Application of WRF/Chem over East Asia:

Part II. Model Improvement and Sensitivity Simulations

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Abstract

To address the problems and limitations identified through a comprehensive evaluation in Part I paper, several modifications are made in model inputs, treatments, and configurations and sensitivity simulations with improved model inputs and treatments are performed in this Part II paper. The use of reinitialization of meteorological variables reduces the biases and increases the spatial correlations in simulated temperature at 2-m (T2), specific humidity at 2-m (Q2), wind speed at 10-m (WS10), and precipitation(Precip). The use of a revised surface drag parameterization further reduces the biases in simulated WS10. The adjustment of only the magnitudes of anthropogenic emissions in the surface layer does not help improve overall model performance, whereas the adjustment of both the magnitudes and vertical distributions of anthropogenic emissions shows moderate to large improvement in simulated surface concentrations and column mass abundances of species in terms of domain mean performance statistics, hourly and monthly mean concentrations, and vertical profiles of concentrations at individual sites. The revised and more advanced dust emission schemes can help improve PM

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predictions. Using revised upper boundary conditions for O₃ significantly improves the column O₃ abundances. Using a simple SOA formation module further improves the predictions of organic carbon and PM_{2.5}. The sensitivity simulation that combines all above model improvements greatly improves the overall model performance. For example, the sensitivity simulation gives the normalized mean biases (NMBs) of -6.1% to 23.8% for T2, 2.7-13.8% for Q2, 22.5-47.6% for WS10, and -9.1% to 15.6% for Precip, comparing to -9.8% to 75.6% for T2, 0.4-23.4% for Q2, 66.5-101.0% for WS10, and 11.4% to 92.7% for Precip from the original simulation without those improvements. It also gives the NMBs for surface predictions of -68.2% to -3.7% for SO₂, -73.8% to -20.6% for NO₂, -8.8% to 128.7% for O₃, -61.4% to -26.5% for PM_{2.5}, and -64.0% to 7.2% for PM₁₀, comparing to -84.2% to -44.5% for SO₂, -88.1% to -44.0% for NO₂, -11.0% to 160.3% for O₃, -63.9% to -25.2% for PM_{2.5}, -68.9% to 33.3% for PM₁₀ from the original simulation. The improved WRF/Chem is applied to estimate the impact of anthropogenic aerosols on regional climate and air quality in East Asia. Anthropogenic aerosols can increase cloud condensation nuclei, aerosol optical depth, cloud droplet number concentrations, cloud optical depth, and decrease surface net radiation, temperature at 2-m, wind speed at 10-m, planetary boundary layer height, and precipitation through various direct and indirect effects. These changes in turn lead to changes in chemical predictions in a variety of ways.

Key words: WRF/Chem, Model Improvement, Sensitivity Simulation, Aerosol Direct Effects, Aerosol Indirect Effects, East Asia

1. Introduction

In Part I paper, the Weather Research and Forecasting model coupled with chemistry (WRF/Chem) (Grell et al., 2005) is applied to East Asia. The performance of WRF/Chem is evaluated using observations from surface networks and satellites and also comparing to that of CMAQ based on a previous application of Wang et al. (2010a, b). Such evaluation and intercomparison help identify limitations and uncertainties in the model inputs and representations of atmospheric processes in WRF/Chem that warrant improvements. The meteorological evaluation indicates that the WRF/Chem free run with only grid nudging of wind vectors in the planetary boundary layer (PBL) cannot reproduce well the observations, leading to large biases in temperature at 2-m (T2), wind speed at 10-m (WS10), and daily precipitation (Precip). On the other hand, the four dimensional data assimilation (FDDA) with analysis nudging of T2 and Q2 above the PBL and also WS10 in the PBL used in MM5 can reproduce WS10 and Precip well and T2 relatively well. However, the use of nudging could squelch the simulated chemistry-meteorology feedback effects because the forcing exerted by the nudging technique may be stronger than the feedback effects. The re-initialization of meteorological variables such as T2, Q2, and WS10 with a re-initialization frequency of 1-4 days has been therefore used in the WRF/Chem applications that focus on the effects of chemistry-meteorology feedback and aerosol direct and indirect effects (e.g., Zhang et al., 2012a, b; Yahya et al., 2014; Hogrefe et al., 2014). Using an online-coupled CMAQ with only aerosol direct effects, Hogrefe et al. (2014) found that nudging has a small dampening effect on the strength of the simulated aerosol direct feedbacks only when nudging coefficients for the temperature, wind speed, and water vapor mixing ratio were reduced by a factor of six comparing the typical nudging

coefficients used in meteorological modeling (e.g., 3×10^{-4} used in Gilliam et al. (2012) and Wang et al. (2015)).

The evaluation of surface carbon monoxide (CO), sulfur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), and particulate matter (PM), column abundances of CO, SO₂, and NO₂, and aerosol optical depth (AOD) indicate possible underestimates of the emissions of those gases and primary PM species as well as inaccurate vertical allocations of their emissions. Several modeling studies over East Asia also reported the underestimates of emissions are a main cause for poor model performance (e.g., X.-H. Liu et al., 2010; X.-Y. Liu, 2015). The significant OC underpredictions at the Tsinghua (THU) site could not be fully explained by the missing of secondary organic aerosol (SOA) formation in the baseline simulations, which also indicates the underestimates in primary OC emissions. The evaluation of surface concentrations of PM_{2.5} and PM₁₀ indicates that the default Shaw (2008) dust emission module is not applicable to East Asia. The significant overpredictions in WS10 by WRF/Chem are mainly due to inaccurate representations of surface roughness that cannot resolve the characteristics of topography, which contribute to higher sea-salt concentrations. The baseline WRF/Chem simulation moderately overpredicts the column abundances of tropospheric ozone (O₃) in January 2005 and underpredicts them in July 2005, due likely to the incorrect representation of the seasonal variations of stratospheric O₃ intrusions to the upper troposphere, which is reflected in the upper boundary conditions (BCs) of O₃. Inaccurate upper BCs of O₃, in particular, a lack of seasonal variations in the upper BCs is identified to be a major factor contributing to the overpredictions of the column abundances of tropospheric O₃.

