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Contributions of international sources to $PM_{2.5}$ in South Korea

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HIGHLIGHTS

- A global model estimated Chinese and domestic contributions to PM2.5 in Korea.
- Influence from China on $PM_{2.5}$ in Korea was the highest during winter and spring.
- Contributions from China were ~60% in January/February and ~20% in August.
- Domestic contributions were also the highest during winter months.

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ABSTRACT

The air quality in Republic of Korea, especially in cities such as Seoul, has been a serious public health concern over the years. The key pollutant in the atmosphere leading to poor air quality in Korea is fine particulate matter ($PM_{2.5}$). Here, we use a 3-D global chemistry model (GEOS-Chem) to conduct source attribution to $PM_{2.5}$ in Korea from international and domestic emissions. The modeling was done for 2015 and 2016 to account for different meteorological conditions. We ran the GEOS-Chem model for both years, conducted model evaluation using ground and aloft observations, and then conducted sensitivity simulations without domestic anthropogenic emissions and Chinese anthropogenic emissions, respectively. Results show that the Chinese influence on $PM_{2.5}$ in Korea varies from month to month with the highest contribution during spring when observed concentrations are also the highest. Chinese contributions to $PM_{2.5}$ concentrations in South Korea reach a maximum of up to ~60% in January and February and gradually decrease until August when they reach a minimum at about 20%. On an annual basis, our analysis estimated that in 2016, Chinese anthropogenic emissions contributed 45% to $PM_{2.5}$ in South Korea. The 2016 contribution from China was generally 3–5% lower than in 2015 because of emissions reductions in China. Compared to the Chinese contribution, the rest of the world contributions (which also include contributions from natural emissions worldwide) were minor except for summer in the South Sea.

1. Introduction

Fine particulate matter ($PM_{2.5}$) has been a significant air pollution concern in Korea over the past few decades. To inform policies to reduce $PM_{2.5}$ concentrations in the country, it is important to fully understand the contribution of different emission sources under different meteorological conditions. This information can be used to develop effective control strategies to improve air quality. There are both domestic and international sources that contribute to $PM_{2.5}$ in Korea, and it has been hypothesized that emissions from China and other neighboring countries can dramatically impact air quality in Korea.

Lee (2014) analyzed the $PM_{2.5}$ data measured in the Seoul Metropolitan Area (SMA) from November 2005 to March 2012 and showed an annual average concentration of 27 μg/m^3, roughly three times the

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WHO standard. Son et al. (2012) analyzed $PM_{2.5}$ and its components in Seoul, Korea from August 2008 to October 2009 comparing those to U.S. conditions and found that chemical characteristics of Seoul's $PM_{2.5}$ were more similar to $PM_{2.5}$ found in the western United States than in the eastern United States, although overall $PM_{2.5}$ levels in Seoul were higher than in the United States. Kim et al. (2018) analyzed $PM_{2.5}$ measurements for four 1-month periods in each season between October 2012 and September 2013 in Seoul and found that the four-season average concentration of $PM_{2.5}$ was 37 μg m ^{3}, with higher concentrations in winter and lower in summer. Their analysis also suggested the influence of transported secondary aerosol from emission sources from upwind urban areas and from China across the Yellow Sea. Bae et al. (2019) analyzed $PM_{2.5}$ concentrations and its major chemical constituents in the SMA from 2012 to 2016 and found that the mean $PM_{2.5}$ concentration in the SMA was 33.7 μg/m^3.

Given that $PM_{2.5}$ concentrations in major cities in Korea have stayed high over the years, much effort has been spent recently to understand the sources and processes contributing to high $PM_{2.5}$ to develop effective policies to reduce its concentrations in the country. To improve air quality in Korea, the government promulgated a stringent environmental policy in 2018, which includes $PM_{2.5}$ standards that should not exceed 15 μg/m^3 on an annual average basis and 35 μg/m^3 for a 24-hr average concentration. Although a decreasing trend of $PM_{2.5}$ concentrations in Seoul was observed in the past (Kim and Lee, 2018), the annual mean $PM_{2.5}$ concentration was 25 μg/m^3 in 2017–~70% higher than the $PM_{2.5}$ standard. Heo et al. (2009) used positive matrix factorization (PMF) to identify sources contributing to $PM_{2.5}$ in Seoul, Korea using every third day $PM_{2.5}$ components data collected from March 2003 to December 2006. They found that major contributors to $PM_{2.5}$ were secondary nitrate (21%), secondary sulfate (21%), gasoline fueled vehicles (17%), biomass burning (12%), and diesel emissions (8%). Using back trajectories, they also showed that the elevated secondary sulfate and nitrate concentrations were possibly due to industrial sources in China. Han et al. (2011) used back trajectory analysis to estimate source contributions and showed that major industrial sources in eastern China could be potential contributors to high $PM_{2.5}$ in rural sites in Korea.

Kim et al. (2017) used the community multiscale air-quality (CMAQ) model to estimate contributions from domestic and foreign emissions to $PM_{2.5}$ in the SMA using the brute force method and showed that foreign emissions contributed ~60% of SMA concentrations of $PM_{2.5}$ in 2014 on average with a maximum of ~70% in March. Bae et al. (2019) performed a set of sensitivity simulations with CMAQ for the east Asia domain for the 2012–2016 period and showed that the annual averaged impact of Chinese emissions on SMA $PM_{2.5}$ concentrations ranged from 41% to 44% during the five years. The KORea-US cooperative Air Quality field study (KORUS-AQ) in May–June 2016 was designed to investigate transboundary influence and source contributions to $PM_{2.5}$ in South Korea under various meteorological conditions (NIER and NASA, 2017). Choi et al. (2019) modeled the KORUS-AQ period using the 3-D chemical transport model, GEOS-Chem, and its adjoint, which have been updated with the latest regional emission inventory, diurnal variations of NH_3 emissions, the implementation of particulate nitrate photolysis, and SOA formation from aromatic oxidations. They found that Chinese contribution accounts for almost 68% of $PM_{2.5}$ in surface air in South Korea during the extreme pollution period of the campaign, whereas an enhanced contribution from domestic sources (57%) occurs for the blocking period, characterized by a high pressure ridge to the north of an area of lower pressure in eastern China. A Rex Block (a high north of a low) limits horizontal transport leading to stagnant conditions (Peterson et al., 2019). Bae et al. (2020) conducted a long-term modeling study for years 2010–2017 using the CMAQ modeling system to estimate contributions of Chinese emissions to $PM_{2.5}$ in different provinces in Korea using two different horizontal resolutions. They estimated that average contributions from Chinese emissions for the 8-year period were 58% and 56% using the 27- and 9-km resolution, respectively.

Most of the air quality modeling conducted to estimate contributions of international sources to $PM_{2.5}$ in Korea has either been seasonal or has mostly focused on Chinese emissions. Also, most of the reported literature has focused on analysis in Seoul, Korea. To inform policies to reduce $PM_{2.5}$ concentrations in the country, it is important to fully understand the contribution of different emission sources (both domestic and international including countries other than China) under different meteorological conditions in different parts of the country. This information can then be used to develop effective control strategies to improve air quality. Here, we report a comprehensive analysis of contributions from China and Rest of the World to $PM_{2.5}$ concentrations in different parts of Korea for two recent years (2015 and 2016) using a global chemistry model with extensive model evaluation. We chose 2016 as the first year for modeling because it coincides with KORUS-AQ, thus providing a rich source of both ground-level and aloft data for model evaluation in Korea. (2015) was selected as the second year for modeling because it was meteorologically different from 2016 and was current enough that similar emissions inventories could be used for both baseline years after accounting for known changes. We report on the detailed model evaluation for both the years followed by modeled estimates of contributions from China, Rest of the World, and Korea to $PM_{2.5}$ concentrations in different parts of Korea.

2. Methods

We used the GEOS-Chem model (v12-01-01) to conduct full-year simulations of coupled gas-phase and aerosol chemistry (Bey et al., 2001; Park et al., 2006). This model uses assimilated meteorological data from Goddard Earth Observing System–Forward Processing (GEOS-FP) from the NASA Global Modeling and Assimilation Office (GMAO) (Lucchesi, 2018). The data are available at http://geoschemdata.computecanada.ca/ExtData/GEOS_0.25x0.3125_CH/GEOS_FP. The GEOS-FP meteorological data have a native horizontal resolution of $0.25^{\circ} \times 0.3125^{\circ}$ (~25 × 25 km^2) with 72 vertical pressure levels and 3-hr temporal frequency (1-h for surface variables and mixed layer depths). Validation of the meteorological data is included in the supplemental information (SI). To minimize the amount of memory required, we reduced the number of vertical levels to 47 by merging layers in the stratosphere. The GEOS-Chem model includes primary black carbon (BC), organic carbon (OC), secondary organic aerosol, and H_2SO_4–H–NO_3–NH_3 aerosol thermodynamics (Park et al., 2003; Heald et al., 2005). The model also includes soil dust in four size bins (Fairlie et al., 2007) and sea salt in two size bins (Jaeglé et al., 2011). A thermodynamic equilibrium model (ISORROPIA II) was applied to calculate gas/particle partitioning of SO_4^{2-}, NO_3^-, and NH_4^+ aerosols (Fountoukis and Nenes, 2007). The model simulation of OC and BC follows that of a previous study by Park et al. (2003). Dry and wet deposition have been described by Zhang et al. (2001) and Liu et al. (2001), respectively.

