Recent Decreasing Trends in Surface PM_{2.5} over East Asia in the Winter-Spring Season: Different Responses to Emissions and Meteorology between Upwind and Downwind Regions

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ABSTRACT

This study developed and evaluated a WRF-Chem modeling system that reflects the effect of recent emission regulations on the PM_{2.5} above East Asia by utilizing an updated anthropogenic emission inventory for 2013–2018. This system accurately reproduced the monthly means, daily variations, and vertical profiles of PM_{2.5} during winter and spring over the Seoul Metropolitan Area (SMA) in South Korea and the North China Plain (NCP) and Yangtze River Delta (YRD) in China. Furthermore, it demonstrated that the decline in PM_{2.5} over the latter nation is attributable to control measures in China that have been in effect since 2013. The most polluted of the three target regions, the NCP, which is also upwind (in contrast to the downwind YRD and SMA), exhibited the largest decrease due to emission reduction. For example, the simulated mean PM_{2.5} concentration for February dropped by 39% over the NCP but by merely 17% over the YRD between 2013 and 2018. Additionally, the SMA displayed only minor changes in the concentration during winter and a weak decreasing trend during spring.

In addition to emission reduction, meteorology significantly modulated the level of PM_{2.5}; it produced larger interannual variations in the downwind regions than the upwind one, accounting for changes in concentration as high as 35% and 45% in the SMA during winter and spring, respectively, versus 11% and 12% in the NCP. Finally, the downwind regions also showed more complex behaviors for the secondary aerosols, which did not always follow the decreasing trends of their precursors.

Keywords: PM_{2.5}, WRF-Chem, Trends, Emissions

1 INTRODUCTION

Particulate matter (PM) pollution has been a serious health concern over East Asia and has been receiving increasing attention over the past decade. After experiencing unprecedented PM pollution levels with record-breaking long durations over China in 2012–2013 winter (Shao *et al.*, 2018) and over South Korea in 2013–2014 winter (Ryu and Min, 2020), public awareness about poor air quality has markedly increased. As a consequence, both countries took significant actions to reduce pollutant emissions. China issued China's Clean Air Action Plan in September 2013, which resulted in considerable reductions in PM_{2.5} concentrations (Zeng *et al.*, 2019). Recently, Zheng *et al.* (2018) estimated that primary PM_{2.5} emissions over China were reduced by 33% from 11.4 Tg in 2013 to 7.6 Tg in 2017. For South Korea, Particulate Matter Comprehensive Plan was enacted in 2017, and as a part of the plan an emergency PM emission reduction action has been effective since 2017.

Air quality modeling systems are often used to evaluate the effectiveness of emission control

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actions and ultimately to develop sustainable control strategies for the future (e.g., Gilliland *et al.*, 2008; Wang *et al.*, 2017). Among various components of air quality modeling, accurate and up-to-date emission estimates are a critical prerequisite for good model performance, especially in rapidly developing countries such as China or South Korea. There are two approaches to estimating anthropogenic emissions: bottom-up and top-down approaches, and both approaches have pros and cons. Bottom-up inventories are generally available at high horizontal resolutions (often higher than $0.5^{\circ} \times 0.5^{\circ}$) and provide inventories of major pollutant species (CO, NO_x, non-methane volatile organic compounds, (NMVOC), SO₂, and PM). Because it takes several years to collect relevant data (e.g., socioeconomic activities and control technologies) in bottom-up approaches, however, bottom-up inventories are not updated in a timely manner and often lag several years. On the other hand, top-down approaches utilizing inverse modeling techniques can provide timely emission estimates even over the regions where reliable and detailed emission information is not readily available (Elguindi *et al.*, 2020). However, due to the nature of methodology that uses observations to constrain atmospheric models, the uncertainties in top-down emission estimates are largely influenced by the uncertainties/qualities of observations (often satellite retrievals) and model's capabilities of predicting physical and chemical processes of pollutants (e.g., transport and chemical transformation). The intercomparison of various bottom-up inventories and top-down estimates by Elguindi *et al.* (2020) showed significant differences among emission estimates, and they found that top-down estimates often exhibit a larger range of uncertainties than bottom-up inventories.

In the present study, we propose corrections/adjustments that can be applied to anthropogenic emission inventories for recent years (2013–2018) over East Asia to reproduce recent decreasing trends in PM_{2.5} concentrations using a WRF-Chem modeling system. East Asia, especially China, has been experiencing rapid economic growth and also emission regulations, and thus realistic up-to-date emission estimates are required and should be used in air quality modeling systems when examining interannual variabilities in pollutant concentrations and projecting onto future scenarios. Because top-down estimates are not available for all major species and due to their coarse horizontal resolutions (e.g., $1.1^{\circ} \times 1.1^{\circ}$ of TCR-2; Miyazaki *et al.*, 2020), we choose a bottom-up-type inventory, the Regional Emission inventory in ASia (REAS) v3.1 developed by Kurokawa and Ohara (2020). As REAS v3.1 is available up to 2015, the emissions after 2015 are estimated in a simple manner utilizing surface observations. Only a few studies aimed to reproduce recent trends in PM_{2.5} concentrations over East Asia using atmospheric chemical transport models. For example, Zhang *et al.* (2019) simulated PM_{2.5} over China for 2013–2017 using the Multi-resolution Emission Inventory (MEIC) inventories corresponding to each simulation year. They showed considerable decreasing trends in surface PM_{2.5} concentrations both from surface observations and model simulations, although their annual mean PM_{2.5} concentrations are underestimated in the northeastern coast (Beijing-Tianjin-Hebei region) and overestimated in the southeastern coast (Yangtze River Delta (YRD) region). In the present study, we evaluate PM_{2.5} over South Korea as well as China at various time scales (from daily to interannual scales) and verify whether REAS v3.1 inventories and emission adjustments are able to reproduce the magnitudes and trends in PM_{2.5} for both the countries. Three target regions are considered in this study, which are the Seoul Metropolitan Area (SMA), the North China Plain (NCP), and the YRD. The NCP region including the Beijing-Tianjin-Hebei region is regarded as the largest anthropogenic emission source region over East Asia and is located northwest of the YRD region (Fig. 1). Thus, when northwesterly synoptic winds prevail in winter and spring seasons, the NCP region serves as the upwind region and the YRD region becomes the downwind region (Fig. S1). Likewise, the SMA is characterized by the downwind region as it is located east of both the NCP and YRD regions under the influence of mid-latitude westerlies. In the present study, we aim to assess the effects of recent changes in anthropogenic emissions on winter–spring season PM_{2.5} and compare the responses of PM_{2.5} to emissions and meteorology in the upwind and downwind regions.