In this study, several inputs and model treatments identified from the model evaluation and intercomparison are first improved, sensitivity simulations with improved model inputs and treatments are then performed to demonstrate their impacts on model performance. The improved WRF/Chem is also applied to estimate the impacts of anthropogenic aerosols on air quality and climate through direct and indirect effects in East Asia. With improved model performance, the simulated effects of anthropogenic aerosols are expected to represent the aerosol-climate interactions in East Asia, which is one of the objectives for this study.

2. Model Improvement and Sensitivity Simulation Design

Based on the evaluations and analyses of the initial application of WRF/Chem and its intercomparison with MM5/CMAQ, several modifications are made for the model inputs and treatments. These include (1) the use of reinitialization of meteorological variables; (2) the adjustments on the anthropogenic emission rates and vertical distributions; (3) the modifications to BCs of O₃ to account for the impact of O₃ intrusions from stratosphere; (4) an update surface drag parameterization that corrects the large positive bias in simulated wind speed; (5) the use of the modified Shaw (2008) dust emission scheme and a new online dust emission scheme based on Zender et al. (2003) as implemented based on Wang et al. (2012); and (6) an addition of a simple treatment of SOA. These modifications are described below.

2.1. Improvements in Model Inputs

2.1.1 Re-initialization of meteorological variables

To accurately simulate meteorology and preserve the chemistry-meteorology feedback effects, T2, Q2, and WS10 are re-initialized daily using the National Centers for Environmental Predictions Final Analysis (NCEP-FNL) reanalysis data in the sensitivity simulations. This re-

initialization technique improves the initial conditions (ICs) of meteorological conditions and ensures that the simulation meteorology is not significantly deviated from the reanalysis data. It thus provides a compromise to allow the simulation of feedbacks while periodically constraining the simulation with re-initialized meteorological conditions based on observations.

2.1.2 Adjustment of Anthropogenic Emissions

No official emission inventories from China have been available for public use until recent years when the Multi-resolution Emission Inventory for China (MEIC) were developed and released by Tsinghua University, China. The MEIC emission inventories include emissions since 2006. The uncertainties of the MEIC emission inventory are estimated to be $\pm 12\%$ for SO_2 , $\pm 31\%$ for nitrogen oxides (NO_x), $\pm 68\%$ for non-methane volatile organic compounds (VOCs), $\pm 70\%$ for CO, $\pm 130\%$ for particles with diameter less than or equal to $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$), and $\pm 132\%$ for particles with diameter less than or equal to $10\ \mu\text{m}$ (PM_{10}) for China. Wang et al. (2015) indicated the uncertainties associated with the spatial allocation of the emissions in MEIC. Earlier emission inventories developed for Asia such as the emissions of Transport and Chemical Evolution over the Pacific (TRACE-P) of Streets et al. (2003) and the Intercontinental Chemical Transport Experiment (INTEX-B) of Zhang et al. (2009) may be subject to larger uncertainties than MEIC in terms of both magnitudes and spatial and temporal allocations. The emissions of Wang et al. (2010a) for 2005 are very similar to those based the INTEX-B emission inventory of Zhang et al. (2009); they are thus subjected to a similar degree of uncertainties as INTEX-B. X. Wang et al. (2010) kept the same emissions of TRACE-P for their WRF/Chem simulation of July 2001 but adjusted the diurnal variations and vertical distributions of emissions in East Asia for July 2001. They found that the model performance is highly sensitive to such

adjustments and the use of the emissions with more realistic diurnal variation and vertical distributions can improve the model performance substantially. X.-Y. Liu et al. (2015) adjusted the 2008 emissions over China for WRF/Chem applications according to the evaluation results from MM5-CMAQ of X.-H. Liu et al. (2010) and their evaluation indicated uncertainties in the vertical allocation of Asian emissions.

In this work, anthropogenic emissions are adjusted with two methods. The first method adjusts the magnitude of emissions using factors shown in Table S1. The second method adjusts the magnitude of emissions using factors shown in Table S2 and also the vertical distribution of emissions. These methods are described in details in the supplemental material.

2.1.3 Modification of Ozone Boundary Conditions

The original O₃ BCs are based on the averaged values extracted from the simulation results of the Goddard Earth Observing System Atmospheric Chemistry Transport Model (GEOS-Chem) model with vertical data interpolations with respect to pressure levels (as described in Wang et al., 2010a). The mixing ratios of O₃ in the BCs are low for the upper layers, resulting in an underestimation of tropospheric O₃ column, especially in July 2005 (see Figure 4 in Part I paper). In this work, O₃ vertical profiles are obtained from the Ozone Monitoring Instrument (OMI) level 2 data, with an averaging of vertical profiles along the boundary of the modeling domain. Such satellite-based vertical profiles account for the contribution of stratospheric O₃ intrusion to tropospheric O₃ in upper layers. These mean BCs for O₃ are applied to correct the biases in the original BCs. As shown in Figure S1, comparing to the original BCs, the modified BCs give much higher values and show greater seasonal variations for altitudes above 12 km. Comparisons of old and new BCs are described in the supplementary material.

2.2. Improvements in Model Treatments

2.2.1 Online Dust Emissions

Dust emissions due to wind blowing only occur on several types of surfaces such as desert and arid areas. The default Shaw (2008) scheme allows additional land types such as shrub-land, grassland, and savanna to generate dust as a function of wind speeds. However, this scheme was developed for application to Mexico, and thus may not be appropriate for East Asia (Zhang et al., 2012b). As shown in Part I paper, the use of Shaw (2008) generates significant amounts of dust particles that contribute to unrealistically high concentrations of $PM_{2.5}$ and PM_{10} , especially in the northern part of the domain. In this work, two methods are explored to examine the impacts of dust emissions on model performance. In the first method, following Zhang et al. (2012b), the surface types that allow dust generation in Shaw (2008) are modified to be mixed shrub land/grassland and barren or sparsely vegetated land, and a scaling factor with value of 0.05 is applied on the resulting dust emissions (referred to as modified Shaw (2008)). In the second method, a more robust dust emission scheme based on Zender et al. (2013) implemented by Wang et al. (2012) is applied, in which dust emission generation is based on surface erodibility and other factors such as wind speed, soil particle size, and soil moistures.