For each of the modeling years, we conducted a baseline GEOS-Chem simulation and two sensitivity simulations using GEOS-Chem and its nested framework for Asia (Harvard University, 2018). We first performed a global GEOS-Chem simulation with $2^{\circ} \times 2.5^{\circ}$ horizontal resolution to provide boundary conditions for the nested simulations. The nested simulation was conducted using the nested framework with $0.25^{\circ} \times 0.3125^{\circ}$ spatial resolution for the domain (see Fig. 1) with boundary conditions from the global run. The two sensitivity simulations were conducted by zeroing out anthropogenic emissions in China and anthropogenic emissions in both China and South Korea, respectively. The differences between the baseline and each sensitivity simulation yield the contributions from China and the rest of the world (ROW), respectively, to $PM_{2.5}$ concentrations in South Korea. The ROW contribution includes contribution from anthropogenic sources outside of Korea and China as well as from natural sources worldwide (including China and Korea).

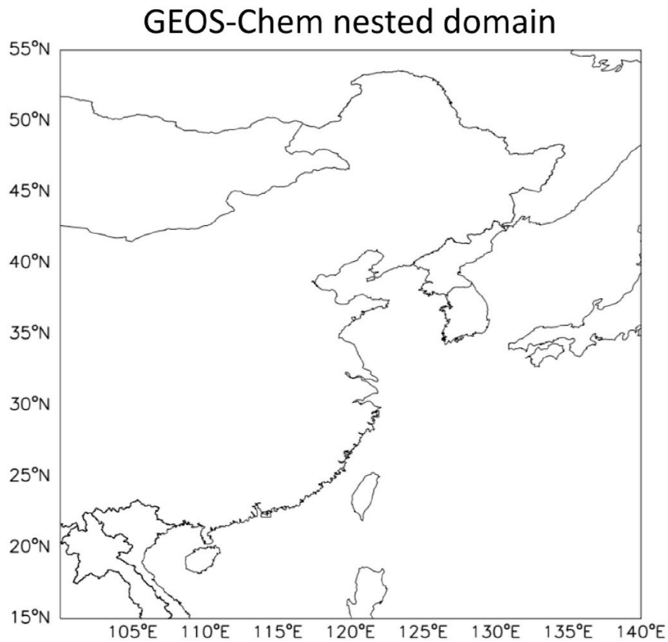

Fig. 1. The modeling domain for the nested GEOS-Chem simulations.

2.1. Emissions inputs for GEOS-Chem simulations

The Community Emissions Data System (CEDS; Hoesly et al., 2018) was used in GEOS-Chem as the global emissions inventory for all regions of the world excluding Korea, China, and several Northeast Asian countries (i.e., North Korea, Japan, Mongolia, and Asia Part of Russia). The CEDS system relies on existing energy consumption data sets as well as regional and country-specific inventories to produce trends over recent decades. The emissions developed with CEDS are available as gridded emission data at 0.1 ° × 0.1 ° horizontal resolution with monthly seasonality. For the regional emissions inventory, we used the NIER/KU-CREATE (National Institute of Environmental Research/Konkuk University – Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment, CREATE hereafter) emission inventory for Northeast Asia (except China). The CREATE inventory has been used to support various research and regulatory applications in Korea and East Asia (Woo et al., 2013; Woo et al., 2020); the latest base year is 2015.

For Korea, weused the Korean official emissions inventory for air pollutants is called *Clean Air Policy* Supporting *System* (CAPSS), which includes seven primary pollutants: CO, SO_2, NOx, VOC, NH_3, PM_{10}, and $PM_{2.5}$. The CAPSS is a comprehensive emissions inventory that has been used to support multiple local and regional air quality studies (for example, Kim et al., 2017, Bae et al., 2019, Bae et al., 2020). Table 1 shows the emissions of seven primary pollutants by different sources for 2015.

China is the most critical country for understanding transboundary influence on Korea because of its large emissions and its location directly upwind of Korea. The Multi-Resolution Emission Inventory for China (MEIC) compiles regional and sectoral emissions for China (MEIC, 2018). The latest year available is for 2017, which is recent enough to represent aggressive implementation of control policies in China. The MEIC was integrated into the Asia mosaic inventory (MIX) and the HTAP global emission inventory, both of which have been widely used by the air quality modeling community. Li et al. (2017) developed the MIX inventory to support the Model Inter-Comparison Study for Asia (MICS-Asia) and the Task Force on Hemispheric Transport of Air Pollution with inclusion of MEIC, CAPSS, and REAS. National total emissions for China by pollutant for Year 2016 (Table 2) are CO 141.9 Tg/yr, NOx 22.5 Tg/yr, SO_2 13.4 Tg/yr, $PM_{2.5}$ 8.1 Tg/yr, VOCs 28.4 Tg/yr, and NH_3 10.3 Tg/yr.

Ratios of Year (2015)–2017 changes in Fig. 2 show decreases of 11% and 7% for CO and NOx, respectively. $PM_{2.5}$ and SO_2 show decreases of 17%, and 38%, respectively, whereas almost no change is found for NMVOCs and NH_3. A large decrease of SO_2 emissions was found in the power and industrial sectors, which represents strong penetration of emission reduction policies and technologies. Based on these rapid changes from year to year, we applied the same inventory for China for 2015 and 2016.

Other East Asian countries—such as North Korea, Mongolia, and Russia—also contribute to transboundary air quality impacts in South Korea even though their emissions are relatively lower. We used CREATE version 3.0 emissions inventory for other North East Asia countries as well. The CREATE inventory compiles regional and sectoral emissions for Korea. The latest year available is 2015, which is recent enough to represent regional emissions estimates in Northeast Asian countries for 2016 as well. The 2015 emissions for North Korea, Japan, Mongolia, and Asian regions of Russia were used from this inventory for both the 2015 and 2016 simulations.

2.2. Observations used for model evaluation

Availability of ambient measurements in Korea is critical to support an air quality model performance evaluation for $PM_{2.5}$. These include surface measurements of $PM_{2.5}$ mass, $PM_{2.5}$ species, and $PM_{2.5}$ precursors (for example, NOx and SO_2). The data sources include the Air Quality Monitoring Station (AQMS) network (http://www.airkorea.or.kr) operated by NIER. The network measures real-time air pollutant

Table 2
Year 2016 emission estimate in MEIC emissions inventory for China.

Units (Tg/yr)	CO	NOx	SO_2	$PM_{2.5}$	VOC	NH_3
Power	4.6	4.6	2.7	0.6	0.1	0.0
Industrial	50.8	9.3	7.7	3.7	9.3	0.3
Residential	60.4	0.9	2.7	3.3	3.9	0.3
Transport	26.2	7.7	0.3	0.5	5.0	0.0
Solvent	0.0	0.0	0.0	0.0	10.1	0.0
Agriculture	0.0	0.0	0.0	0.0	0.0	9.6
Total	141.9	22.5	13.4	8.1	28.4	10.3

Table 1
Year 2015 emission estimates in the CAPSS emissions inventory.