In Section 2, details of the modeling system and datasets are described. Because we use all input data that are openly available to public, one can follow and apply the methods to a modeling system for other regions or studies. In Section 3, the results of impact of emissions on recent PM_{2.5} and the roles of meteorology in the upwind and downwind regions are presented and discussed. Conclusions are given in Section 4.

Fig. 1. Monthly mean of hourly allocated (a) OC, (b) BC, and (c) PM_{2.5} emissions in February 2014. (b), (e), and (f) are the same as (a), (b), and (c), respectively but in May 2014. The emissions over South Korea are enlarged in the right-lower corner. The boundaries of North China Plain (NCP), Yangtze River Delta (YRD), and Seoul Metropolitan Area (SMA) are depicted with their abbreviations in (d).

2 METHODS AND DATA

2.1 WRF-Chem Modeling

We use the WRF-Chem model version 4.1.2 (Skamarock *et al.*, 2019). The model domain of this study is shown in Fig. 1 together with examples of monthly means organic carbon (OC), black carbon (BC), and PM_{2.5} emissions. The horizontal grid size is 20 km, and the number of grids in the zonal and meridional direction is 180 and 150, respectively. The number of total vertical layers is 40 with 11 layers located below 2 km. Initial and boundary conditions for meteorology are derived using the ERA-5 reanalysis (at 3-hour intervals for boundary conditions). The list of physics options used in this study is given in Table 1. Six sets of 1-month simulations for February and May are conducted for the years of 2013–2018. All simulations start at 15 UTC from 24 January to 28 February, and at 15 UTC from 24 April to 31 May. The first week of outputs are used as spin-up and not used in the analyses. To avoid the growth of errors in meteorology with time in the month-long simulations, four-dimensional data assimilation (FDDA) is employed in all the simulations. Analysis nudging for winds, temperature, and humidity at 3-hour intervals is applied above the boundary layer. The nudging coefficient for all the nudged variables is set to $6 \times 10^{-4} \text{s}^{-1}$ following Pan *et al.* (2015).

For gas and aerosol chemistry and dynamics, we use MOZART-4 mechanism coupled with MOSAIC aerosol module (Zaveri *et al.*, 2008) as described in Knote *et al.* (2014) and Hodzic *et al.* (2014). This mechanism is called *MOZART_MOSAIC_4bin_KPP* in the WRF-Chem. The details of

Table 1. Model configuration for meteorology.

Description/Physic scheme	Selection	Reference
Horizontal grid size (number of grids, $n_x \times n_y$)	20 km (180×15)	
Number of vertical layers (top pressure)	40 (40 hPa)	
Initial/boundary conditions, updated intervals	ERA-5, 3-hour	Hersbach <i>et al.</i> (2020)
Shortwave and longwave radiation	RRTMG	Iacono <i>et al.</i> (2008)
Boundary layer	MYNN	Nakanishi and Niino (2006, 2009)
Land surface	Noah	Tewari <i>et al.</i> (2004)
Urban canopy model (UCM) and parameters used in the UCM	WRF–single-layer UCM and UCM parameters for Seoul Metropolitan Area	Kusaka <i>et al.</i> (2001) Ryu and Baik (2013) for the UCM parameters
Microphysics	Morrison two-moment	Morrison <i>et al.</i> (2009)
Cumulus	Grell-Freitas	Grell and Freitas (2014)

Table 2. Model configuration for gas and aerosols.

Description	Selection	Reference
Gas-aerosol chemistry	MOZART-MOSAIC	Hodzic <i>et al.</i> (2014); Knote <i>et al.</i> (2014)
SOA chemistry	Empirical parameterization based on anthropogenic and biomass burning CO emissions.	Hodzic and Jimenez (2011)
Photolysis	Updated TUV using cloud optical depth computed in RRTMG shortwave scheme	Ryu <i>et al.</i> (2018, 2019)
Dry deposition	Wesely (1989)	
Wet deposition	Neu and Prather (2012) for soluble gases Wang <i>et al.</i> (2014) for below-cloud scavenging of aerosols	
Initial/boundary conditions, updated intervals	CAMS, 3-hour	Inness <i>et al.</i> (2019)
Anthropogenic emissions	REAS v3.1	Kurokawa and Ohara (2020)
Biogenic emissions	MEGAN v2.04	Guenther <i>et al.</i> (2012)
Biomass burning emissions	GFAS v1.2	Kaiser <i>et al.</i> (2012); Rémy <i>et al.</i> (2017)
Dust/sea salt emissions	GOCART	Zhao <i>et al.</i> (2010)

schemes relevant to chemistry and aerosols are summarized in Table 2. Secondary organic aerosols (SOAs) are included following the approach of Hodzic and Jimenez (2011). For below-cloud scavenging of aerosols, the semi-empirical formulae developed by Wang *et al.* (2014) are implemented in the present study. As in Wang *et al.* (2014), the scavenging coefficients depend on the aerosol size and rain intensity. Note that a constant factor of 5×10^2 is applied to the scavenging coefficients estimates for better agreements with observed scavenging coefficients given in Bae *et al.* (2010). We consider rain rates as a sum of resolved rain (from microphysics scheme) and parameterized rain (from cumulus scheme).

Unlike many previous studies that used atmospheric composition forecasts as initial and boundary conditions for trace gases and aerosols, for example forecasts from MOZART global chemistry model (e.g., Ryu *et al.*, 2019) or GEOS-Chem (e.g., Mao *et al.*, 2016), we use the CAMS reanalysis (Inness *et al.*, 2019) as initial and boundary conditions at 3-hour intervals. The CAMS reanalysis is a global atmospheric composition reanalysis with horizontal resolutions of ~ 80 km and it is available since 2003. Several satellite retrievals such as total column CO, tropospheric column NO₂, aerosol optical depth, total and partial O₃ column, and O₃ profiles are assimilated in CAMS with the ECMWF's Integrated Forecasting System (Inness *et al.*, 2019). Ryu and Min (2021) evaluated the performance of CAMS against surface observations for 2003–2018 over South Korea and showed satisfactory performance of CAMS in capturing daily variations as well as annual mean trends. Therefore, it is expected that realistic gas and aerosols are provided as boundary forcing in our simulations. The details of conversion of CAMS species to the WRF-Chem MOZART-MOSAIC species can be found in Table S1. The WRF-Chem preprocessor tool, mozbc, is

utilized to ingest CAMS reanalysis and create initial and lateral boundary conditions for various gas and aerosol species.