2.2.2 Modified Surface Drag Parameterization for Correction of Wind Speed Bias

WS10 is significantly overpredicted by WRF/Chem for all months as shown in Table 2 in Part I paper. The overpredictions in WS10 are likely caused by low surface drag due to the inappropriate representation of surface roughness because the detailed surface structure cannot be reproduced at a coarse grid resolution of 36-km. To correct this WS10 bias, following Mass

and Ovens (2005), the surface drag is increased by 1.5 times (which is applied to the friction velocity) when calculating wind speeds in the YSU PBL scheme.

2.2.3 Secondary Organic Aerosol Formation

The 8-bin option of MOSAIC coupled with CBM-Z in WRF/Chem v3.3.1 does not simulate SOA, which was identified as one of the reasons for underpredictions of OC at the THU site in Beijing shown in Figure 6 in Part I paper. In this work, a simple SOA module based on an aerosol yield approach is added in the CBM-Z and MOSAIC chemistry option used in this work. In this simple approach, SOA is generated from several gas precursors using aerosol yield coefficients derived from lab experiments of Odum et al. (1997), Lim and Ziemann (2005), Kroll et al. (2006), and Lee et al. (2006), as described in the supplementary material. The advantages and limitations of the aerosol yield approach along with other approaches have been discussed in several papers (e.g., Pun et al., 2002; Yu et al., 2004, 2007; Yu, 2005). While this is clearly not the best approach for simulating SOA, it does take the reactivity of the parent organic species and the availability of oxidants into account and has several advantages such as ease for implementation into 3-D models and computational efficiency; it has been therefore used in a number of 3-D models in the past and in this work.

2.3. Sensitivity Simulation Design

Sensitivity simulations are conducted to examine the improvements in the model performance due to the aforementioned modifications. Table 1 summarizes the differences in the model configuration between baseline and sensitivity simulations. The WRF/Chem baseline simulation in Part I paper (referred to as ORIGEMIS) uses continuous run for meteorology with only grid nudging for winds in the PBL, the original anthropogenic emission inputs as those used

in CMAQ simulations of Wang et al. (2010a, b), the original BCs for O₃, and default settings of schemes (i.e., the original PBL scheme, the dust emission scheme of Shaw (2008), and the original aerosol module MOSAIC that does not simulate SOA). Since the online-coupled WRF/Chem simulations are computationally more expensive than offline-coupled MM5/CMAQ, only a limited number of sensitivity simulations can be performed with limited computer resources. Four sensitivity simulations are carefully designed to demonstrate the impacts of the improvements in model inputs and treatments. In the first sensitivity simulation (referred to as ADJEMIS1), anthropogenic emissions are adjusted using the first adjustment method, the modified Shaw (2008) is used for dust emission scheme, and meteorological variables are reinitialized daily. It uses, however, the original BCs for O₃ and does not simulate SOA. Comparison of ADJEMIS1 with the baseline simulation, ORIGEMIS, allows the assessment of the impacts of reinitialization and modified anthropogenic and dust emissions on meteorological performance, and gases and PM performance, respectively. In the second sensitivity simulation (referred to as ADJEMIS2), anthropogenic emissions are adjusted using the second method. It also uses the new O₃ BCs, the daily reinitialization of meteorological variables, the wind speed bias correction method, the new dust emission scheme of Zender et al. (2003), and the simple SOA module. Comparison of ADJEMIS2 with ADJEMIS1 allows an assessment of the overall impacts of the new O₃ BCs, modified anthropogenic emissions using the second method and dust emissions using the Zender et al. (2003) on gases and PM species, and the simple SOA module on performance of gases (in particular column O₃ abundance), PM_{2.5} (in particular, OC), and PM₁₀. In order to examine the individual impacts of the corrected wind speed and the simple treatment of SOA, two one-month sensitivity simulations (i.e., the 3rd and 4th sensitivity

simulations) are performed using the same configuration of ADJEMIS2 but turning off the wind speed correction and SOA generation, respectively (WCoff and SOAoff, respectively). January 2005 is selected for the WCoff sensitivity simulation, since January has the largest overprediction in WS10 based on WRF/Chem initial applications (as shown in Table 2 in Part I paper). July 2005 is chosen for the SOAoff sensitivity simulation, since July has high emissions of biogenic VOCs (i.e., isoprene and terpene) that are important precursors of SOA.

To estimate the impact of anthropogenic aerosol direct and indirect effects, the 5th sensitivity simulation, PMoff, is performed by turning off anthropogenic PM emissions, and secondary PM formation through aerosol and cloud chemistry, but using the same model configuration and ICs/BCs as the simulation ADJEMIS2 that has the best overall model performance. The differences between ADJEMIS2 and PMoff represent the direct and indirect effects of anthropogenic aerosols, following the method used in Zhang et al. (2010) (which focused on the effects of both anthropogenic and natural aerosols). The PMoff simulation is performed for all the four months to estimate the seasonal variations of such effects.

3. Evaluation and Intercomparison of Simulation Results

3.1 Evaluation of Sensitivity Simulations and Comparison with the Baseline Simulations

Table 2 summarizes the performance statistics of surface meteorological variables against NCDC data for the baseline simulation (i.e., ORIGEMIS) and the four sensitivity simulations (i.e., ADJEMIS1, ADJEMIS2, WCoff, and SOAoff). Comparing to ORIGEMIS, ADJEMIS1 significantly reduces the biases in simulated T2 and Precip, particularly in January, April, and July for T2 and in January and October for Precip. Comparing to observations from the National Climatic Data Center (NCDC), the mean biases (MBs) in T2 are reduced from 1 to 0.4 °C, -1.4

to -0.8°C , -1.5 to -0.8°C , and 0.2 to 0.1°C and the normalized mean biases (NMBs) change from 75.6% to 33.1%, -9.8% to -6%, -6.0% to -3.3%, and 1.5% to -0.8% in January, April, July, and October, respectively. The NMBs change from 92.7% to 24.6%, 13.6% to 10.4%, 11.4% to -4.6%, and 89.8% to 1.1% for Precip in January, April, July, and October, respectively.