Unit: Gg/year	CO	NOx	SOx	PM_{10}	$PM_{2.5}$	NMVOCs	NH_3
Power	55	151	91	4	4	7	1
Industrial	43	229	190	78	41	186	40
Residential	72	83	29	2	1	3	1
On-Road Mobile	246	370	0	10	9	46	10
Non-Road Mobile	136	304	39	15	14	40	0
Solvent	0	0	0	0	0	555	0
Agriculture	0	0	0	0	0	0	231
Other	241	21	2	125	30	173	13
Total	**793**	**1158**	**352**	**233**	**99**	**1011**	**297**

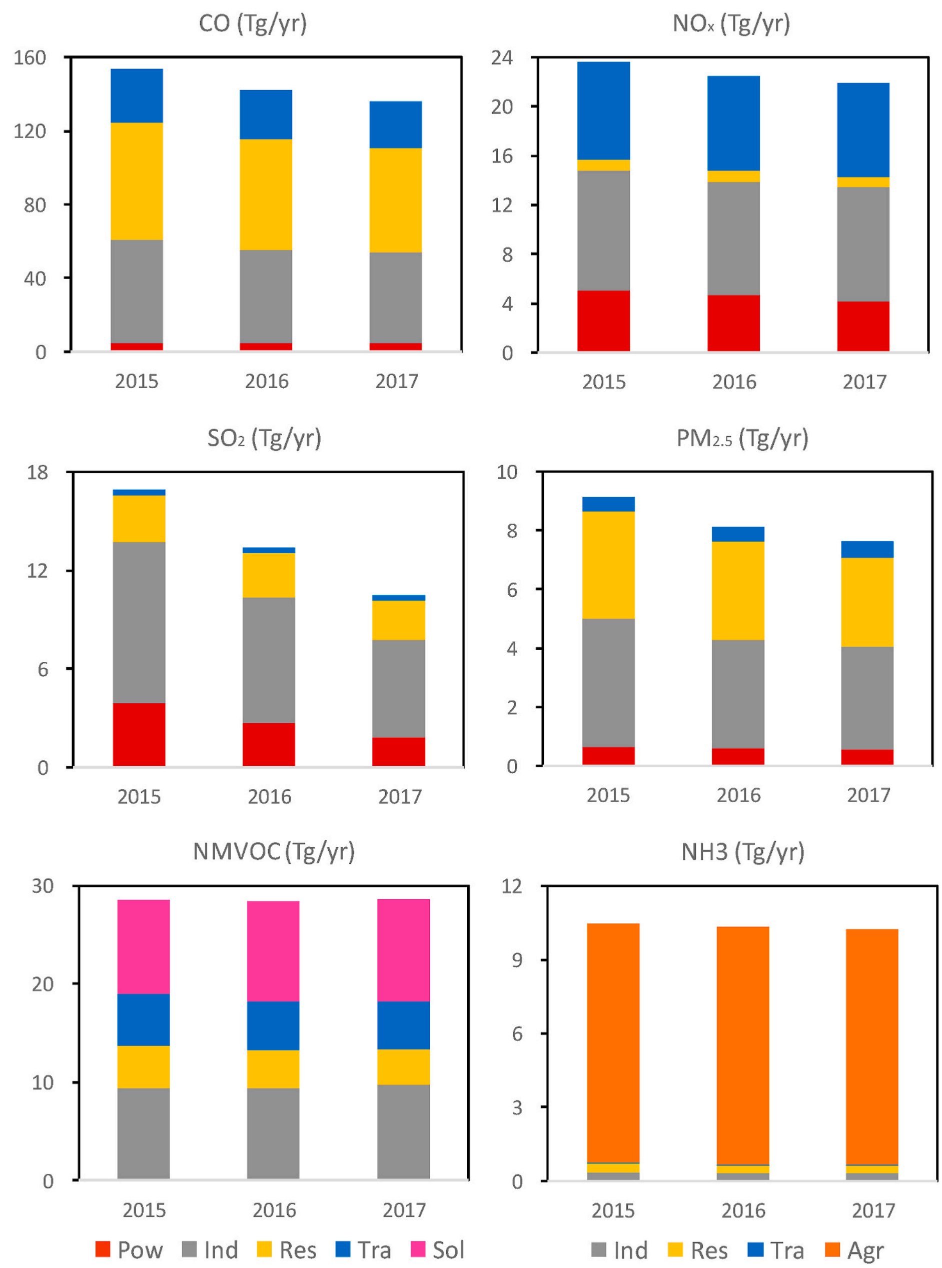

Fig. 2. 2015–2017 emissions trends in China: Pow, power; Ind, industry; Res, residential; Tra, transport; Sol, solvent; Agr, agriculture.

concentrations and provides hourly concentrations for CO, NO_2, O_3, $PM_{2.5}$, PM_{10}, and SO_2, which are available to the public. NIER also operates 6 p.m. supersites in Korea that provide continuous measurement data for speciated PM components (Korea Ministry of Environment, 2018). We obtained all the data summarized in Table 3, which was used for the model performance evaluation for $PM_{2.5}$. In addition, we used aerosol optical depth (AOD) measurements from satellites to support model performance evaluation over East Asia and rest of the globe – these results are shown in the SI.

Table 3
Summary of air quality data used for model evaluation.

Data Species	Sources	Spatial Resolution	Temporal Resolution	Period of Data Availability	Measurement Location
O_3, SO_2, NO_2, CO, PM_{10}	Air Korea	More than 250 sites	Hourly	2014–2017	Korea
$PM_{2.5}$	Air Korea	More than 250 sites	Hourly	2015–2017	Korea
MODIS AOD	NASA	1° x 1°	Monthly	2014–2017	Globe
GOCI AOD	Yonsei University	6 × 6 km^2	Hourly	2014–2017	East Asia
AERONET AOD	NASA	More than 30 sites	Daily	2014–2017	East Asia
Speciated PM components	NIER	6 sites	Daily	2015–2016	Korea

3. Results

3.1. Evaluation of the 2016 GEOS-Chem simulation

Fig. 3 shows comparisons between simulated monthly mean $PM_{2.5}$ concentrations and observations from the Air Korea network and six supersites in South Korea for 2016. We find good agreement between the model and both observation data sets, with correlation coefficients (0.62 and 0.81) and regression slopes of ~1.3. In particular, the model successfully reproduces the observed $PM_{2.5}$ concentrations at Baengnyeong Island (black circles on the right panel in Fig. 3), which is situated in the Yellow Sea and therefore an ideal site to monitor transboundary pollution influences from China. The model appears to successfully capture transboundary transport of aerosols from upwind regions including China.

We also evaluated simulated chemical components comprising $PM_{2.5}$, including SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC using the observations from six supersites in the peninsula. Fig. 4 shows scatter plot comparisons of the simulated vs. observed monthly mean SO_4^{2-}, NO_3^-, and NH_4^+ concentrations at six supersites. The model generally underestimates SO_4^{2-} concentrations but overestimates NO_3^- concentrations in surface air. The low bias of SO_4^{2-} may indicate low SO_2 emissions in the model. Formation of the two inorganic ions is tightly related through thermodynamic equilibrium so that too much NH_3 available from insufficient SO_4^{2-} neutralization could produce too much NH_4NO_3 in the model. This is evident in Fig. 4.

Fig. 5 also shows scatter plot comparisons of the simulated vs. observed aircraft every 1 min SO_4^{2-}, NO_3^-, and NH_4^+ concentrations below 2 km on board DC-8 during the KORUS-AQ campaign. The simulated values are sampled along the flight track every 1 min for comparisons so as to capture spatial variation in the observations. In this comparison, we also find a similar underestimate in SO_4^{2-} concentrations in the low troposphere but high bias in NO_3^- concentrations.

Finally, we examined the carbonaceous components of $PM_{2.5}$ in the model. Fig. 6 shows scatter plot comparisons of the simulated vs. observed monthly mean OC and BC concentrations at six supersites. BC does not show too much discrepancy between the model and the observations, but OC is too high in the model—especially in the surface air relative to the observations. This is likely caused by an increase of primary OC emissions in the KU-CREATE emission inventory. When we compare the model with the aircraft observations from the DC-8 during the KORUS-AQ campaign (see Fig. 7), we could not find a significant high bias in the model below 2 km altitude; the model even shows a slight low bias, which has been an issue in the past (Heald et al., 2005).

While there are no statistical benchmarks proposed in the literature for evaluation of global chemistry models like GEOS-Chem, such benhmarks have been proposed for regional chemical transport models (CTMs) or regional applications that include GEOS-Chem (e.g., Emery et al., 2017; Huang et al., 2021). Although the benchmarks proposed for regional applications may not be applicable for GEOS-Chem at coarser resolution, we provide statistical metrics in the SI and compare against benchmarks recommended by Huang et al. (2021) that are proposed for China (the definitions of different metrics can be found in that paper).

Table S-1 shows statistical model performance evaluation using daily data, as speciation data are only available at daily interval. As one can see, the baseline model simulation captures the observations relatively well and most benchmarks satisfy the recommended criteria for $PM_{2.5}$ and its chemical components except for sulfate, whose correlation coefficients are slightly lower than the recommended criteria. As noted earlier, model overestimates nitrate, ammonium, and OC concentrations.

3.2. Evaluation of 2015 GEOS-Chem simulation

As with the 2016 simulation, we compared simulated monthly mean $PM_{2.5}$ concentrations with observations from the Air Korea network and six supersites in South Korea for 2015 (see Fig. 8). We find good agreement between the model and both observation data sets, with correlation coefficients (0.7). Just like for 2016, the model successfully reproduces the observed $PM_{2.5}$ concentrations at Baengnyeong Island (black circles on the left panel in Fig. 8). The model again appears to successfully capture transboundary transport of aerosols from upwind regions including China.

