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 develops and evaluates a WRF-Chem modeling system that utilizes updated anthropogenic emissions for 2013–2018 over East Asia to reflect $PM_{2.5}$ changes due to recent emission regulations. The WRF-Chem model reasonably reproduces monthly means, daily variations, and vertical profiles in $PM_{2.5}$ in winter and spring over South Korea and China. Results show that the recent declines in $PM_{2.5}$ over China are attributed to China's emission control action effective since 2013. The effects of emission reductions are found to be larger in the severely polluted region (i.e., North China Plain, NCP) than less polluted regions or downwind regions (Yangtze River Delta, YRD, and Seoul Metropolitan Area, SMA). The simulated February-mean $PM_{2.5}$ is reduced by 39% during 2013–2018 over the NCP and by 17% over the YRD. The SMA shows small changes in $PM_{2.5}$ in winter but weak decreasing trends in spring. In addition to emissions, meteorology is found to considerably modulate $PM_{2.5}$. The effects of meteorology on the interannual variations in $PM_{2.5}$ are larger in the downwind regions (SMA and YRD regions) than in the upwind region (NCP), which can be up to 35% in winter and 45% in spring over the SMA. The corresponding effects over the NCP are 11% in winter and 12% in spring. More complex variations in secondary aerosols that do not always follow the decreasing trends in primary aerosol emissions are found in the downwind regions than in the upwind regions.

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 actions and ultimately to develop sustainable control strategies for the future (e.g., Gilliland et al., 2008; G. 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°×0.5°) and provide inventories of major pollutant species (CO, NO_X, non-methane volatile organic compound (NMVOC), SO_2, 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°×1.1° of TCR-2, Miyazaki et al., 2020), we choose a bottom-up type inventory, REASv3.1 (Regional Emission inventory in Asia) developed by Kurokawa and Ohara (2020). As REASv3.1 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 chemistry-transport models. For example, Zhang et al. (2019) simulated $PM_{2.5}$ over China for 2013–2017 using the MEIC (Multi-resolution Emission Inventory) 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 REASv3.1 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, supplementary). 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.

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 one-month simulations for February and May are conducted for the years of 2013–2018. All simulations start from 15 UTC 24 January to 28 February, and from 15 UTC 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 6×10^{-4} 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 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 X. Wang et al. (2014) are implemented in the present study. As in X. 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 NO_2, aerosol optical depth, total and partial O_3 column, and O_3 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 (Supplementary). 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 using 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 REASv3.1 (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 Zheng et al., 2018), whereas they are similar for the year 2017. Considering generally large uncertainties in PM emissions, e.g., ±94% for REASv3 $PM_{2.5}$ 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 (Supplementary). 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.

REASv3.1 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 NH_3 (fertilizer, manure management, and miscellaneous). The monthly total emissions provided by REASv3.1 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.

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

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 Gyeong-gi Traffic Information Center of South Korea are utilized (https://gits.gg.go.kr/gtdb/web/trafficDb/TrafficVolume/SangTimeVolumeTraffic.do), and the weekly and diurnal profiles for weekday, Saturday, and Sunday are obtained based on the 2018 traffic volume data (Figs. 3a and 3b). 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. 3c) 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. 3d), the second one for NO_X and SO_X in urban areas (the residential NO_X profile in urban areas in Fig. 3d), and the third one for all species in rural areas (the residential profile in rural areas in Fig. 3d). 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.

After temporal allocation, the hourly emissions at 0.25°×0.25° are spatially interpolated (regridded) to the 20 km×20 km WRF-Chem grid using the Earth System Modeling Framework (ESMF) software embedded in the NCAR Commend Language (NCL). For NMVOC, REASv3.1 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 (Supplementary). The $PM_{2.5}$ 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 (https://www.epa.gov/air-emissions-modeling/speciate, last access: 25 June 2020) (Table S3, Supplementary). 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 (Supplementary). 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 SO_2. Zheng et al. (2015) reported that heterogeneous oxidation of SO_2 largely increases sulfate concentration by a factor of 2.3. The