ADJEMIS1 and ADJEMIS2 also show stronger correlations with observations than those predicted from the base simulations. For example, the values of correlation coefficient, R , for Precip change from 0.1-0.2 in ORIGEMIS to 0.3 in ADJEMIS1 and to 0.3-0.4 in ADJEMIS2. The MBs and NMBs of Precip from ADJEMIS1 are even smaller than those of MM5 in April, July, and October, and comparable to that of MM5 in January (see MM5 performance statistics in Table 2, Part I paper). This is expected because of the use of reinitialization in ADJEMIS1 that ensures a consistency between the simulated meteorology and the NCEP reanalysis data. Figure 1 compares the Tropical Rainfall Measuring Mission (TRMM)-derived and simulated spatial distributions of Precip. The spatial distributions of Precip simulated by ADJEMIS1 and ADJEMIS2 are more comparable to the satellite-derived Precip over northeastern and southern China, North and South Korea, Japan, and western Pacific in all months. The reinitialization used in both ADJEMIS1 and ADJEMIS2 also improves the performance of Q2 in January and October (despite slightly worsening that in April and July) and WS10 in all months. However, the MBs and NMBs in WS10 simulated by ADJEMIS1 remain large, ranging from 1.5 to 2.6 m s^{-1} and from 56.4 to 84.3%, respectively, in the four months. Such large NMBs in WS10 are greatly reduced to 22.5-47.6% in ADJEMIS2 because of the use of the modified surface drag parameterization for wind speed bias correction. The comparison of WCoff and ADJEMIS2 in January shows the net impact of such a bias correction, reducing the MB of 2.6 to 1.5 m s^{-1} and

the NMB of 84.1% to 47.6%. Among the three simulations, ADJEMIS2 has the best overall meteorological performance. It either outperforms or performs similarly to MM5.

Table 3 summarizes the performance statistics of chemical predictions against surface and satellite data. The evaluation includes the surface concentrations of CO, NO₂, NO, SO₂, O₃, PM_{2.5}, and PM₁₀ against the surface observations from mainland China, Hong Kong, Taiwan, and Japan, and the column abundances of CO, NO₂, SO₂, O₃ and AOD against satellite-derived observations. ADJEMIS1 adjusts anthropogenic emissions using the method 1 by increasing the emissions of all gaseous species (except for NH₃) and PM species in all emission layers by 30% or 50% as shown in Table S1. As a consequence, the degree of underpredictions of surface concentrations of CO, NO, NO₂, and SO₂ at all sites is reduced (except for CO in Hong Kong in April). However, because of increased emissions at all emission levels, the column abundances of CO, NO₂, and SO₂ are moderately to significantly overpredicted in some months. The column abundances of CO in January and October change from slight to moderate overpredictions. The column abundances of NO₂ change from underpredictions to overpredictions with NMBs of 3.7% to 42.9% (comparing to -30.6% to -0.01% from ORIGEMIS) and those of SO₂ change from underpredictions to overpredictions or from moderate to significant overpredictions with NMBs of 23.3-172.6 % for SO₂ (comparing to -40.0% to 26.1% from ORIGEMIS). As shown in Figure S2, the simulated column abundances of CO in January and October from ADJEMIS1 give large positive biases over southern China comparing to the MOPPIT derived CO column. As shown in Figure S3, ADJEMIS1 moderately overpredicts the column abundances of NO₂ in July and October in eastern China. As shown in Figure S4, ADJEMIS1 also overpredicts the column abundances of SO₂, in particular, in July and October in eastern China with NMBs of

132.2% and 172.6%, respectively. These results indicate that the adjustment factors of anthropogenic emissions used in ADJEMIS1 may not be appropriate in terms of magnitudes despite some improvements for surface mixing ratios.

ADJEMIS2 adjusts anthropogenic emissions using the method 2 by increasing the emissions of SO₂, NH₃, and PM species at the surface level and CO and NO_x at higher levels by 30% or 300% as shown in Table S2, and also by modifying the vertical profiles of the emissions. As shown in Table 3, the performance statistics of simulated mixing ratios and column abundances of NO, NO₂, and SO₂ against surface and satellite observations show moderate to large improvement in ADJEMIS2 compared to ORIGEMIS. For example, for the four month simulations, the NMBs of surface NO and NO₂ concentrations range from -86.3% to -52.7% and from -51.4% to -20.6% compared to the original -96.8% to -92.2% and -75.5% to -61.7% in Taiwan, respectively. The NMBs of surface SO₂ concentrations range from -57.5% to -48.1% compared to the original -80.4% to -71.6% in China, from -57.9% to -3.7% compared to the original -84.2% to -59.1% in Hong Kong, and from -39.0% to -6.3% compared to the original -65.5% to -44.5% in Taiwan. For surface CO concentrations, ADJEMIS1 and ADJEMIS2 give similar performance, both are slightly better than the original ORIGEMIS in Hong Kong in all months except for April and in Taiwan and Japan for all months. For surface O₃ concentrations, comparing to ORIGEMIS, ADJEMIS1 shows better performance in Taiwan for January and October and in Japan for January and April but worse performance in Taiwan and Japan for other months and in Hong Kong in all months. By contrast, ADJEMIS2 significantly improves performance of surface O₃ concentrations at all sites in all months except for Japan in April and July. For example, the NMBs are -8.8% to 32.1% comparing to the original 13.6-83.8% in

Taiwan for all months, and 4.2-12.8% comparing to the original 25.8-33.3% in Japan for January and October. During April and July, the NMBs of surface O₃ concentrations in Japan are slightly worse than those from ORIGEMIS (e.g., -15.7% vs. -11.0% in April, and 25.3% vs. 16.3% in July) but still within the acceptable range.

Comparing to ADJEMIS1, ADJEMIS2 gives much less overpredictions for column abundances of CO, NO₂, and SO₂ in all months (except for column NO₂ in July when ADJEMIS2 gives slightly higher overpredictions than ADJEMIS1), in particular, it shows substantial improvement for the performance of column SO₂ comparing to both ADJEMIS1 and ORIGEMIS. For example, the NMBs for column SO₂ change from -17.2% to 89.5% in ADJEMIS2 comparing to the original -40.0% to 26.1% in ORIGEMIS, and 23.3% to 172.6% ADJEMIS1. The statistics for column gases are overall within the acceptable ranges of performance reported in the literature, especially considering the relatively poor data quality of SO₂ satellite observations which may contribute to some uncertainties in the model performance. As shown in Figures S2-S4, the simulated column abundances of CO, NO₂, and SO₂ from ADJEMIS2 also agree better with the satellite observations than ADJEMIS1 in terms of the spatial distributions, magnitudes, and correlations for all the months.