We also evaluated chemical components comprising $PM_{2.5}$, including SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC using the observations from six supersites in the peninsula. Fig. 9 shows scatter plot comparisons of the simulated vs. observed monthly mean SO_4^{2-}, NO_3^-, and NH_4^+ concentrations at six supersites. The model generally underestimates SO_4^{2-} concentrations but overestimates NO_3^- concentrations in surface air. Once again, the low bias of SO_4^{2-} may indicate low SO_2 emissions in the model.

Finally, we examined the carbonaceous components of $PM_{2.5}$ in the model. Fig. 10 shows scatter plot comparisons of the simulated vs. observed monthly mean OC and BC concentrations at six supersites. Like the 2016 simulation, BC shows a high bias between the model and the observations particularly in Seoul and OC is too high in the model—especially in the surface air relative to the observations. This again suggests that an increase of primary OC emissions in the KU-CREATE emission inventory may have contributed to this positive bias.

Similar to 2016, we also conducted statistical model performance using daily data, as shown in Table S-2. The performance is similar to 2016 in that the baseline model simulation captures the observations relatively well and most benchmarks satisfy the recommended criteria for $PM_{2.5}$ and its chemical components.

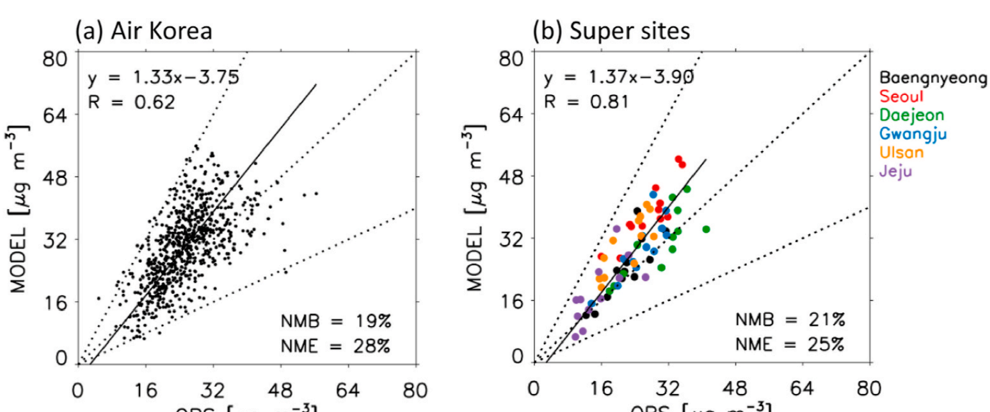

Fig. 3. Scatter plot comparisons of monthly mean $PM_{2.5}$ concentrations between the GEOS-Chem baseline simulation versus observations from the (a) Air Korea network and (b) six supersites in South Korea for 2016. The normalized mean bias (NMB) and normalized mean error (NME) are shown inset.

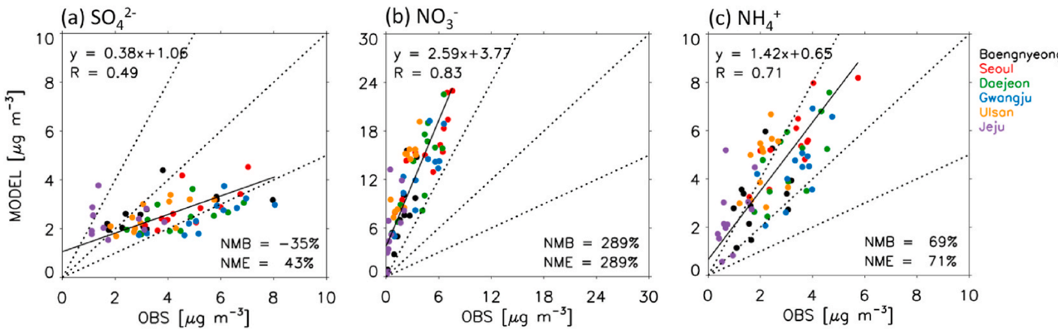

Fig. 4. Scatter plot comparisons of monthly mean (a) SO_4^{2-}, (b) NO_3^-, and (c) NH_4^+ concentrations between the model simulation results versus observations from the six supersites in South Korea for 2016. The normalized mean bias (NMB) and normalized mean error (NME) are shown inset.

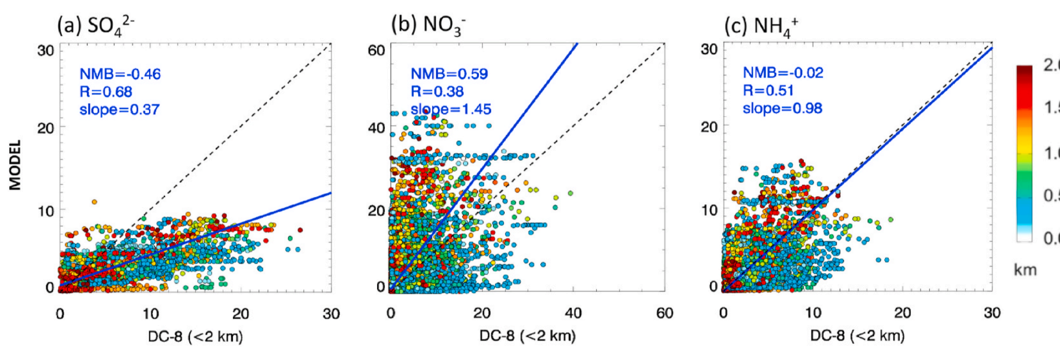

Fig. 5. Scatter plot comparisons of (a) SO_4^{2-}, (b) NO_3^-, and (c) NH_4^+ concentrations (μg m^{-3}) every 1 min between the model simulation versus aircraft observations below 2 km from DC-8 during the KORUS-AQ campaign. The colors of the circles represent the observed altitude from the DC-8. The normalized mean bias (NMB), correlation coefficient (R), and slope are shown inset. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

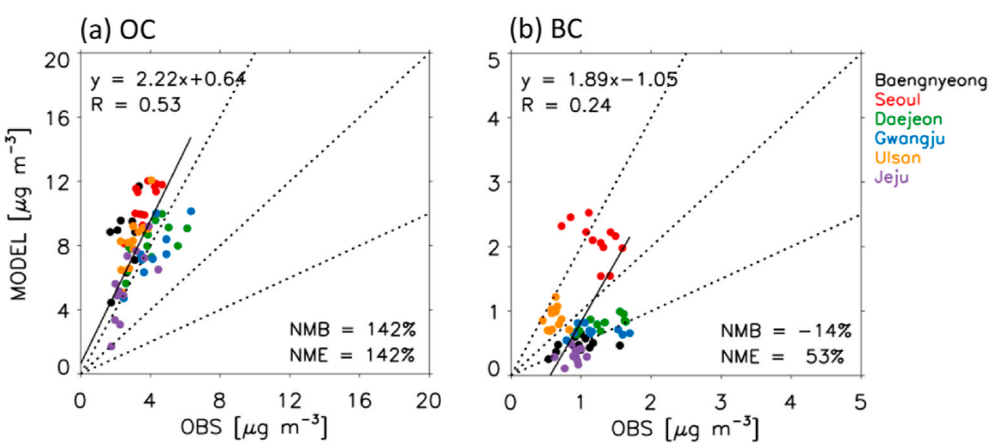

Fig. 6. Scatter plot comparisons of monthly mean (a) OC and (b) BC concentrations between the model simulation versus observations from the six supersites in South Korea for 2016. The normalized mean bias (NMB) and normalized mean error (NME) are shown inset.

3.3. Contribution from China, Korea, and other international sources

As mentioned previously, GEOS-Chem sensitivity simulations were used to quantify domestic and transboundary contributions to $PM_{2.5}$ concentrations in South Korea. We conducted the baseline simulation and two sensitivity simulations with no anthropogenic emissions in China and then no anthropogenic emissions in both China and South Korea. A contribution of Chinese anthropogenic emissions to $PM_{2.5}$ concentrations was calculated by subtracting the first sensitivity simulation from the baseline simulation. Similarly, we computed a contribution of the rest of world emissions to $PM_{2.5}$ concentrations by subtracting the second sensitivity simulation from the baseline simulation. The rest of the world includes all countries other than China and South Korea and therefore includes North Korea as well as natural emissions from the whole domain. The remaining contribution was deemed to be from domestic anthropogenic sources.