To investigate how sensitive PM_{2.5} levels are to the changes in emissions, we conduct a series of sensitivity simulations in which the 2014 emissions are used in all years. The reasons for choosing 2014 are that 1) the simulated PM_{2.5} levels show a good agreement with surface observations, and 2) OC emissions over South Korea are the highest in 2014 over the recent 6 years (2010–2015). The WRF-Chem simulations use the time-varying emissions as the control simulations.

2.2 Emissions

2.2.1 Anthropogenic emissions

Anthropogenic emissions play an essential role in controlling the levels of atmospheric pollutants, and hence the accurate representation of anthropogenic emissions is critical in air quality modeling. Here we have updated emission inventories for 2013–2018 based on the existing inventories over Asia and surface PM observations. We use the REAS version 3.1 recently developed by Kurokawa and Ohara (2020) for 2013–2015 as it is available from 1950 to 2015. For 2016–2018, the 2015 emissions are scaled down linearly or kept the same (Fig. 2). In the present study, the reduction amounts are first estimated for 2018. For China, the best agreement between the 2018 simulations and surface observations is obtained when PM emissions were reduced by 39% from the 2015 emissions. Between 2015 and 2018, 13% reduction is applied per year assuming linear decreases from 2015 to 2018. The reductions in OC and BC emissions until 2017 are similar to those estimated by Zheng *et al.* (2018) (Fig. 2). The PM_{2.5} emissions from REAS v3.1 (2010–2015) are larger than those estimated by Zheng *et al.* (2018) particularly during 2014–2015. Our estimate for 2016 PM_{2.5} is also somewhat larger than that estimated by Zheng *et al.* (2018) by 15% (9.56 Tg in this study vs. 8.1 Tg in Zheng *et al.*, 2018), whereas they are similar for the year 2017. Considering generally large uncertainties in PM emissions, e.g., ±94% for REAS v3 PM_{2.5} emissions over China in 2015, this discrepancy can be regarded as acceptable.

For South Korea, OC and BC emissions are decreased by 10% each year from those in 2015, but PM_{2.5} emissions are kept the same as in 2015 by reflecting the recent trends. The trends of other emissions from China and South Korea are shown in Fig. S2. The description of how emissions are modified or estimated is given in Text S1. Note that the emissions from the other regions (e.g., Taiwan and Japan) after 2016 are not modified and so remain the same as in 2015.

REAS v3.1 provide monthly emissions at 0.25° × 0.25° resolutions and includes four major sectors (residential, industry, energy, and transport), three additional sectors for NMVOC (extraction, solvent, and waste treatment), and three others for NH₃ (fertilizer, manure management, and miscellaneous). The monthly total emissions provided by REAS v3.1 are allocated into hourly emissions for use in WRF-Chem simulations. We apply weekly, holiday, and diurnal profiles (Fig. 3) to monthly emissions for each species and each sector as follows:

$$E_{s,i} = E_{s,m} \frac{W_{s,j}}{\sum_{j=1}^{nday} W_{s,j}} \frac{D_{s,i}}{\sum_{i=1}^{24} D_{s,i}} \quad (1)$$

where $E_{s,i}$ is the hourly emission at local time i for sector s , $E_{s,m}$ is the monthly emission for month m and sector s , $W_{s,j}$ is the weekly profile for day j and sector s , $D_{s,i}$ is the diurnal profile at local time i and sector s , and $nday$ is the number of days in month m . Note that the sum values of weekly (Monday through Sunday) and diurnal profiles are 7 and 24, respectively. In case a weekday belongs to a holiday, the Sunday profile is applied to the temporal allocation. For China, there are special workdays that are originally weekend days but assigned to workdays to compensate for the absence of economic activities during long holiday seasons. In this case, the special workdays are assumed to be the same as weekdays, and so the weekday profiles are applied to the temporal allocation. The majority of weekly and diurnal profiles by sectors, except for the transportation sector, are adopted from the CAMS-TEMPO dataset (Guevara *et al.*, 2020) and MACC fixed temporal profile by Denier van der Gon *et al.* (2011). For transportation, 2018 traffic volume data monitored by Gyeonggi Traffic Information Center of South Korea are utilized (<https://gits.gg.go.kr/gtdb/web/trafficDb/TrafficVolume/SangTimeVolumeTraffic.do>), and the

Fig. 2. Trends of year total (a) OC, (b) BC, and (e) $\text{PM}_{2.5}$ emissions over South Korea (left) and over China (right) for 2010–2015 from REASv3.1 inventory. The black dots indicate the year total emissions used (and assumed for 2016–2018) in this study. The blue squares indicate the estimated emissions by Zheng *et al.* (2018).

weekly and diurnal profiles for weekday, Saturday, and Sunday are obtained based on the 2018 traffic volume data (Figs. 3(a) and 3(b)). Unlike the CAMS-TEMPO dataset that considers some spatial differences in temporal profiles by country or urban/rural areas, the same temporal profiles are applied to all the grids except for industry and residential diurnal profiles. Slightly different industry diurnal profiles are applied to South Korea and China (Fig. 3(c)) by considering high nighttime NO_2 concentration over China. However, these differences in industry diurnal profiles are found to have little impact on nighttime NO_2 concentration over China (not shown). For the residential sector, the CAMS-TEMPO has three different diurnal profiles: the first one for CO, NMVOC, aerosols, and NH_3 in urban areas (the residential CO profile in urban areas in Fig. 3(d)), the second one for NO_x and SO_x in urban areas (the residential NO_x profile in urban areas in Fig. 3(d)), and the third one for all species in rural areas (the residential profile in rural areas in Fig. 3(d)). In the present study, however, we adopt the CO-type residential profile in urban areas only for CO, and the profiles for the other species in urban areas follow the NO_x -type profile. All species use the same residential profile in rural areas.

Fig. 3. (a) Weekly profiles of various sectors and diurnal profiles of (b) transportation, (c) industry and energy, (d) residential, and (e) solvent, waste treatment and agriculture sectors. The weekly profiles for residential sector are the same as those for industrial sector, and those for waste treatment and agriculture sectors are the same each other.