Figure 2 compares observed and simulated surface concentrations of CO, NO, NO₂, SO₂, O₃, and PM₁₀ in Hong Kong. Comparing to ORIGEMIS, ADJEMIS1 and ADJEMIS2 show moderate to large improvement in hourly concentrations of CO, NO, NO₂, and SO₂. ADJEMIS2 outperforms ADJEMIS1 for NO, NO₂, and SO₂, particularly in April and July. Figure 3 compares the simulated vertical profiles of SO₂ in the Henan province in northern China from all simulations to that from the aircraft measurement in April 2008 reported by He et al.

(2012). While both ORIGEMIS and ADJEMIS1 show a peak mixing ratio of SO₂ at altitudes of 200-250 meters above surface, the observations show a nearly monotonic decrease with height with the highest mixing ratio of SO₂ occurring at 100 meters. He et al. (2012) found similar mismatching of the simulated vertical profiles with those observed and speculated the problems in simulating vertical movement and transport. Such a monotonically-decreasing distribution with height is also found in the aircraft measurement of both NO₂ and SO₂ by Xue et al. (2010) in Changchun, Lanzhou, Jinan, and Wuxi, located in northeastern, northwestern, central, and eastern China, respectively. These aircraft measurements indeed indicated a need to adjust the vertical distributions of SO₂ and NO₂ emissions used in the model baseline simulations as described in the method 2 for emission adjustments. The total emissions of SO₂ and NO₂ are therefore redistributed with higher percentages of SO₂ and NO₂ emissions at the surface layer than those in the original emissions. Figure 3 shows a decrease of SO₂ mixing ratios with height monotonically by ADJEMIS2, consistent with observed vertical profiles of SO₂.

ADJEMIS1 significantly increases the overpredictions in the surface mixing ratios of O₃ at nearly all sites due to increased emissions of O₃ precursors, including NO_x, VOCs, and CO, and the fact that the impacts of increased NO_x emissions on O₃ during daytime dominate over the impacts of the titration of O₃ by NO_x at night. The performance of column abundances of O₃ remains similarly as ADJEMIS1 uses the original BCs for O₃. In ADJEMIS 2, VOCs emissions remain unchanged throughout all emission layers, the emissions of CO remain unchanged in surface layer but are increased in non-surface emission layers in April and July, and the emissions of NO_x are increased in the surface layer in all months because of increases in total emissions of NO_x and their re-distribution vertically. The more appropriate vertical allocations

of NO_x emissions used in ADJEMIS2 lead to much less overpredictions of surface O₃ due to an enhanced titration by more surface NO_x at night than those in ADJEMIS1 and ORIGEMIS, coupled with lower mixing ratios in the BCs of O₃ in the surface layer (see Figure S1), resulting in a significant improvement in surface O₃ performance by ADJEMIS2. For example, the NMBs of O₃ range from 26.8% to 128.7% comparing to the original 88.0% to 160.3% in Hong Kong and from -8.8% to 32.1% comparing to the original 13.6% to 83.8% in Taiwan. Figure 2 shows the improved hourly concentrations of O₃ in Hong Kong in all the four months. With the modified O₃ BCs, the column abundances of tropospheric O₃ are also improved, with NMBs from -17.0% to 28.5% comparing to the original -32.3% to 45.4% against TOMS data, and NMBs from -1.1% to 17.1% comparing to the original -19.4% to 27.2% against OMI data. Figure 4 compares observed and simulated O₃ concentrations in six major cities in Japan for the four months. Among the three simulations, ADJEMIS2 performs the best in January, July, and October, and ADJEMIS1 performs the best in April. Comparisons of spatial distributions of tropospheric O₃ column in Figure 5 also demonstrate the improvement of the simulated tropospheric O₃ column against observed O₃ column in all the four months due to the change in O₃ BCs in ADJEMIS2, which gives a better overall agreement with satellite-derived column O₃ in terms of both magnitudes and spatial correlations among the three WRF/Chem simulations.

Comparing to ORIGEMIS, the adjustment of surface emissions of PM species and precursors of secondary PM such as SO₂, NO_x, and VOCs in ADJEMIS1 increases the surface concentrations of PM species including SO₄²⁻, NO₃⁻, ammonium (NH₄⁺), EC, and OC (Figures not shown). However, the use of modified Shaw (2008) in ADJEMIS1 significantly reduces the concentrations of OIN (Figures not shown), leading to a net decrease in surface PM_{2.5}

concentrations in all areas except for an area in eastern China and western Sichuan province where anthropogenic emissions dominate $PM_{2.5}$ concentrations (see Figure S5). The modification of Shaw (2008) in ADJEMIS1 allows dust emissions over deserted areas located in the northwestern China (rather than the northern and northeastern China), making dust emissions more consistent with observations of dust storms in China.

For PM_{10} , ADJEMIS1 shows a large decrease in PM_{10} concentrations in the eastern part of the domain (in particular, eastern and northern China, North and South Korea, and Japan), but an increase in northwestern China where the major dust sources are located (see Figure S6). As a result of reduced $PM_{2.5}$ and PM_{10} concentrations, the performance of surface PM concentrations becomes worse at most sites in most months. By contrast, the adjustment of anthropogenic emissions of primary PM and the precursors of secondary PM as well as the use of the dust emission scheme of Zender et al. (2003) in ADJEMIS2 lead to large improvement in PM predictions comparing to results from both ORIGEMIS and ADJEMIS1. For example, the NMBs of PM_{10} range from -19.7% to 4.6% comparing to the original -37.8% to -2.9% in mainland China and from -30.4% to 7.2% comparing to the original -47.4% to -6.9% in Hong Kong. The values of R range from -0.2 to 0.4 comparing to the original 0.1 to 0.3 in China and from 0.4 to 0.8 comparing to the original 0.1 to 0.7 in Hong Kong. Similarly, the performance of $PM_{2.5}$ in Taiwan is improved in ADJEMIS2 relative to ORIGEMIS for three out of four months, with the NMBs ranging from -56.8%, -37.2%, and -26.5% comparing to the original -61.0%, -41.1%, and -63.9% for January, April, and July, respectively. As shown in Figure 2, ORIGEMIS fails to reproduce observed hourly concentrations of PM_{10} in Hong Kong in terms of magnitudes and diurnal variations, ADJEMIS1 reduces underpredictions in April and July, but increases