3.3.1. Contributions for 2016

First, we look at the monthly spatial plots from the nested GEOS-Chem results from the baseline simulation as shown in Fig. 11. Simulated $PM_{2.5}$ concentrations in surface air show high values in China and its downwind regions including the Yellow Sea, mostly in winter and spring. During the summer, East Asian summer monsoons bring relatively clean air from the northwestern Pacific and result in much lower

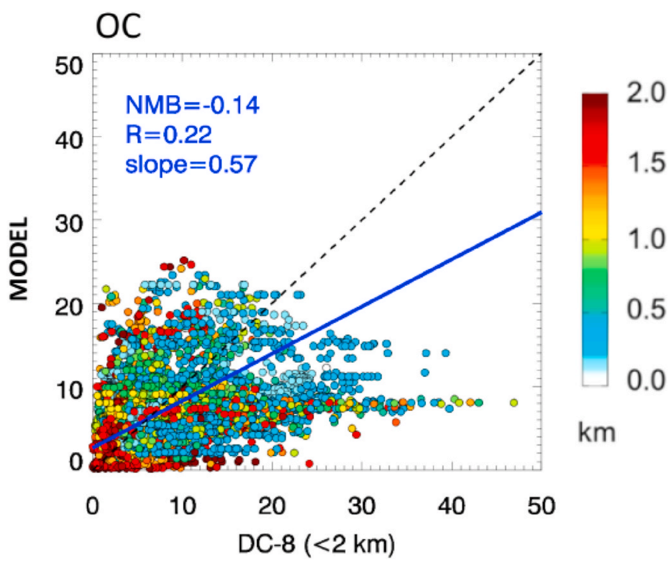

Fig. 7. Scatter plot comparisons of OC concentrations every 1 min between the model simulation versus aircraft observations below 2 km from DC-8 during the KORUS-AQ. campaign. The colors of the circles represent the observed altitude from the DC-8. The normalized mean bias (NMB), correlation coefficient (R), and slope are shown inset. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

$PM_{2.5}$ concentrations throughout Asia. The seasonal variation of $PM_{2.5}$ in Korea is mostly affected by the East Asian monsoons, which are a key factor for determining synoptic meteorological patterns in this region. Fig. 11 indicates that one can expect higher contribution from China to $PM_{2.5}$ concentrations in South Korea during the winter and spring months.

Fig. 12 shows the five cities that we chose to show contributions from different source regions – these cities were chosen because they show relatively high observed $PM_{2.5}$ concentrations and are spread across the country. Figs. 13 and 14 show simulated monthly mean contributions to $PM_{2.5}$ concentrations in the South Korea domain (126-129.5E, 34.5–38N) as well as different cities for 2016. Plots for Ulsan and Gangreong are shown in the SI. We find that at those cities and on average over South Korea the total $PM_{2.5}$ values are generally higher in winter and spring than those in summer and are the highest in March. Contributions from China are the highest in winter and spring months. There is a large variability in contributions from China between the winter and summer months, whereas contributions from domestic sources don't vary as much from season to season. For example, in Seoul domestic contributions are quite high in winter (December), in spring (March, April), in summer (June, July), and in fall (September). Gangreong (shown in the SI) is a bit different from other cities, as the domestic contributions are considerably smaller than the Chinese contributions in most months of the year. Taean is another such city with relatively higher contributions from China. Taean is a coastal city on the western coast with direct influence from Chinese emissions, so it is not surprising that it has relatively higher contributions from China. Gangreong is a city on the eastern coast with lower domestic emissions, thus shows relatively higher contributions from China.

3.3.2. *Contributions for 2015*

The nested GEOS-Chem results from the baseline simulation for 2015 are similar to those for 2016 and the spatial patterns of the monthly PM2.5 concentrations are shown in the SI. Figs. 15 and 16 show simulated monthly mean contributions to $PM_{2.5}$ concentrations in South

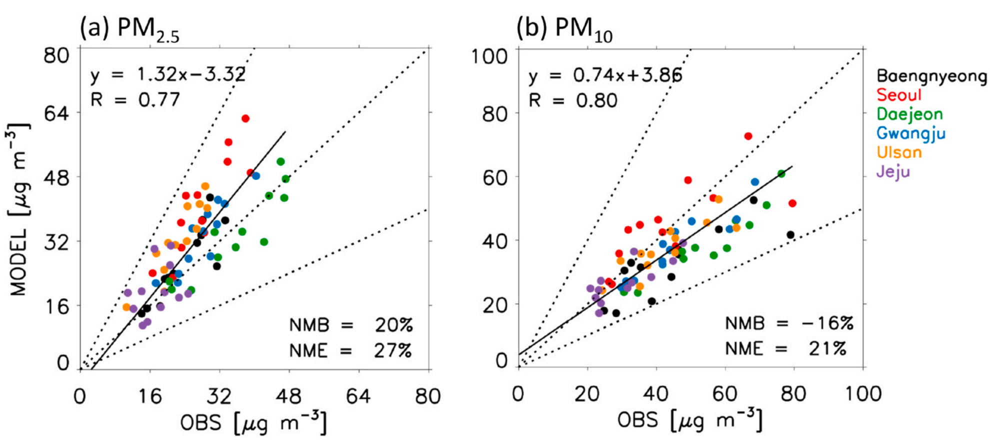

Fig. 8. Scatter plot comparisons of monthly mean (a) $PM_{2.5}$ and (b) PM_{10} concentrations between the model simulation versus observations from six supersites in South Korea for 2015. The normalized mean bias (NMB) and normalized mean error (NME) are shown inset.

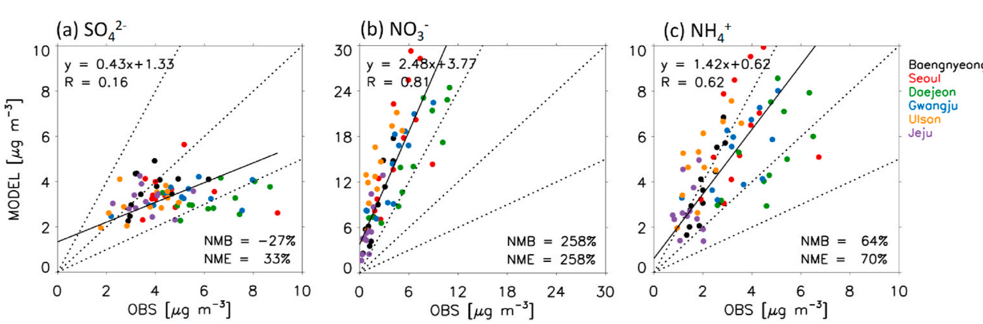

Fig. 9. Scatter plot comparisons of monthly mean (a) SO_4^{2-}, (b) NO_3^-, and (c) NH_4^+ concentrations between the model simulation versus observations from six supersites in South Korea for 2015. The normalized mean bias (NMB) and normalized mean error (NME) are shown inset.

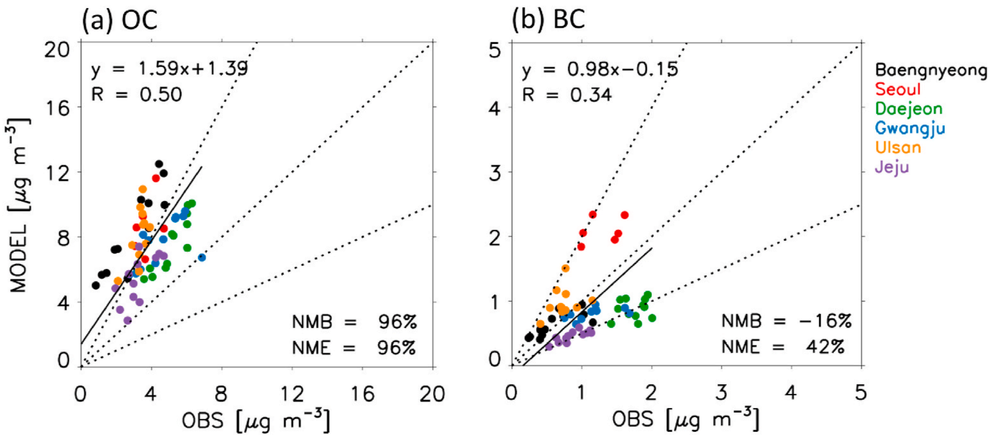

Fig. 10. Scatter plot comparisons of monthly mean (a) OC and (b) BC concentrations between the model simulation versus observations from the six supersites in South Korea for 2015. The normalized mean bias (NMB), and normalized mean error (NME) are shown inset.