After temporal allocation, the hourly emissions at $0.25^{\circ} \times 0.25^{\circ}$ are spatially interpolated (regrided) to the $20 \text{ km} \times 20 \text{ km}$ WRF-Chem grid using the Earth System Modeling Framework (ESMF) software embedded in the NCAR Command Language (NCL). For NMVOC, REAS v3.1 provides its own speciated VOC emissions. Some of them are directly used in the WRF-Chem, and the other lumped species are speciated for use in MOZART-4 mechanism. The details of mapping of REAS VOC species to MOZART-4 VOC species are given in Table S2. The $\text{PM}_{2.5}$ emissions also need to be speciated. They are speciated into sulfate, nitrate, ammonium, and inorganic aerosols

based on the weighting factors obtained from the SPECIATE tool developed by EPA (Table S3; <https://www.epa.gov/air-emissions-modeling/speciate>, last access: 25 June 2020). We use the simple PM_{2.5} speciation in the SPECIATE, and also adopt volatility basis set (VBS) PM speciation for transport sector. The details of how PM_{2.5} emissions are speciated are described in Text S2. Note that in this study the PM_{2.5} emissions excluding OC and BC emissions are increased by ~39% to compensate for the omission of heterogeneous sulfate formation via heterogeneous oxidation SO₂. Zheng *et al.* (2015) reported that heterogeneous oxidation of SO₂ largely increases sulfate concentration by a factor of 2.3. The increase in PM_{2.5} concentration excluding BC and OC in Zheng *et al.* (2015) due to heterogeneous chemistry is about 44%. Although the heterogeneous sulfate formation is not included in our study, the heterogeneous hydrolysis of dinitrogen pentoxide (N₂O₅) that produces particulate nitrate is considered. In the present study, the emissions from point sources are placed in the third lowest model level. According to Mar *et al.* (2016), the vertical allocation of emissions is not expected to significantly affect pollutant concentrations at the surface.

2.2.1 Biomass burning emissions

The GFAS version 1.2 is used in this study (Kaiser *et al.*, 2012; Rémy *et al.*, 2017). The GFAS assimilates Fire Radiative Power observations from MODIS instruments and provides biomass burning emissions at daily time scales. Rémy *et al.* (2017) extended the system's capability to include information of injection heights derived from fire observations and ECMWF meteorology forecasts. The GFAS horizontal resolution is 0.1° and available since 2003. In this study, vertical profiles of fire emissions are prescribed using two GFAS parameters: mean altitude of maximum injection and top of plume based on the profile given in Cussac *et al.* (2020). Fig. 4 illustrates an example of vertical profiles of injection fraction. In the present study, the profile below mean altitude of maximum injection is assumed to follow a logarithm function and that above is to follow an exponential function. The coefficient, a_2 , in Fig. 4 is equal to the top of plume. The injection fractions at the mean altitude of maximum injection and at the lowest model level are set to 0.2 and 0.025, respectively, as a first guess. After obtaining a_1 , b_1 , and b_2 in Fig. 4 with the height information and the prescribed functions, the initially determined vertical profile with a_1 ,

Fig. 4. An example of vertical profile of GFAS injection fraction estimated using mean altitude of maximum injection and top of plume parameters.

b_1 , a_2 , and b_2 are normalized so that the sum of the vertical profile is equal to 1. The GFAS emissions, which are basically column total emissions, at a grid point are then vertically allocated using the normalized vertical profile. The ESMF software is also used for GFAS emission regridding. Note that the daily GFAS emissions are converted to constant hourly emissions (no diurnal variation in biomass emissions over the course of the day), and then the hourly biomass emissions are added to the hourly anthropogenic emissions. The time series of biomass burning emissions during the study period are illustrated in Fig. S3.

2.3 Observations

2.3.1 Surface observation data over South Korea

We use gridded surface $\text{PM}_{2.5}$ observations data at $0.25^\circ \times 0.25^\circ$ over South Korea constructed by Ryu and Min (2021). The reason for using the gridded observations is that monitoring stations are non-uniformly located, and a simple average over stations can lead to a biased average with more weight being put on urban areas where the majority of stations are located rather than outside the cities. We choose the Seoul Metropolitan Area as our target region for model validation and take a spatial average over the SMA (Fig. 1(d)). The SMA include Seoul, the capital of South Korea, and two nearby provinces, Incheon and Gyeonggi. Surface $\text{PM}_{2.5}$ observations over South Korea are available since 2015. Daily $\text{PM}_{2.5}$ observations over Seoul, however, are available since 2013, so daily $\text{PM}_{2.5}$ over the SMA prior to 2015 are obtained from daily $\text{PM}_{2.5}$ over Seoul and daily PM_{10} data over the two regions. Note that surface PM_{10} observations are available since 2001 over South Korea. The ratio of daily PM_{10} over the SMA to daily PM_{10} over Seoul is applied to daily $\text{PM}_{2.5}$ over Seoul, assuming that $\text{PM}_{2.5}/\text{PM}_{10}$ ratio over the SMA is the same as the ratio over Seoul.

2.3.2 China surface reanalysis

We utilize surface $\text{PM}_{2.5}$ reanalysis data over China recently developed by Kong *et al.* (2020). The dataset has $15 \text{ km} \times 15 \text{ km}$ spatial resolutions at hourly time scales for the period of 2013–2018. They assimilated surface observations at more than 1000 monitoring stations over China in the Nested Air Quality Prediction Modeling System using the ensemble Kalman filter. The datasets show excellent performance in reproducing the magnitude and variability of surface $\text{PM}_{2.5}$ and also high accuracy compared to independent observations over China (Kong *et al.*, 2020). As it is hard to access Chinese surface observation data outside China, we use the surface reanalysis data as pseudo-observation data. The gridded reanalysis data can mitigate the problems arising from large spatial heterogeneity of surface observations. For model validation, the gridded $\text{PM}_{2.5}$ data are averaged over the NCP and the YRD regions (Fig. 1(d)), and daily $\text{PM}_{2.5}$ data over the two regions are used.