underpredictions in many days in January and October. For comparison, ADJEMIS2 shows a much improved hourly variations of PM_{10} concentrations than ORIGEMIS and ADJEMIS1. Figure 4 compares observed and simulated PM_{10} concentrations in six major cities in Japan. Among the three simulations, ADJEMIS2 gives the best performance in April, July, and October. It performs the best in Sapporo, Tokyo, Nagoya, and Osaka but worse than ORIGEMIS in Nagasaki and Tsukuba in January. Figure 6 compares observed and simulated concentrations of $PM_{2.5}$ and its composition at two sites in Beijing, China: Tsinghua University (THU) which is a suburban site, and Miyun which is a rural site. At each site, two sets of observations are available, the first set contains daily $PM_{2.5}$ concentrations at THU (see Figure 6a), and the second set contains weekly mean concentrations of $PM_{2.5}$ and its composition at THU and Miyun (see Figures 6a and 6b), which are used to calculate monthly mean concentrations at both sites. At THU, the observed daily $PM_{2.5}$ concentrations are only available during April 21-30, July, and October. In April, ORIGEMIS gives the best agreement among the three simulations. In July, $PM_{2.5}$ concentrations are significantly underpredicted by ORIGEMIS, overpredicted by ADJEMIS2, but well predicted by ADJEMIS 1. In October, ORIGEMIS underpredicts most peak concentrations but overpredicts minimal concentrations, particularly during October 14-15, ADJEMIS1 and ADJEMIS2 give similar results, with overall better agreement for most peak and minimal values by ADJEMIS2. In January, ADJEMIS1 and ADJEMIS2 predict much higher daily $PM_{2.5}$ concentrations than ORIGEMIS at THU. Although observed daily $PM_{2.5}$ concentrations are not available, the comparison of the monthly-mean observed and simulated $PM_{2.5}$ concentrations shows that ADJEMIS2 gives the best agreement to the observations among the three simulations. ADJEMIS2 also performs the best in July and

October for $\text{PM}_{2.5}$, whereas ORIGEMIS performs the best in April for monthly mean $\text{PM}_{2.5}$ concentrations. ADJEMIS1 and ADJEMIS2 predict better SO_4^{2-} , NO_3^- , and NH_4^+ than ORIGEMIS in January, April, and October, the opposite is true for July. For OC, ADJEMIS2 gives the best agreement with observations among the three simulations in all months. For EC and OIN, ORIGEMIS performs the best for all the months. As shown in Figure 6b, at Miyun, ORIGEMIS fails to reproduce the observed weekly variation of $\text{PM}_{2.5}$, ADJEMIS1 gives the best agreement in January, July, and October whereas ADJEMIS2 gives the best agreement in April. For monthly mean $\text{PM}_{2.5}$ concentrations, ADJEMIS1 gives the best agreement in January, July, and October whereas ORIGEMIS gives the best agreement in April but it cannot reproduce the weekly variations. Similar to THU, ADJEMIS1 and ADJEMIS2 predict better SO_4^{2-} and NH_4^+ than ORIGEMIS in all the four months, NO_3^- in January and April, whereas ORIGEMIS gives better NO_3^- in July and October. These site-specific results indicate that accurately simulating temporal variations (i.e., diurnal, weekly) at individual sites are challenging, rendering a need for the model to better capture the characteristics of in-situ $\text{PM}_{2.5}$ and its composition at small temporal scales.

ADJEMIS2 gives better OC predictions than ADJEMIS1 and ORIGEMIS for two main reasons. First, it has higher primary OC emissions in the surface layer. Second, it includes a simple SOA formation. A sensitivity simulation (i.e., SOAoff) is performed in July to estimate the impact of the SOA formation on total OC concentrations and performance. Note that SOAoff uses the same configurations as ADJEMIS2, the differences between the two simulations can be attributed to the inclusion of the SOA formation module. Figures 6a and 6b compare simulated concentrations of $\text{PM}_{2.5}$ and its composition by SOAoff with observations

and results from other simulations at THU and Miyun in July. Comparing to SOAoff, ADJEMIS2 gives an OC concentration that is higher by $5 \mu\text{g m}^{-3}$ at THU, because of the inclusion of a simple SOA formation module. Comparing to ADJEMIS1, ADJEMIS2 gives an OC concentration that is higher by $12.1 \mu\text{g m}^{-3}$ at THU, the simple SOA module contributes to 41% of OC increase, and higher primary OC emissions contribute to the remaining differences. SOAoff gives slightly lower concentrations of secondary inorganic aerosols and thus $\text{PM}_{2.5}$ at both THU and Miyun sites because lower concentrations of OH and HO_2 radicals are available for oxidation of SO_2 and NO_x to form SO_4^{2-} and NO_3^- , respectively. As shown in Figure S7, comparing to SOAoff that turns off the simple SOA module, the simple SOA formation module used in ADJEMIS2 increases the domain mean concentrations of OC and $\text{PM}_{2.5}$ by $1.5 \mu\text{g m}^{-3}$ and by up to $10\sim 20 \mu\text{g m}^{-3}$ in the VOCs source regions in central and eastern China in July. It also affects the gas-phase chemistry to some extent. For example, it reduces the mixing ratios of VOCs such as toluene and isoprene domainwide by converting their condensable products to SOA, which slightly increases the mixing ratios of NO_x because less OH and HO_2 radicals are available for oxidation of NO_x . The impact of reduced VOCs dominates over the increased NO_x , leading to decreases in the O_3 mixing ratios for most regions by 0-1.5 ppb, except for the southwestern China, and northern Burma, Thailand, Laos, and Vietnam where O_3 mixing ratio increases due likely to the pristine and biogenic VOCs rich atmosphere, making O_3 formation in those regions NO_x -limited.