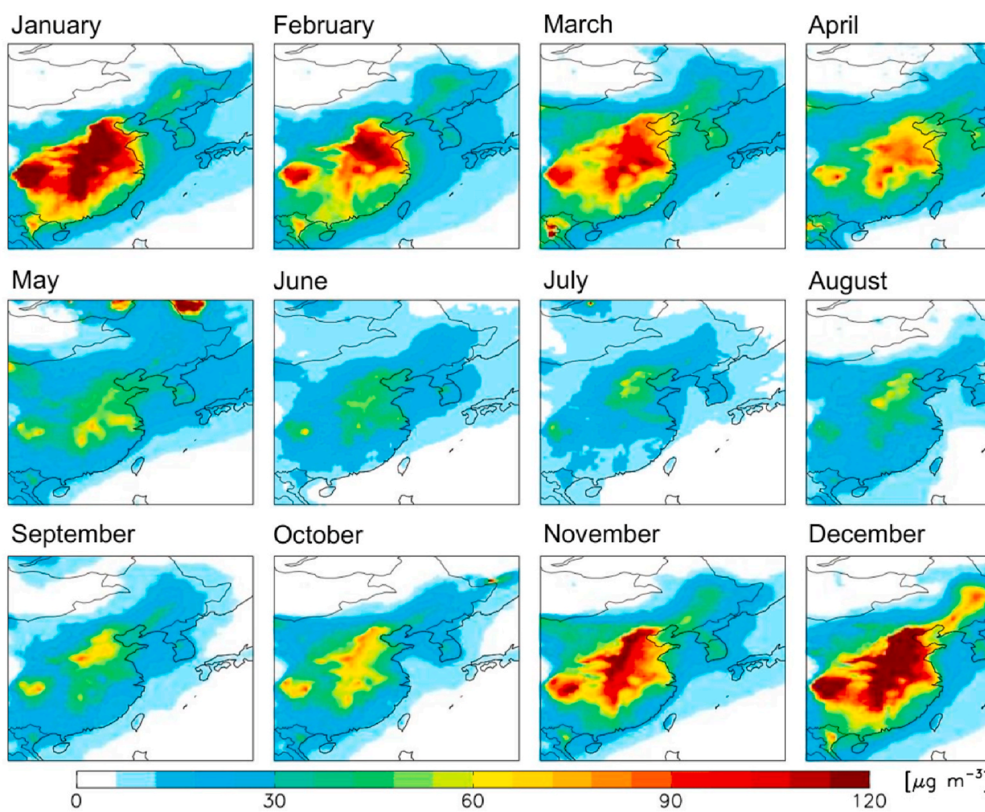

Fig. 11. Monthly mean surface $PM_{2.5}$ concentration from the model simulation for 2016.

Korea (126-129.5E, 34.5–38N) and three cities (Seoul, Taean, and Samcheonpo) for 2015 – Ulsan and Gangreong results are shown in the SI. Values are generally higher in winter and spring than those in summer and are the highest in March, although some differences exist depending on the eastern versus western part of the peninsula.

Figs. 15 and 16 also show that the Chinese contributions are generally dominant in cold seasons, whereas they are relatively less important in warm seasons. The contributions from the rest of the world are also shown. Compared to the Chinese contribution, the ROW contributions are relatively minor except for summer when they appear to be important mostly in the southern sea (not shown). The rest of the patterns in different cities are similar to what was seen for 2016.

3.3.3. Differences between contributions for 2015 and 2016

Tables 4 and 5 summarize the transboundary transport contributions from China to $PM_{2.5}$ concentrations in South Korea and at five representative cities in 2015 and 2016, respectively, focusing on the months of March and April when the synoptic conditions are favorable for the long-range transport of pollutants. In March, compared to 2015, values of $PM_{2.5}$ concentrations in South Korea are lower in 2016. The lower values in 2016 are in part owing to the reduction of anthropogenic emissions in China. In terms of Chinese contributions to $PM_{2.5}$ in South Korea and at five cities, we can also see a decrease from 2015 to 2016 by 3–5% except for Gangreong. The transboundary transport is largely determined by the synoptic meteorological conditions. The transboundary transport contributions from China to annual mean $PM_{2.5}$ concentrations in South Korea and at five representative cities are also

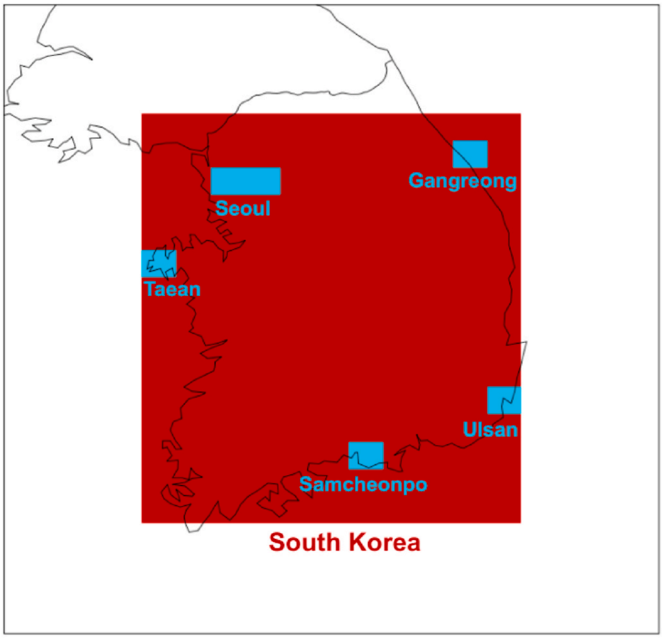

Fig. 12. South Korea domain and five selected city locations (Seoul, Taean, Samcheonpo, Ulsan, and Gangreong).

summarized in Table 6. Although a decrease in annual mean $PM_{2.5}$ concentrations from 2015 to 2016 is simulated, the Chinese contributions for both years are largely consistent nationwide with different degrees for individual cities.

Fig. 17 shows spatial distributions of $PM_{2.5}$ concentrations and wind vectors at 850 hPa in GEOS-Chem simulations for March 2015 and 2016, respectively. Their differences between two years are also shown in Fig. 17. As we can see, March 2016 winds at 850 hPa are easterly in the peninsula and Yellow Sea, indicating less efficient long-range transport of air pollutants from China in March 2016.

However, despite the anthropogenic emission reduction in China in 2016 relative to 2015, the transboundary transport contributions to $PM_{2.5}$ in South Korea have increased from 2015 to 2016 in the month of April. We find that this is mostly caused by favorable synoptic conditions for long-range transport in April 2016, which is illustrated in Fig. 18. We can see the prevailing westerly winds in April 2016, which brings pollutants from China into the Korean peninsula. The variation of the transboundary transport influences from March to April indicates the important role of synoptic meteorological conditions in determining transboundary transport of pollutants from China to Korea.

4. Discussion

We conducted simulations for 2015 and 2016 using the GEOS-Chem 3-D global chemical transport model and its nested framework to estimate contributions to PM2.5 in Korea from China, Korea and rest of the world. The simulations for each year included one baseline and two sensitivity simulations with no anthropogenic emissions in China and in the Korean peninsula. Our evaluation of the model against observations showed that the baseline results were adequate to be used for contribution analysis for China and the rest of the world.

We estimate that the Chinese contributions to $PM_{2.5}$ concentrations in South Korea for 2015 and 2016 were dominant in cold seasons, up to ~60% in January and February on a monthly mean basis, whereas they were less important in warm seasons reaching a minimum at about 20% in August. Compared to the Chinese contribution, the rest of the world contributions (which also include contributions from natural emissions all over the world) were minor except for summer in the South Sea. However, the daily contributions changed widely and could sometimes be very high in summer. On average, the rest of the world contributions can be as high as 30% of monthly average in July (2015) and August (2016) when the total concentrations are usually the lowest. We found that variations in the transboundary transport contributions were strongly influenced by meteorology but also declined in response to emission reductions, as expected. For example, the Chinese contribution to $PM_{2.5}$ concentrations in Korea in April 2016 was higher than that of 2015 despite the decrease in Chinese anthropogenic emissions because of the favorable synoptic conditions for long-range transport in April 2016. On an absolute basis, Chinese contributions were the highest in