241 increase in $PM_{2.5}$ concentration excluding BC and OC in Zheng et al. (2015) due to heterogeneous
242 chemistry is about 44%. Although the heterogeneous sulfate formation is not included in our study,
243 the heterogeneous hydrolysis of dinitrogen pentoxide (N_2O_5) that produces particulate nitrate is
244 considered. In the present study, the emissions from point sources are placed in the 3rd lowest model
245 level. According to Mar et al. (2016), the vertical allocation of emissions is not expected to
246 significantly affect pollutant concentrations at the surface.
247
248 **2.2.1 Biomass burning emissions**
249 The GFAS version 1.2 is used in this study (Kaiser et al., 2012; Rémy et al., 2017). The
250 GFAS assimilates Fire Radiative Power observations from MODIS instruments and provides
251 biomass burning emissions at daily time scales. Rémy et al. (2017) extended the system's capability
252 to include information of injection heights derived from fire observations and ECMWF
253 meteorology forecasts. The GFAS horizontal resolution is 0.1° and available since 2003. In this
254 study, vertical profiles of fire emissions are prescribed using two GFAS parameters: mean altitude
255 of maximum injection and top of plume based on the profile given in Cussac et al. (2020). Figure
256 4 illustrates an example of vertical profiles of injection fraction. In the present study, the profile
257 below mean altitude of maximum injection is assumed to follow a logarithm function and that
258 above is to follow an exponential function. The coefficient, a_2, in Fig. 4 is equal to the top of plume.
259 The injection fractions at the mean altitude of maximum injection and at the lowest model level are
260 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
261 height information and the prescribed functions, the initially determined vertical profile with a_1, b_1,
262 a_2, and b_2 are normalized so that the sum of the vertical profile is equal to 1. The GFAS emissions,
263 which are basically column total emissions, at a grid point are then vertically allocated using the
264 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 timeseries of biomass burning emissions during the study period are illustrated in Supplementary (Fig. S3).

2.3 Observations

2.3.1 Surface observation data over South Korea

We use gridded surface $PM_{2.5}$ observations data at 0.25°×0.25° 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 (SMA) as our target region for model validation and take a spatial average over the SMA (see subfigure in Fig. 1d). The SMA include Seoul, the capital of South Korea, and two nearby provinces: Incheon and Gyeong-gi. Surface $PM_{2.5}$ observations over South Korea are available since 2015. Daily $PM_{2.5}$ observations over Seoul, however, are available since 2013, so daily $PM_{2.5}$ over the SMA prior to 2015 are obtained from daily $PM_{2.5}$ 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 $PM_{2.5}$ over Seoul, assuming that $PM_{2.5}/PM_{10}$ ratio over the SMA is the same as the ratio over Seoul.

2.3.2 China surface reanalysis

We utilize surface $PM_{2.5}$ reanalysis data over China recently developed by Kong et al. (2020). The dataset has 15 km×15 km spatial resolutions at hourly time scales for the period of

2013–2018. They assimilated surface observations at more than 1,000 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 $PM_{2.5}$ 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 $PM_{2.5}$ data are averaged over the North China Plain (NCP) and the YRD regions (see Fig. 1d), and daily $PM_{2.5}$ data over the two regions are used.

It should be noted that $PM_{2.5}$ 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 $PM_{10}/PM_{2.5}$ ratio is greater than or equal to 2.5 and observed daily PM_{10} is greater than or equal to 125 μg 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 (Supplementary). 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 $PM_{2.5}$

Figure 5 compares the spatial distribution of monthly mean $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ over China (e.g., the NCP and YRD) at the exception of a few regions of central China where $PM_{2.5}$ levels are largely overestimated (Henan, Chongqing, and southwestern Sichuan). In May, the model reproduces reasonably well $PM_{2.5}$ over the NCP, YRD, and central China except for Henan province. Over the dust regions of southern Mongolia and northern China (Inner Mongolia), the model tends to underestimate $PM_{2.5}$ (especially in May) likely due to underestimated dust aerosols. The lower $PM_{2.5}$ 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 in Supplementary). The large biases in the long-term averaged $PM_{2.5}$ seen over central China for both seasons imply that the emissions are likely overestimated in REASv3.1 and thus need to be revised for this region.