ADJEMIS1 increases the secondary $\text{PM}_{2.5}$ concentrations over highly-polluted regions but decreases OIN throughout the atmosphere, which results in a moderate improvement in AOD in July, but worse agreement in other months (i.e., slightly more overpredictions in AOD in

October and worse underpredictions in January and April) in terms of both performance statistics and spatial correlation (see Table 3 and Figure 7). For comparison, ADJEMIS2 performs much better in all months except for October. The NMBs of AOD are -49.1% to 11.7% in ADJEMIS1, comparing to NMBs of -38.7% to 5.6% in ORIGEMIS. The correlations coefficients of AOD against satellite observations increase from 0.53, 0.51, 0.52, and 0.57 in ORIGEMIS to 0.61, 0.67, 0.56 and 0.60 in ADJEMIS1 for January, April, July, and October 2005, respectively. Comparing to ORIGEMIS, the NMBs in AOD by ADJEMIS2 are reduced for January, April, and July 2005 with the NMBs of -18.9% to 19.6%, (despite a greater overprediction in October, with an NMB of 56.1%) (see Table 3) and R values of simulated AOD against observations are improved from 0.5-0.6 to 0.6-0.7. Figure 7 shows improvement in AOD by ADJEMIS2 in terms of spatial correlations in January, April, and July 2005.

Simulated radiative and cloud variables respond to changes in meteorological and chemical predictions in ADJEMIS1 and ADJEMIS2. Figures S8 and S9 compare satellite-derived downward shortwave radiation (GSW) and downward longwave radiation (GLW), respectively, with those simulated by WRF/Chem in the four months. ORIGEMIS underpredicts GSW over most of the domain, particularly over western and northeastern China, North and South Korea, Japan, and western Pacific, leading to underpredictions in domain mean GSW in all the four months. ADJEMIS1 and ADJEMIS2 give similar GSW, both show higher GSW over most of the domain, especially over the aforementioned regions, leading to better agreement in terms of both domain mean values and spatial correlations in the four months. For GLW, ORIGEMIS simulates it well in terms of domain mean values and spatial correlations over land areas but it overpredicts GLW over oceanic areas such as western Pacific in all the four months.

Such overpredictions over oceanic areas are greatly reduced in ADJEMIS1 and ADIEMIS2. Figures S10 and S11 compare satellite-derived cloud liquid water path (LWP) and cloud fraction (CF), respectively, with those simulated by WRF/Chem in the four months. For LWP, ORIGEMIS shows large overpredictions in July, but moderate to large underpredictions in other months. Comparing to ORIGEMIS, ADJEMIS1 and ADIEMIS2 reduce the overpredictions of LWP in July and the underpredictions of LWP in January, and they also improve the spatial correlations of LWP in January, July, and October. As shown in Figure S11, ORIGEMIS underpredicts domain mean CF in all the four months. Such underpredictions occur in the entire domain in January, April, and October, but only in northern and central China in July during which large overpredictions actually occur in eastern and southwestern parts of the domain. ADJEMIS1 and ADIEMIS2 greatly reduce such overpredictions in July and improve the spatial correlations for all the four months. They also reduce the overpredictions domain mean in January, April, and October, although it increases the underpredictions in the domain mean in July.

3.2 Comparison with CMAQ Results

Comparing to CMAQ performance described in Part I paper, ADIEMIS2 performs much better for surface concentrations of CO, NO, NO₂, and SO₂ in all months and PM₁₀ in all months except for January in Hong Kong, the surface concentrations of CO, NO, NO₂, O₃, and PM_{2.5} in all months and those of SO₂ and PM_{2.5} in all months except for January in Taiwan, the surface concentrations of CO, NO, NO₂, and O₃ in all months and those of PM₁₀ in all months except for January, and those of SO₂ in July and October in Japan, and the surface concentrations of SO₂ in all months except for April and those of PM₁₀ in April and July in mainland China. ADIEMIS2

performs better in July and similarly in January for column CO, better in January and April but worse in July and October for column NO₂, better in January and April but worse in July and October for NO₂, much better in January and July for column SO₂ and column O₃ against TOMS and OMI data, much better in January, April, and July but much worse in October for AOD.

Part I paper identifies that the differences in dry and wet deposition treatments between CMAQ and WRF/Chem may be responsible for some of the differences in simulated chemical concentrations. To further examine such differences, WRF/Chem is modified to write out dry and wet deposition fluxes of relevant species in the ADJEMIS2 simulation. The simulated wet deposition fluxes and chemical concentrations of SO₄²⁻, NO₃⁻, and NH₄⁺ in July are compared with the observations from the Acid Deposition Monitoring Network in East Asia Data (EANET). Figure 8 compares observed and simulated concentrations of SO₄²⁻, NO₃⁻, and NH₄⁺ in July at the 27 EANET sites. Among the 18 sites where the concentration observations are available, WRF/Chem significantly underpredicts the concentrations of SO₄²⁻ at all sites except for Terelj, Primarskaya, and Chiang Mai, and overpredicts those of NO₃⁻ at most sites except for Rishiri, Tappi, Hedo, Ulaanbaatar, Terelj, Chiang Mai, Hanoi, and Hoa Binh. The underpredictions and overpredictions compensate each other, leading to an overall good agreement for NH₄⁺ at most sites with exceptions at Hedo, Ljira, and Cheju when large underpredictions in NH₄⁺ occur. The underpredictions in the concentrations of SO₄²⁻ may be caused by underestimates of SO₂ emissions (which is supported by underpredictions in the surface SO₂ concentrations shown in Table 3), overpredictions of SO₂ dry deposition, and overpredictions in WS10. The overpredictions of NO₃⁻ may be caused by the overpredictions of HNO₃, the inaccurate partitioning of total nitrate in the gas-phase and particulate phase, as well

as the overpredictions in Precip and underpredictions in T2 (note that both meteorological conditions favor the formation of NO_3^-). Comparing to WRF/Chem, CMAQ gives higher concentrations of SO_4^{2-} and better agreement with observations at most sites although the large underpredictions remain. CMAQ gives much lower concentrations of NO_3^- and better agreement with observations at most sites than WRF/Chem. The two models give similar performance for NH_4^+ at most sites.