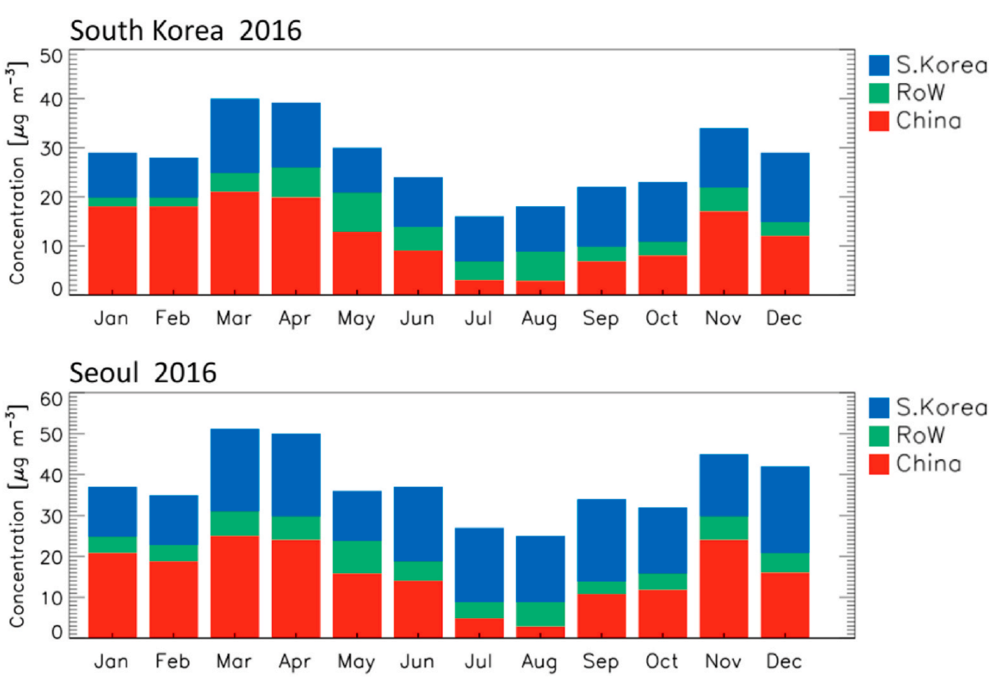

Fig. 13. Simulated contributions of emissions from China (red), the rest of the world (green), and South Korea (blue) to monthly mean $PM_{2.5}$ concentrations in 2016 in South Korea (upper) and Seoul (lower). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

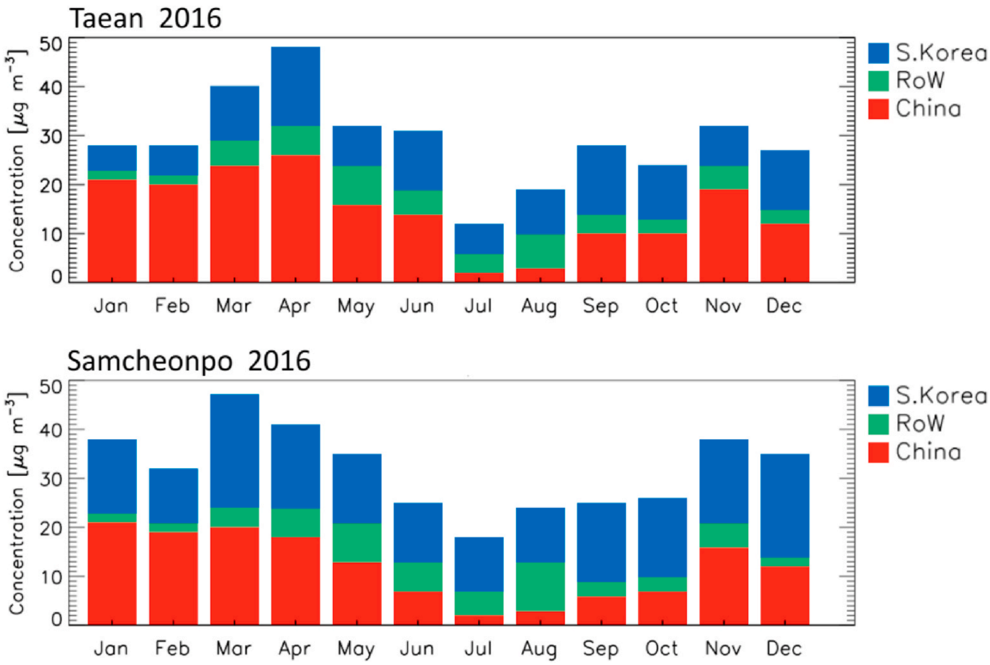

Fig. 14. Same as Fig. 13 except for taean (upper) and samcheonpo (lower).

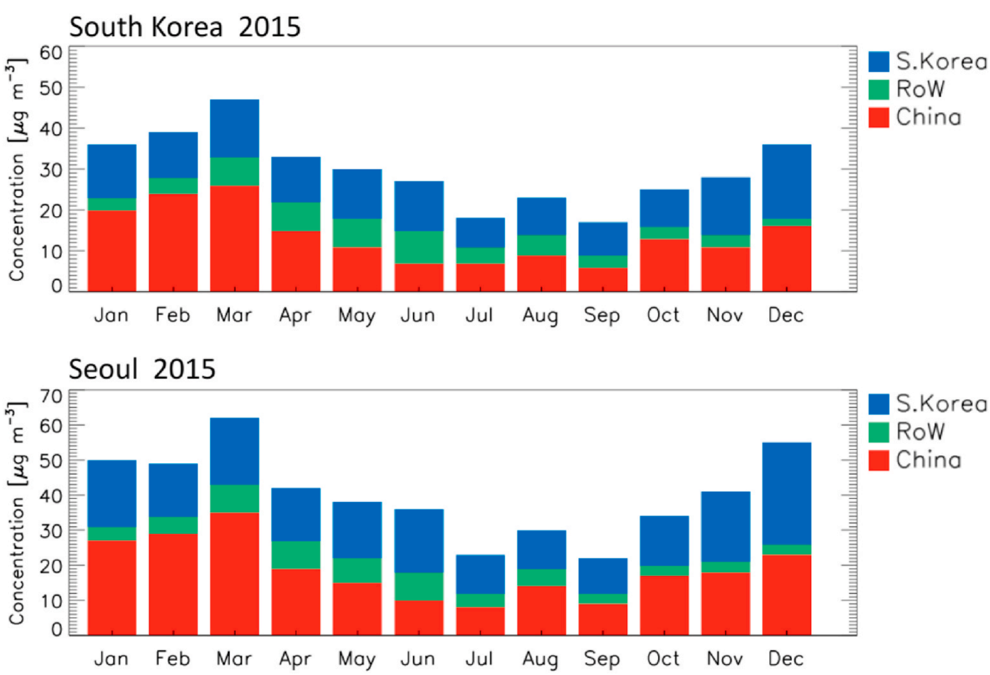

Fig. 15. Simulated contributions of emissions from China (red), the rest of the world (green), and South Korea (blue) to monthly mean $PM_{2.5}$ concentrations in 2015 in South Korea (upper) and Seoul (lower). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

March and April in 2016, whereas for 2015 they were the highest in March but decrease in April. The 2016 contribution from China was generally 3–5% lower than in 2015 because of emissions reductions except for the anomaly in April 2016 when the contributions were higher than in April 2015. Since meteorology can play such an important role in determining the influence of Chinese emissions to $PM_{2.5}$ in Korea, a more thorough evaluation of the impact of different meteorological conditions may be needed when planning policy measures to control $PM_{2.5}$.

When examining individual urban locations, the results are similar to the overall results in the whole Korean domain with some important differences. On an annual basis, Taean and Gangreong had the highest percentage contribution from Chinese emissions ranging between 51 and 55 percent between the two cities and the two modeling years. As mentioned earlier Taean is a coastal city on the western coast with direct influence from Chinese emissions and Gangreong is a coastal city on the eastern coast with relatively lower domestic emissions. Samcheonpo and Ulsan were at the other extreme with annual average contribution from Chinese emissions between 38 and 39 percent. Seoul was somewhere in the middle with the annual average contribution from Chinese emissions at 43 and 47 percent in 2016 and 2015, respectively. Gangreong also had the highest monthly average contribution from Chinese emissions at

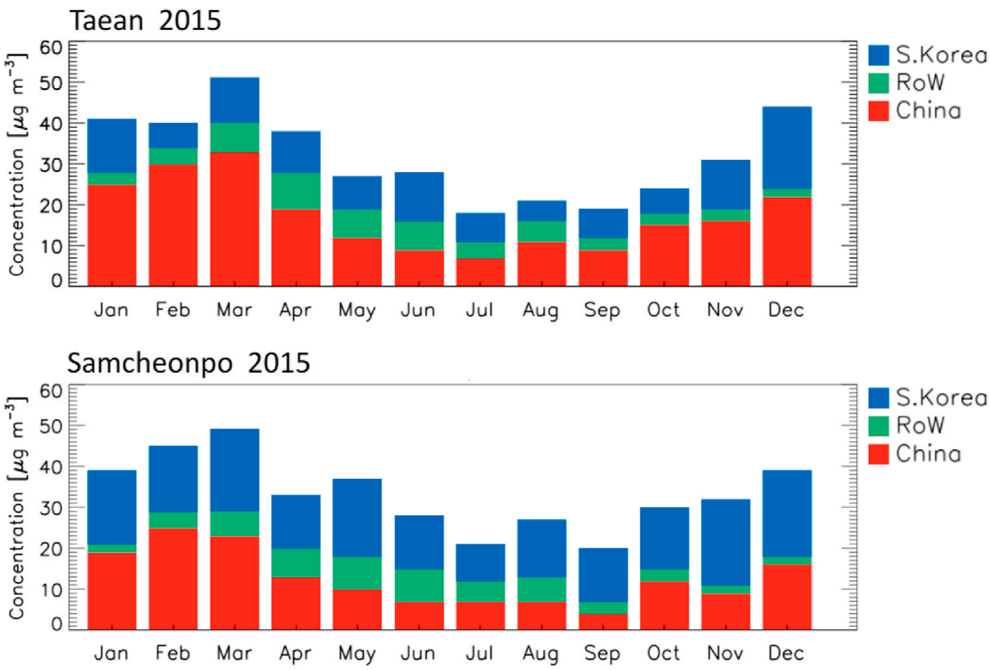

Fig. 16. Same as Fig. 15 except for taean (upper) and samcheonpo (lower).