3.2 Temporal Variations—Trends, Interannual and Daily Variations

Figure 6 shows the monthly mean surface $PM_{2.5}$ concentrations simulated and observed over the SMA, NCP, and YRD for February and May 2013–2018. Both simulated and observed $PM_{2.5}$ concentrations show decreasing trends, and the control simulations capture reasonably well the trends and monthly mean values. The observed $PM_{2.5}$ over the SMA shows small decreasing trends in February (–0.21 μg m^{-3} $year^{-1}$) and slightly stronger decreasing trends in May (–2.3 μg m^{-3} $year^{-1}$). The trends in simulated $PM_{2.5}$ are –1.81 μg m^{-3} $year^{-1}$ in February and –2.68 μg m^{-3} $year^{-1}$ in May. The model is not able to capture the slightly elevated $PM_{2.5}$ 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, $PM_{2.5}$ in February over the NCP decreases at a rate of –9.6 μg m^{-3} $year^{-1}$ in the observations and –8.4 μg m^{-3} $year^{-1}$ in the control simulations. For February, the NCP $PM_{2.5}$ 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 $PM_{2.5}$ are given in Table 3. The systematic RMSE is based on the difference between linearly-fitted model 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 ±~25% (Table 3), the RMSE values are smaller than 30 μg 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 $PM_{2.5}$. We show that the interannual variations in monthly mean $PM_{2.5}$ due to those in meteorology is not negligible and can be up to 10.5 μg m^{-3} in February (the difference between 37.0 μg m^{-3} in 2014 and 26.5 μg m^{-3} in 2018 in the sensitivity simulations) and by 13.3 μg m^{-3} in May (the difference between 36.5 μg m^{-3} in 2013 and 23.2 μg m^{-3} in 2018) over the SMA with the fixed emissions. The monthly mean $PM_{2.5}$ is approximately 30 μg m^{-3} over SMA. This result indicates that about 35% (45%) of monthly mean $PM_{2.5}$ can be modulated by meteorology in winter (spring) at maximum. Similarly, the YRD reveals large interannual variations in $PM_{2.5}$ of 19.0 μg m^{-3} in February and of 12.0 μg 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 $PM_{2.5}$ due to meteorology of 10.6 μg m^{-3} in February and 6.5 μg 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, $PM_{2.5}$ 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 $PM_{2.5}$ over these regions can be substantially influenced by the long-range transport of $PM_{2.5}$ from the upwind regions. A similar result is found from a case study by (Kang et al., 2019), highlighting a large contribution of $PM_{2.5}$ 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 of the non-negligible role of meteorology in the interannual variations in $PM_{2.5}$, the decreasing trends of $PM_{2.5}$ are hardly explained only by meteorology (Fig. 6). In other words, the decreases in $PM_{2.5}$ are mostly due to the reductions in emissions especially for China. Similar result was found for the past when $PM_{2.5}$ concentration increased over eastern China during 1985–2005 (Yang et al. 2016). They showed that the increasing trends of $PM_{2.5}$ are largely influenced by anthropogenic emissions (10.5±6.2 μg m^{-3} $decade^{-1}$) than by meteorology (e.g., 1.8±1.5 μg m^{-3} $decade^{-1}$) during 1985–2005. These results underscore the effectiveness of emission controls and are consistent with many recent studies 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 HTAPv2) 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.

3.3 Potential Sources of Errors

In Fig. 6a, 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 ~3 μg m^{-3}. In the daily timeseries (Fig. 7m), 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