It should be noted that $\text{PM}_{2.5}$ data when influenced by Asian dust are excluded in this study because the model is found to have poor performance in capturing dust events (e.g., marked by yellow circles in Figs. 7 and 8). A dust day for China is defined when observed $\text{PM}_{10}/\text{PM}_{2.5}$ ratio is greater than or equal to 2.5 and observed daily PM_{10} is greater than or equal to $125 \mu\text{g m}^{-3}$. For South Korea, we use measurement and identification of Asian dust day by the Korea Meteorological Administration (<https://www.weather.go.kr/weather/asiandust/observday.jsp>) to filter out dust days. A further study that includes improved dust emissions and associated physics is required in the future.

2.4 2016 KORUS-AQ Campaign Data

The Korea-United States Air Quality (KORUS-AQ) field campaign has taken place over Korean Peninsula and nearby sea during May–June 2016. We use 15 flight days in May 2016 as our simulation covers only May. The 15 flight tracks used in this study are shown in Fig. S4. As organic carbon, black carbon, sulfate, nitrate, and ammonium aerosols are measured, those species from the simulations are accordingly used in the evaluation. The samples over the SMA are separated from the outside regions to focus on the highly polluted area (the SMA). The simulated aerosol mass concentrations are converted to mass concentrations at standard temperature (273.15 K) and pressure (1013.25 hPa). The vertical levels are binned with 200 m intervals and the lowest layer is centered at 100 m above ground.

3 RESULTS AND DISCUSSION

3.1 Spatial Distribution of 6-year (2013–2018) Averaged Monthly $\text{PM}_{2.5}$

Fig. 5 compares the spatial distribution of monthly mean $\text{PM}_{2.5}$ concentrations averaged for 2013–2018 from the China surface reanalysis and the WRF-Chem control simulations. Both the reanalysis and WRF-Chem control simulations exhibit substantially higher $\text{PM}_{2.5}$ levels over China in February than in May due to 1) higher pollutant emissions in winter than in spring and 2) more frequent stable and stagnant atmospheric conditions in winter than in spring. In February, the model generally captures the spatial distribution and magnitude of surface $\text{PM}_{2.5}$ over China (e.g., the NCP and YRD) with the exception of a few regions of central China where $\text{PM}_{2.5}$ levels are largely overestimated (Henan, Chongqing, and southwestern Sichuan). In May, the model reproduces reasonably well $\text{PM}_{2.5}$ over the NCP, YRD, and central China except for Henan Province. Over the

Fig. 5. Spatial maps of 6-year (2013–2018) averaged monthly $\text{PM}_{2.5}$ in February from (a) the China surface reanalysis (CNRA) and (b) the WRF-Chem simulations. (c) and (d) are the same as (a) and (b), respectively, but in May.

dust regions of southern Mongolia and northern China (Inner Mongolia), the model tends to underestimate $\text{PM}_{2.5}$ (especially in May) likely due to underestimated dust aerosols. The lower $\text{PM}_{2.5}$ over southern China (e.g., Guangxi, Guangdong, and Fujian) in the control simulations than in the reanalysis is likely due to the overestimated wet deposition in the subtropical regions (see the 6-year averaged precipitation in Fig. S5). The large biases in the long-term averaged $\text{PM}_{2.5}$ seen over central China for both seasons imply that the emissions are likely overestimated in REAS v3.1 and thus need to be revised for this region.

3.2 Temporal Variations—Trends, Interannual and Daily Variations

Fig. 6 shows the monthly mean surface $\text{PM}_{2.5}$ concentrations simulated and observed over the SMA, NCP, and YRD for February and May 2013–2018. Both simulated and observed $\text{PM}_{2.5}$ concentrations show decreasing trends, and the control simulations capture reasonably well the trends and monthly mean values. The observed $\text{PM}_{2.5}$ over the SMA shows small decreasing trends in February ($-0.21 \mu\text{g m}^{-3} \text{ year}^{-1}$) and slightly stronger decreasing trends in May ($-2.3 \mu\text{g m}^{-3} \text{ year}^{-1}$). The trends in simulated $\text{PM}_{2.5}$ are $-1.81 \mu\text{g m}^{-3} \text{ year}^{-1}$ in February and $-2.68 \mu\text{g m}^{-3} \text{ year}^{-1}$ in May. The model is not able to capture the slightly elevated $\text{PM}_{2.5}$ concentrations observed in February 2017–2018, which leads to the larger decreasing trends in magnitude in the control simulations. Further discussion on the model errors and potential sources of errors is given in Section 3.3. For China, $\text{PM}_{2.5}$ in February over the NCP decreases at a rate of $-9.6 \mu\text{g m}^{-3} \text{ year}^{-1}$ in the observations and $-8.4 \mu\text{g m}^{-3} \text{ year}^{-1}$ in the control simulations. For February, the NCP $\text{PM}_{2.5}$ is reduced by 43% during 2013–2018 in the observations and by 39% in the control simulations. The corresponding reduction rates over the YRD are 21% in the observations and 17% in the control simulations. For May, the reduction rate over the NCP is underestimated (27% in the control simulations and 41% in the observations), but the simulated reduction rate over YRD (35%) is similar to the observed one (33%). The performance statistics of the normalized mean bias (NMB), systematic root-mean-square error (RMSE), and unsystematic RMSE computed using daily mean $\text{PM}_{2.5}$ are given in Table 3. The systematic RMSE is based on the difference between linearly fitted model

Fig. 6. Timeseries of mean $\text{PM}_{2.5}$ in February 2013–2018 over the (a) SMA, (b) NCP, and (c) YRD. (d), (e), and (f) are the same as in (a), (b), and (c), respectively, but for mean $\text{PM}_{2.5}$ in May. The “OBS”, “CNTR”, and “emis2014” in the legend indicate observations, control simulation, and sensitivity simulation that uses 2014 emissions for all years, respectively. The dotted lines indicate linear regression lines, and the small numbers indicate the slopes of linear regression lines in $\mu\text{g m}^{-3} \text{ year}^{-1}$.

Table 3. Performance statistics for 2013–2018. The unit of normalized mean bias (NMB) is %, and the unit of systematic root-mean-square error (RMSEs) and unsystematic root-mean-square error (RMSEu) is $\mu\text{g m}^{-3}$.