Table 4
Average contribution change from 2015 to 2016 in month of March.

Location	2015 p.m.$_{2.5}$ [μg m^{-3}]	2016 p.m.$_{2.5}$ [μg m^{-3}]	China Contribution 2015		China Contribution 2016	
			[μg m^{-3}]	[%]	[μg m^{-3}]	[%]
South Korea	47.5	40.7	26.9	57	21.2	52
Seoul	62.2	51.3	35.2	57	25.2	49
Taean	51.3	40.0	33.0	64	24.1	60
Samcheonpo	49.2	47.4	23.0	47	20.3	43
Ulsan	45.6	42.8	21.5	47	19.0	44
Gangreong	41.2	30.4	26.5	64	20.6	68

Table 5
Average contribution change from 2015 to 2016 in month of April.

Location	2015 p.m.$_{2.5}$ [μg m^{-3}]	2016 p.m.$_{2.5}$ [μg m^{-3}]	China Contribution 2015		China Contribution 2016	
			[μg m^{-3}]	[%]	[μg m^{-3}]	[%]
South Korea	33.2	39.8	15.3	46	20.8	52
Seoul	42.7	50.2	19.8	46	24.5	49
Taean	38.6	48.2	19.6	51	26.8	56
Samcheonpo	33.6	41.2	13.6	41	18.9	46
Ulsan	31.4	37.4	13.3	42	16.9	45
Gangreong	26.1	26.6	14.0	54	15.7	59

68 percent in March 2016.

From a policy point of view, it is instructive to examine transboundary contributions on days with the highest $PM_{2.5}$ concentrations, as Korea has a daily $PM_{2.5}$ standard. For the top-10 modeled days of highest average daily $PM_{2.5}$ concentrations in Korea in 2015, all of which occur either in winter or spring, the average contribution from Chinese anthropogenic emissions was 71% and 8% was from rest of the world. The corresponding numbers for 2016 were 62% and 9% from Chinese emissions and rest of the world, respectively.

When compared to the previous studies, our results for Chinese contributions are similar, although we show that the Chinese contributions reduced from 2015 to 2016 because of emissions reductions in China. Given that Chinese emissions further reduced in 2017 (Fig. 2) and are expected to reduce further given their emissions reductions plan, the relative contribution of Chinese emissions to $PM_{2.5}$ in Korea may have changed depending on how emissions in Korea and other countries have behaved in the same time period. In addition, we showed that meteorology can play an important role in transboundary pollution. Therefore, it is important to conduct modeling for more recent years to obtain current estimates.

Table 6
Annual average contribution change from 2015 to 2016.

Location	2015 p.m.$_{2.5}$ [μg m^{-3}]	2016 p.m.$_{2.5}$ [μg m^{-3}]	China Contribution 2015		China Contribution 2016	
			[μg m^{-3}]	[%]	[μg m^{-3}]	[%]
South Korea	30.3	28.3	14.2	47	12.9	46
Seoul	40.5	38.0	19.1	47	16.2	43
Taean	32.3	29.6	17.7	55	15.1	51
Samcheonpo	33.8	32.3	13.0	39	12.4	38
Ulsan	32.4	30.7	12.2	38	11.6	38
Gangreong	20.1	18.6	10.6	53	9.9	54

One caveat with the contribution analysis using brute force methods as used in our study is that there is no estimate of uncertainty that may be associated with the approach. Although we showed the model performance was similar to other modeling studies that have used similar approaches in the past, one way to increase confidence in our results is to examine model performance on days where meteorological conditions would minimize Chinese emission contribution to Korean monitoring locations. We found two days (July 21 and July 26) in 2015 when the meteorological conditions indicated no transport from China on previous few days. The Chinese contribution to PM2.5 in Korea was less than 1% for those days confirming what the meteorology indicated. The model performance for those two days is shown below in Table 7. The model performs quite well for those two days indicating confidence in the model in predicting $PM_{2.5}$ concentrations when most of the

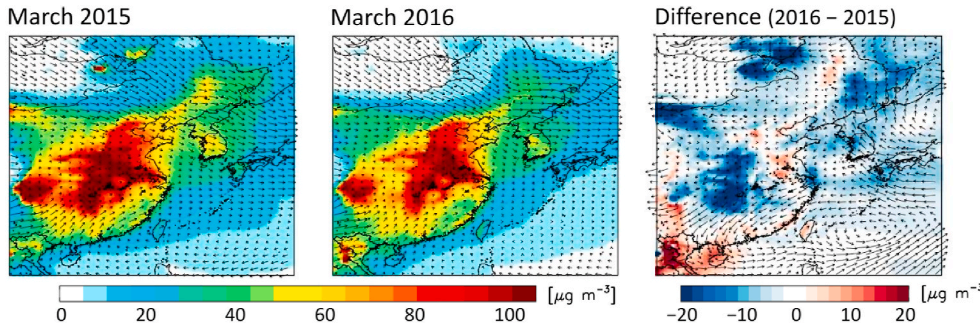

Fig. 17. Monthly surface $PM_{2.5}$ concentrations and 850 hPa wind filed from the nested GEOS-Chem model in East Asia for March 2015 and 2016 and difference between two months.

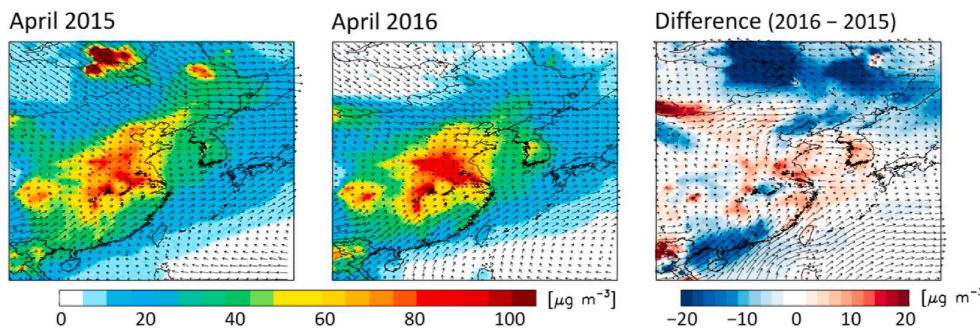

Fig. 18. Same as Fig. 17 except for April 2015 and 2016.

Table 7
Statistical model performance for days with the least contribution from China.

Metrics	Unit	$PM_{2.5}$[a]
R		0.58
IOA		0.74
NMB	%	3
NME	%	28
FB	%	1
FE	%	27
RMSE	$\mu g\ m^{-3}$	5.4
MB	$\mu g\ m^{-3}$	0.5
ME	$\mu g\ m^{-3}$	4

[a] The contribution from China is 1% (July 21, July 26, 2015).

contribution is from local sources.

CRediT authorship contribution statement

Naresh Kumar: Conceptualization, Methodology, Writing – original draft, Supervision, Project administration, Funding acquisition. **Rokjin J. Park:** Methodology, Formal analysis, Investigation, Resources, Writing – review & editing, Supervision. **Jaein I. Jeong:** Software, Validation, Formal analysis, Investigation, Visualization. **Jung-Hun Woo:** Methodology, Formal analysis, Investigation, Resources, Writing – review & editing, Supervision. **Younha Kim:** Software, Formal analysis, Investigation, Visualization. **Jeremiah Johnson:** Formal analysis, Investigation, Visualization. **Greg Yarwood:** Methodology, Resources, Writing – review & editing, Supervision. **Suji Kang:** Conceptualization, Resources, Project administration. **Sungnam Chun:** Conceptualization, Resources, Supervision, Project administration, Funding acquisition. **Eladio Knipping:** Conceptualization, Methodology, Writing – review & editing, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118542.

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