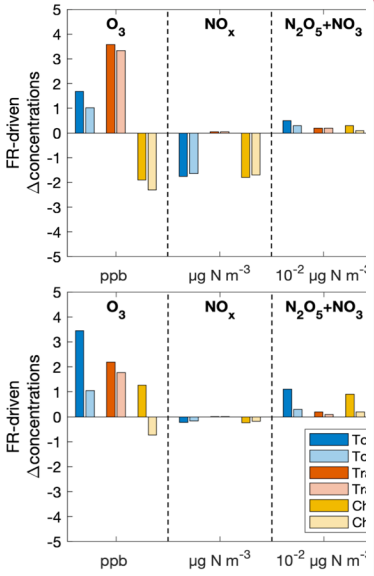

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Figure 6. Contributions of foreign anthropogenic emissions of total aerosols and NMVOCs to atmospheric oxidizing capacity and oxidation products over the eastern China as simulated by the GEOS-Chem model for 2015 January (top) and July (bottom). Total foreign anthropogenic contributions are split into contributions from direct transport and chemical interactions according to the legend. O_3 concentrations are 24-hour average concentrations. Nitrogen-related species are presented in the unit of μg N m^{-3} for the convenience of nitrogen budget calculation.

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Table 1. Configuration summary of GEOS-Chem simulations in this study. Emission abbreviations are elaborated in Section 2.2.

Foreign contribution type			FR_Total		FR_Transport		FR_NMVOCs		FR_NO$_x$	
Simulation name			Base	CHAnth	FRAnth	NoAnth	No_FRAnth-NMVOCs	FRAnth-NMVOCs	No_FR-AnthNO$_x$	FRAnth-NO$_x$
Emissions	CHAnth (MEIC+CH_AFCID)		Y	Y	N	N	Y	N	Y	N
	FRAnth (CEDS+FR_AFCID)	VOC	Y	N	Y	N	N	Y	Y	N
		NO$_x$					Y	N	N	Y
		OTR[1]					Y	N	Y	N
	Other (Shipping, aircraft, natural)		Y	Y	Y	Y	Y	Y	Y	Y
Resolution			0.5° x 0.625°		0.5° x 0.625°		2° x 2.5°		2° x 2.5°	
Simulation period			2015/1,4,7,10		2015/1,4,7,10		2015/1,7		2015/1,7	
Met fields			MERRA2		MERRA2		MERRA2		MERRA2	

[1]OTR refers to other species including SO_2, NH_3, BC, OC, CO, etc.

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