409 specific episodes are similarly found over the NCP and the YRD: the large biases in monthly mean
410 $PM_{2.5}$ are often due to the large errors in specific episodes (e.g., 2–4 and 15–19 February 2016, and
411 12–14 February 2017 over the NCP; and 16–17 February 2015, 15–19 February 2016, and 4–5
412 February 2017 over the YRD). Identifying and quantifying the sources of errors is difficult and not
413 the main goal of this study. However, here we propose some potential rationales for the sources of
414 errors. In general, the model bias errors show large daily variations; that is, the bias errors are
415 sporadically large in short-term episodes rather than are consistently present throughout the month.
416 In some cases, however, persistent biases throughout the month are found (e.g., February 2018 over
417 the SMA, May 2013 over the NCP, and May 2018 over the YRD). In these cases, the systematic
418 RMSEs are larger than the unsystematic RMSEs (Table 3). This means that there are considerable
419 systematic errors that can be corrected by linear scaling because the systematic RMSE is computed
420 as an RMSE of linear regressed model results against observations. The systematic errors in these
421 cases are likely originating from emissions because the scaling up of emissions acts to
422 monotonically and linearly increase $PM_{2.5}$ concentration throughout the month (Figs. 7 and 8). The
423 reductions in systematic RMSEs with the increased emissions (the base year of 2014) in February
424 2018 over the SMA and in May over the YRD support this assertion (Figs. 7p and 8r, and Table
425 S4 in Supplementary). Dry deposition can also be a source of systematic errors, but it is less likely
426 because we use the same dry deposition scheme throughout all years and the monthly means of
427 simulated $PM_{2.5}$ do not show a constant bias throughout years relative to the observed ones. On the
428 other hand, the larger unsystematic RMSE than systematic RMSE can be interpreted as larger
429 contributions from intermittent sources such as meteorology and wet deposition, which is the case
430 for most runs (Table 3). The large daily variation in bias errors indeed reflects the large
431 unsystematic errors. A systematic and in-depth research will be required in the future to identify
432 and quantify the sources of errors.

3.4 $PM_{2.5}$ Composition

The changes in $PM_{2.5}$ composition during 2013–2018 are shown in Fig. 9. Because there are few comprehensive observations of $PM_{2.5}$ composition over the study area, we only present the model simulation results. For the SMA, the trends of $PM_{2.5}$ 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 $PM_{2.5}$ concentrations decrease. Sulfate aerosols are also slightly higher in February 2015 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, T. 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 SO_2 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

Figure 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. 10a). 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. 8j). 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 all 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 agreements 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.

3.6 Anthropogenic Emission Rates versus Surface $PM_{2.5}$

In this subsection, the relationship between emission rates and surface $PM_{2.5}$ concentrations is explored to provide an insight into how much $PM_{2.5}$ changes in accordance with

481 changes in emissions. In the emission rates, primary PM emissions are only considered. That is,
482 the contribution of secondary aerosol formation is not considered because the secondary aerosol
483 formation is largely affected by levels of precursors, meteorological conditions (e.g., temperature,
484 relative humidity, and UV intensity), and other factors. It is noteworthy that the contributions and
485 associated uncertainties of SOA to $PM_{2.5}$ concentrations would be small in winter (~4–9 %), but
486 become larger in spring (~10–20 %) (Fig. 9).

487 As expected, the NCP clearly shows a linear relationship between emissions and surface
488 $PM_{2.5}$ concentrations in winter (February) (Fig. 11). The relationship between emissions and $PM_{2.5}$
489 is less clear over the YRD than over the NCP (Fig. 11). The YRD results for February 2013–2014
490 are off from the linear tendency embracing the results for the NCP and 2015–2018 YRD. Thus, the
491 results for February 2013–2014 YRD are not included in the linear regression. One potential reason
492 for this is the much stronger winds over the YRD in February 2013–2014 than in February 2015–
493 2018, which could facilitate ventilation and less accumulation of pollutants over the YRD during
494 2013–2014. The 1000 hPa mean wind speed in February over the YRD from the ERA-5 reanalysis
495 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,
496 and 1.0 m s^{-1} in 2018. It would be interesting to explore why $PM_{2.5}$ in February 2013–2014 over
497 the YRD shows different responses to emissions in a future study.