YEAR	SMA FEBRUARY				NCP FEBRUARY				YRD FEBRUARY				SMA MAY				NCP MAY				YRD MAY			
	NMB	RMSES	RMSEU	RMSE	NMB	RMSES	RMSEU	RMSE	NMB	RMSES	RMSEU	RMSE	NMB	RMSES	RMSEU	RMSE	NMB	RMSES	RMSEU	RMSE	NMB	RMSES	RMSEU	
2013	-9.5	2.8	6.6		-8.4	17.8	24.8		-3.8	7.1	16.2		1.5	1.7	10.4		-17.2	13.3		9.3		-14.8	8.1	12.9
2014	5.4	10.4	17.8		-3.4	11.5	16.8		-4.5	2.8	14.3		2.4	2.8	6.6		-1.4	0.9		13.2		-11.1	6.6	14.2
2015	2.3	3.2	10.4		7.5	6.7	17.4		18.6	13.4	16.0		16.9	4.9	9.8		-6.8	5.3		9.1		-19.2	11.1	11.0
2016	-2.7	0.7	6.2		20.9	14.3	15.6		17.7	10.4	16.9		-8.8	6.8	6.7		0.7	1.4		7.2		-2.7	3.1	10.9
2017	-22.9	8.4	5.3		-8.2	11.8	13.1		2.4	1.5	18.0		5.6	4.0	10.2		-1.0	2.3		7.3		-1.4	2.7	8.8
2018	-26.4	9.0	6.4		-0.8	11.3	14.2		0.8	14.8	13.9		-7.9	3.4	5.1		2.5	2.0		4.9		-17.1	9.1	7.5

values (fitted model simulated values to observations) and observations, and the unsystematic RMSE is the RMSE between linearly fitted model values and model-simulated values (Willmott, 1981). The NMBs are in general within $\pm 25\%$ (Table 3), the RMSE values are smaller than $30 \mu\text{g m}^{-3}$, and the correlation coefficients are generally greater than 0.7 (Figs. 7 and 8). The model performs better in spring (May) than in winter (February) in terms of monthly mean values (Fig. 6). The performance of temporal correlation is found to be slightly degraded in May as compared to that in February particularly over the YRD (Fig. 8).

One may ask a question of how much interannual variations in meteorology contribute to the interannual variations in $\text{PM}_{2.5}$. We show that the interannual variations in monthly mean $\text{PM}_{2.5}$ due to those in meteorology is not negligible and can be up to $10.5 \mu\text{g m}^{-3}$ in February (the difference between $37.0 \mu\text{g m}^{-3}$ in 2014 and $26.5 \mu\text{g m}^{-3}$ in 2018 in the sensitivity simulations) and by $13.3 \mu\text{g m}^{-3}$ in May (the difference between $36.5 \mu\text{g m}^{-3}$ in 2013 and $23.2 \mu\text{g m}^{-3}$ in 2018) over the SMA with the fixed emissions. The monthly mean $\text{PM}_{2.5}$ is approximately $30 \mu\text{g m}^{-3}$ over SMA. This result indicates that about 35% (45%) of monthly mean $\text{PM}_{2.5}$ can be modulated by meteorology in winter (spring) at maximum. Similarly, the YRD reveals large interannual variations in $\text{PM}_{2.5}$ of $19.0 \mu\text{g m}^{-3}$ in February and of $12.0 \mu\text{g m}^{-3}$ in May with the fixed emissions, which correspond to 27.6% in February and 26.9% in May of its monthly means. On the other hand, the NCP shows relatively small interannual variations in $\text{PM}_{2.5}$ due to meteorology of $10.6 \mu\text{g m}^{-3}$ in February and $6.5 \mu\text{g m}^{-3}$ in May. These correspond to 10.9% in February and 11.8% in May of its monthly means in the sensitivity simulations. As can be seen in the emission distributions (Fig. 1), the emissions from the NCP are very large and larger by 40% than those from the YRD and by 200% than those from the SMA. Thus, $\text{PM}_{2.5}$ over the NCP is mainly influenced by the local emissions in particular in winter when stagnant conditions often prevail. On the other hand, the YRD and SMA are located downwind of the NCP and China, respectively, so $\text{PM}_{2.5}$ over these regions can be substantially influenced by the long-range transport of $\text{PM}_{2.5}$ from the upwind regions. A similar result is found from a case study by Kang *et al.* (2019), highlighting a large contribution of $\text{PM}_{2.5}$ transported from the NCP to the YRD during a cold frontal episode. Therefore, the role of meteorology in the downwind regions can be larger than that in the upwind region.

Despite the non-negligible role of meteorology in the interannual variations in $\text{PM}_{2.5}$, the decreasing trends of $\text{PM}_{2.5}$ are hardly explained only by meteorology (Fig. 6). In other words, the decreases in $\text{PM}_{2.5}$ are mostly due to the reductions in emissions especially for China. Similar result was found for the past when $\text{PM}_{2.5}$ concentration increased over eastern China during 1985–2005 (Yang *et al.*, 2016). They showed that the increasing trends of $\text{PM}_{2.5}$ are largely influenced by anthropogenic emissions ($10.5 \pm 6.2 \mu\text{g m}^{-3} \text{ decade}^{-1}$) than by meteorology (e.g., $1.8 \pm 1.5 \mu\text{g m}^{-3} \text{ decade}^{-1}$) during 1985–2005. These results underscore the effectiveness of emission controls and are consistent with many recent studies for China

Fig. 7. Daily PM_{2.5} in February in (a, b, c) 2013; (d, e, f) 2014; (g, h, i) 2015; (j, k, l) 2016; (m, n, o) 2017; and (p, q, r) 2018 over the SMA (left column), the NCP (middle column), and the YRD (right column), respectively. The “rmse” and “corr” indicate the root-mean-square-error and linear correlation coefficient, respectively. The yellow circles indicate the days on which Asian dust events occur. Note that the y-axis scale for South Korea differs from that for China.

based on surface observations (Ding *et al.*, 2019; Zhai *et al.*, 2019) and satellite observations (Ma *et al.*, 2019). Also, these results suggest that the use of outdated emissions (e.g., the base year of 2014) likely leads to large errors in PM_{2.5} as well as other pollutants, which was confirmed by a numerical study by Chen *et al.* (2019). The large adjustments made in the China surface reanalysis data by Kong *et al.* (2020) also support large discrepancies between their *a priori* emissions (2010 HTAP v2) and true emissions. Therefore, it is recommended that appropriate and reasonable trends in emissions should be considered in air quality modeling over East Asia in particular if one is interested in recent years and the future.

Fig. 8. Same as in Fig. 7, but for May.

3.3 Potential Sources of Errors

In Fig. 6(a), it is seen that the simulated $PM_{2.5}$ values in February 2017–2018 is smaller than the observed $PM_{2.5}$ values. The higher emissions (the base year of 2014) in the sensitivity simulations than in the control simulations result in only small increases in $PM_{2.5}$ by $\sim 3 \mu g m^{-3}$. In the daily time series (Fig. 7(m)), the largest underestimation is found in 4–5 February 2017 and this mainly contributes to the negative bias of the February mean in 2017. The increased emissions lead to a slightly better agreement with observations during this episode in the sensitivity simulations than in the control simulations, but $PM_{2.5}$ is still largely underestimated. Large errors during specific episodes are similarly found over the NCP and the YRD: The large biases in monthly mean $PM_{2.5}$ are often due to the large errors in specific episodes (e.g., 2–4 and 15–19 February 2016 and 12–14 February 2017 over the NCP and 16–17 February 2015, 15–19 February 2016, and 4–

5 February 2017 over the YRD). Identifying and quantifying the sources of errors is difficult and not the main goal of this study. However, here we propose some potential rationales for the sources of errors. In general, the model bias errors show large daily variations; that is, the bias errors are sporadically large in short-term episodes rather than are consistently present throughout the month. In some cases, however, persistent biases throughout the month are found (e.g., February 2018 over the SMA, May 2013 over the NCP, and May 2018 over the YRD). In these cases, the systematic RMSEs are larger than the unsystematic RMSEs (Table 3). This means that there are considerable systematic errors that can be corrected by linear scaling because the systematic RMSE is computed as an RMSE of linear regressed model results against observations. The systematic errors in these cases are likely originating from emissions because the scaling up of emissions acts to monotonically and linearly increase $\text{PM}_{2.5}$ concentration throughout the month (Figs. 7 and 8). The reductions in systematic RMSEs with the increased emissions (the base year of 2014) in February 2018 over the SMA and in May over the YRD support this assertion (Figs. 7(p) and 8(r), and Table S4). Dry deposition can also be a source of systematic errors, but it is less likely because we use the same dry deposition scheme throughout all years and the monthly means of simulated $\text{PM}_{2.5}$ do not show a constant bias throughout years relative to the observed ones. On the other hand, the larger unsystematic RMSE than systematic RMSE can be interpreted as larger contributions from intermittent sources such as meteorology and wet deposition, which is the case for most runs (Table 3). The large daily variation in bias errors indeed reflects the large unsystematic errors. A systematic and in-depth research will be required in the future to identify and quantify the sources of errors.

3.4 $\text{PM}_{2.5}$ Composition

The changes in $\text{PM}_{2.5}$ composition during 2013–2018 are shown in Fig. 9. Because there are few comprehensive observations of $\text{PM}_{2.5}$ composition over the study area, we only present the model simulation results. For the SMA, the trends of $\text{PM}_{2.5}$ composition are quite complex. That is, not all species show the same trends. For example, the primary aerosols (OC, BC, and inorganic aerosols) over the SMA increases during February 2014–2015 while nitrate, ammonium and the total $\text{PM}_{2.5}$ concentrations decrease. Sulfate aerosols are also slightly higher in February 2015

Fig. 9. Timeseries of February mean $\text{PM}_{2.5}$ composition in February for 2013–2018 over the (a) SMA, (b) NCP, and (c) YRD. (d), (e), and (f) are the same as in (a), (b), and (c), respectively, but for May mean $\text{PM}_{2.5}$ in May. NO_3 is the nitrate, NH_4 is the ammonium, SO_4 is the sulfate, and OIN is the other inorganic aerosols.

than in February 2014. After 2015, the majority of PM_{2.5} components generally decrease, but some species show opposite tendencies (i.e., increasing tendencies compared to their previous year concentrations). The influence of meteorology and its interannual variations are presumed to contribute to the complex behaviors of secondary aerosols. As an example, Wang *et al.* (2020) demonstrated that upward transport of pollutants and increased humidity accompanied by a cold front facilitate heterogeneous and aqueous-phase oxidation of precursors, leading to increases in secondary inorganic aerosol formation and concentrations.

For China, the trends of primary aerosols are generally similar to those of total PM_{2.5}. On the other hand, the secondary aerosols do not always follow the decreasing trends of total PM_{2.5} especially in winter. For example, the concentration of nitrate aerosols over the NCP is higher in 2018 than in 2017, even though NO_x emissions over China continue to decrease. Sulfate aerosols show decreasing tendencies in general over the NCP, but their tendencies over the YRD are less clear, suggesting that the large reduction in SO₂ emissions seem effective in the large source region. It is also noteworthy that the mass fraction of secondary aerosols is higher in the downwind regions (51% over the SMA and 45% over the YRD in February) than in the upwind region (32% over the NCP in February), and is even higher in spring (58% over the SMA, 47% over the YRD, and 43% over the NCP in May) than in winter. Because the trends and magnitudes of secondary aerosols are complex and do not always depend on their precursors' emissions, more comprehensive understanding and regulation policies are therefore required to reduce the levels of secondary aerosols over East Asia.

3.5 Vertical Profiles during KORUS-AQ Campaign

Fig. 10 evaluates the vertical distribution of aerosols simulated by the model against the observations during the KORUS-AQ campaign in May 2016. The simulated aerosol profiles averaged over all samples show a good agreement with the observations in general (Fig. 10(a)). The aerosols over the SMA are found to be slightly underestimated in the lower boundary layer (below ~0.5 km) and also in the upper boundary layer (1–1.5 km). The underestimation in the lower boundary layer is consistent with the slight underestimation in PM_{2.5} at the surface (Fig. 8(j)). The aerosols outside the SMA, on the other hand, are slightly overestimated near the surface and also in the mid-to-upper boundary layer. The purpose of this evaluation is to examine the model's capability in capturing overall vertical profiles of aerosols over relatively large areas (the SMA or entire campaign domain). Because of the relatively coarse horizontal resolution (20 km), a point-by-point comparison between modeled and observed aerosols shows not only good agreement but also large discrepancies (not shown). As the land surface characteristics and spatial distributions of emissions are highly heterogeneous over South Korea, high-resolution simulations will be required for a more thorough and detailed evaluation of the modeling system.

Fig. 10. Vertical profiles of aerosols (organic and black carbon, sulfate, nitrate, and ammonium) from the KORUS air-borne observations and from the WRF-Chem simulations averaged over (a) all region, (b) the SMA, and (c) outside the SMA.