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

504 The relationship between emissions and $PM_{2.5}$ 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 more 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 less contribution of emissions to $PM_{2.5}$ in May than in February. For example, even if the monthly mean PM emission rate is the same as 0.2 $\mu g\ m^{-2}\ s^{-1}$, the monthly mean $PM_{2.5}$ would be 79.5 $\mu g\ m^{-3}$ in February and 54.2 $\mu g\ m^{-3}$ 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 presented the WRF-Chem modeling system that utilizes up-to-date data including updates in emissions during 2013–2018 to account for the recent trends in $PM_{2.5}$ in the winter-spring season. The WRF-Chem model shows reasonable performance in reproducing monthly means, trends, daily variations, and vertical profiles in $PM_{2.5}$ in the study area, even though some biases are found between observations and simulations. The recent decreasing trends in $PM_{2.5}$ over China are attributed to the reductions in emissions regulated since 2013. Therefore, it is strongly recommended that reasonably estimated trends in emissions should be used in air quality modeling for the recent years and the future over East Asia. The effects of emission reductions on the decreases in $PM_{2.5}$ are larger in the severely polluted upwind region (43% reduction in February and 41% in May during 2013–2018 based on the NCP observations) than the downwind region (21% in February and 33% in May from the YRD observations). The trends of $PM_{2.5}$ over the SMA are very week in winter, but are decreasing in spring at a rate of –2.3 $\mu g\ m^{-3}\ year^{-1}$ from the observations and –2.7 $\mu g\ m^{-3}\ year^{-1}$ from the simulations. Even though the decreasing trends in

$PM_{2.5}$ are mainly explained by the decreasing trends in emissions, meteorology is found to considerably modulate $PM_{2.5}$ concentrations and also its chemical composition, thus influencing the interannual variations in $PM_{2.5}$. The effects of meteorology are larger in the downwind regions (the SMA and YRD) than in the intense source region (the NCP), and larger in spring than in winter. With the fixed emissions throughout all years, monthly mean $PM_{2.5}$ can be altered by meteorology by up to 45% in spring and 35% in winter over the SMA, by ~27% over the YRD, and by ~11% over the NCP. The meteorology is also presumed to more influence the levels of secondary aerosols than those of primary aerosols as secondary aerosols are found to increase sometimes even their precursors' emissions decrease. The sources of model errors could be either by errors in emissions or in meteorology or by both. A consistent bias appearing throughout a month reflected by a large systematic RMSE is likely due to the errors in emissions. The sporadic large biases appearing in short-term episodes suggest that the misrepresentation in meteorology is likely responsible for the biases. For a judicious evaluation of emission controls, therefore, more efforts to minimize errors in meteorology should be dedicated.

The relationships between emission rates and $PM_{2.5}$ concentration presented in this study can be used to estimate emissions for other studies. For example, one can apply the relationships and estimate how much emissions are reduced under unusual and unexpected events such as the COVID-19 pandemic if historic $PM_{2.5}$ observations are available.

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786 **Table 1.** Model configuration for meteorology

Description/Physic scheme	Selection	Reference
Horizontal grid size (number of grids, nx×ny)	20 km (180×15)	
Number of vertical layers (top pressure)	40 (40 hPa)	
Initial/boundary conditions, updated intervals	ERA-5, 3-hour	(Hersbach et al., 2020)
Shortwave and Longwave radiation	RRTMG	(Iacono et al., 2008)
Boundary layer	MYNN	(Nakanishi and Niino, 2009, 2006)

Land surface	Noah	(Tewari et al., 2004)
Urban canopy model (UCM) and parameters used in the UCM	WRF-Single Layer UCM UCM parameters for Seoul Metropolitan Area	(Kusaka et al., 2001) (Ryu and Baik, 2013) for the UCM parameters
Microphysics	Morrison two moment	(Morrison et al., 2009)
Cumulus	Grell-Freitas	(Grell and Freitas, 2014)

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800 **Table 2.** Model configuration for gas and aerosols

Description	Selection	Reference
Gas-aerosol chemistry	MOZART-MOSAIC	(Hodzic et al., 2014; Knote et al., 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	(Ryu et al., 2019, 2018)

	shortwave scheme	
Dry deposition	(Wesely, 1989)	
Wet deposition	(Neu and Prather, 2012) for soluble gases (Wang et al., 2014) for below-cloud scavenging of aerosols	
Initial/boundary conditions, updated intervals	CAMS, 3-hour	(Inness et al., 2019)
Anthropogenic emissions	REASv3.1	(Kurokawa and Ohara, 2020)
Biogenic emissions	MEGANv2.04	(Guenther et al., 2012)
Biomass burning emissions	GFASv1.2	(Kaiser et al., 2012; Rémy et al., 2017)
Dust/sea salt emissions	GOCART	(Zhao et al., 2010)

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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 μg m^{-3}.

	SMA February			NCP February			YRD February			SMA May			NCP May			YRD May		
year	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	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