Fig. 11. Scatter plot of monthly mean primary PM (OC, BC, and $\text{PM}_{2.5}$) emissions and monthly mean surface $\text{PM}_{2.5}$ from the WRF-Chem control simulations. The gray solid line is the linear regression line computed using the results of 2013–2018 February for the NCP and 2015–2018 February for the YRD. The gray dotted line is the linear regression line computed using all May results. The two ending digits of each corresponding year are denoted by small number with the color same as the marker, but slightly darker color is used for the SMA. The year is basically marked right above or below the marker, but adjusted for the SMA to avoid overlaps.

3.6 Anthropogenic Emission Rates versus Surface $\text{PM}_{2.5}$

In this subsection, the relationship between emission rates and surface $\text{PM}_{2.5}$ concentrations is explored to provide an insight into how much $\text{PM}_{2.5}$ changes in accordance with changes in emissions. In the emission rates, primary PM emissions are only considered. That is, the contribution of secondary aerosol formation is not considered because the secondary aerosol formation is largely affected by levels of precursors, meteorological conditions (e.g., temperature, relative humidity, and UV intensity), and other factors. It is noteworthy that the contributions and associated uncertainties of SOA to $\text{PM}_{2.5}$ concentrations would be small in winter (~4–9%), but become larger in spring (~10–20%) (Fig. 9).

As expected, the NCP clearly shows a linear relationship between emissions and surface $\text{PM}_{2.5}$ concentrations in winter (February) (Fig. 11). The relationship between emissions and $\text{PM}_{2.5}$ is less clear over the YRD than over the NCP (Fig. 11). The YRD results for February 2013–2014 are off from the linear tendency embracing the results for the NCP and 2015–2018 YRD. Thus, the results for February 2013–2014 YRD are not included in the linear regression. One potential reason for this is the much stronger winds over the YRD in February 2013–2014 than in February 2015–2018, which could facilitate ventilation and less accumulation of pollutants over the YRD during 2013–2014. The 1000 hPa mean wind speed in February over the YRD from the ERA-5 reanalysis data is 2.9 m s^{-1} in 2013, 3.6 m s^{-1} in 2014, 1.4 m s^{-1} in 2015, 0.57 m s^{-1} in 2016, 1.2 m s^{-1} in 2017, and 1.0 m s^{-1} in 2018. It would be interesting to explore why $\text{PM}_{2.5}$ in February 2013–2014 over the YRD shows different responses to emissions in a future study.

The SMA shows a positive relationship between emissions and $\text{PM}_{2.5}$ in February, and the slope of the linear regression is quite different from that for China winter cases. As the SMA data are rather densely placed and only two major clusters (2014 and 2015 versus 2013 and 2016–2018) are visible, we are cautious to draw a conclusive interpretation. More samples or simulations covering a longer period would be required to understand how emissions and $\text{PM}_{2.5}$ are related over the SMA.

The relationship between emissions and $\text{PM}_{2.5}$ in spring is generally linear, but the data points are much more scattered than those in winter. The linear regression line is plotted using all the May results for the three regions. The greater spread in May is likely due to the more significant role in meteorology in May. The distinctively smaller y-axis intercept and the smaller slope of the

regression line in May than in February also support the lower contribution of emissions to $\text{PM}_{2.5}$ in May than in February. For example, even if the monthly mean PM emission rate is the same as $0.2 \mu\text{g m}^{-2} \text{s}^{-1}$, the monthly mean $\text{PM}_{2.5}$ would be $79.5 \mu\text{g m}^{-3}$ in February and $54.2 \mu\text{g m}^{-3}$ in May based on our regression results. The increased role in meteorology in May relative to that in February suggests that more accurate modeling or forecast of meteorology is particularly required in spring.

4 CONCLUDING REMARKS

We present a WRF-Chem modeling system that utilizes current data, including an updated emission inventory for 2013–2018, to explain the recent trends in $\text{PM}_{2.5}$ during winter and spring. In spite of several discrepancies between the observations and the simulations, the model realistically captures the monthly means, trends, daily variations, and vertical profiles of this pollutant in the studied regions. It also attributes the decreasing trends in $\text{PM}_{2.5}$ concentrations over China to the emission regulations enacted in 2013. Therefore, we strongly recommend the use of accurate emission trend estimates in modeling both the recent and the future air quality of East Asia. Emission reduction caused larger decreases in the most polluted region, the upwind NCP (43% and 41% for February and May, respectively, between 2013 and 2018 based on the observations), than the two cleaner, downwind regions (e.g., in the YRD, 21% and 33% were the corresponding percentages). Additionally, the levels in the SMA showed little change during winter, although they decreased slightly during spring (at $-2.3 \mu\text{g m}^{-3} \text{year}^{-1}$ and $-2.7 \mu\text{g m}^{-3} \text{year}^{-1}$ based on the observations and the simulations, respectively).

Even though control measures mainly drove the decline in $\text{PM}_{2.5}$, meteorological conditions played a significant role in modulating both the concentration and the chemical composition of this pollutant, ultimately influencing its interannual variation. The effects of meteorology were more prominent in the downwind regions (the SMA and YRD) than the upwind one (the NCP); moreover, they were stronger during spring than winter. Specifically, when assuming a fixed rate of emission for the entire study period, this factor altered the mean monthly $\text{PM}_{2.5}$ concentration by as much as 45% and 35% during spring and winter, respectively, over the SMA. Differences of $\sim 27\%$ for the YRD and $\sim 11\%$ for the NCP were also estimated. As the secondary aerosols sometimes increased even while their precursors decreased, we presume that meteorology exerts a greater influence on the levels of the secondary aerosols. A steady bias that persists throughout a month of data, which is reflected by a large systematic RMSE, likely arises from errors in the emission data, but a sizable bias that sporadically appears probably stems from errors in the meteorological data. Therefore, in order to better assess the effects of emission reduction, more efforts should be dedicated to accurately representing meteorological conditions.

We have characterized the relationship between emission rates and $\text{PM}_{2.5}$ concentrations, which can be applied to estimate emissions in other studies. For example, one can evaluate the decrease in emissions during unusual or unexpected events, such as the COVID-19 pandemic, when historic $\text{PM}_{2.5}$ observations are available.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <https://doi.org/10.4209/aaqr.200654>

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