Figure 9 compares observed and simulated wet deposition of SO_4^{2-} , NO_3^- , and NH_4^+ at the 27 EANET sites in July. Different from the concentration predictions, the wet deposition fluxes of SO_4^{2-} , NO_3^- , and NH_4^+ at all sites are all underpredicted by WRF/Chem. The largest underpredictions occur at Kanghwa in China, Imsil in South Korea, Yushara and Oki in Japan, and Hanoi and Hoa Binh in Vietnam. Precip at Kanghwa, Imsil, Yushara, and Oki are largely underpredicted (see Figure 1), which may contribute in part to the underpredictions in the wet deposition flux of SO_4^{2-} (another reason is the underpredictions in SO_4^{2-} concentrations as shown in Figure 8) and to a larger extent to the underpredictions in the wet deposition flux of NO_3^- and NH_4^+ because the concentrations of NO_3^- are indeed overpredicted and those of NH_4^+ are either overpredicted or slightly underpredicted. Precip at Hanoi and Hoa Binh are overpredicted (see Figure 2 in Part I paper), the underpredictions of the concentrations of SO_4^{2-} , NO_3^- , and NH_4^+ at the two sites are the major contributor to the underpredictions of their wet deposition fluxes. Comparing to WRF/Chem, CMAQ gives higher wet deposition fluxes of SO_4^{2-} at most sites and those of NH_4^+ at some sites, which brings the simulated results into better agreement with observations (except for NH_4^+ at Haplo, Cheju, and Banryu). CMAQ gives poorer agreement than WRF/Chem for the wet deposition fluxes of NO_3^- at most sites. Further, large

underpredictions in the wet deposition fluxes of SO_4^{2-} , NO_3^- , and NH_4^+ remain for CMAQ predictions. Figure S12 compares the spatial distributions of wet deposition fluxes simulated by the two models. They differ significantly in terms of spatial distributions, magnitudes, and domain mean for wet deposition fluxes of SO_4^{2-} . Differences also exist in the simulated spatial distributions of wet deposition fluxes of NO_3^- and NH_4^+ by both models, but the magnitudes and domain mean values are more similar than those for SO_4^{2-} . Such differences can be attributed to different precursor concentrations of those PM species, Precip predictions, and wet deposition parameterizations. In addition to large differences in simulated wet deposition fluxes, the two models also give different dry deposition predictions, as shown in Figure S13. CMAQ gives lower dry deposition fluxes of SO_2 and O_3 but higher values for NO_2 and NH_3 . Such differences are attributed to different gas-phase chemistry and dry deposition treatments. As described in Part I paper, the differences in dry deposition fluxes explain in part the differences in the simulated concentrations of chemical species such as SO_2 and O_3 .

4. Effects of anthropogenic aerosols on regional climate and air quality

The meteorological and chemical evaluation shows that ADJEMIS2 gives the best overall performance. This simulation is therefore used as the benchmark to examine the effects of anthropogenic aerosols in East Asia. A sensitivity simulation (i.e., PMoff) using WRF/Chem with the improved model inputs and treatments but turning off primary PM emissions and secondary PM formation is performed and comparing to ADJEMIS2. The differences between ADJEMIS and PMoff are caused by the direct and indirect effects of anthropogenic aerosols. Figures 10 and S14 show the absolute differences of PM_{10} concentrations, cloud condensation nuclei at a supersaturation of 0.5% ($\text{CCN}_{0.5}$), AOD, cloud droplet number concentration

(CDNC), cloud optical depth (COT), surface net shortwave radiation (SNSR), T2, total precipitation, planetary boundary layer height (PBLH), WS10, and surface mixing ratios of max 8-hr O₃ and CO for the four months between the ADJEMIS2 and PMoff. Anthropogenic aerosols contribute significantly to the mass (and number) concentrations of PM in East Asia in all months in 2005, with the highest in January, followed by October, April, and July. Due to the increase of anthropogenic aerosols, column CCN_{0.5} increases over the whole domain by $2.4\text{--}3.3 \times 10^9 \text{ cm}^{-2}$ (or by 59.7-70.3%) and AOD increases by about 0.15 to 0.30 (or by 162.9-307.3%). The largest increases in AOD occur in eastern China in all months and in both eastern and northeastern China in July and October. The domain mean AOD is the highest in July (because of the highest column PM concentrations in July), followed by October, April, and January. By acting as CCN, anthropogenic aerosols can increase column CDNC by 1.5×10^6 to $2.4 \times 10^6 \text{ cm}^{-2}$ (or from 143.8% to 183.3%), COT by 1.7 to 3.0 (or by 47.6-63.3%) during the four months. Large increases in CDNC and COT occur in the same areas where AOD increases, and spread eastward in all months, particularly in January, April, and October. Domain mean CDNC and COT are the highest in July, followed by October, January, and April. Through direct and indirect effects, anthropogenic aerosols can reduce SNSR by up to 49.4 W m^{-2} in January, up to 8.7 W m^{-2} in April, up to 50.5 W m^{-2} in July, and up to 47.6 W m^{-2} in October (or by up to 18.7-38.6%), with the domain mean decreases of 7.1, 9.8, 11.7, and 7.8 W m^{-2} , respectively. The areas with the largest decreases in SNSR correspond to areas where AOD is high. As direct responses to the reductions in SNSR and latent heat, T2 reduces by up to $1.1 \text{ }^\circ\text{C}$ in January, $0.55 \text{ }^\circ\text{C}$ in April, $0.53 \text{ }^\circ\text{C}$ in July, and $0.86 \text{ }^\circ\text{C}$ in October (or by factors of 2951, 630, 392, and 1179, respectively), with the domain mean decreases of 0.05, 0.05, 0.07, and $0.07 \text{ }^\circ\text{C}$, respectively (or

by 2.9%, 0.6%, 0.4%, and 1.2%, respectively). As a result of cooling at the surface caused by reduced SNSR and warming in the PBL caused by aerosol absorption, PBLH decreases by up to 84.3-122.1 m (or by up to 13.0-28.2%), with the domain mean decreases of 6.2-13.1 m, indicating an increased atmospheric stability that can further worsen the air pollution in East Asia. Anthropogenic aerosols can reduce WS10 by up to 0.3-0.5 m s⁻¹ (or by up to 56.8-88.6%). Slower wind speeds over source regions may prevent ventilation of pollutants, worsening air pollution. Anthropogenic aerosols can suppress the formation of warm and mixed-phase clouds, reducing the total precipitation by up to 3.7 to 14.9 mm day⁻¹ (or by up to 94.1% to 100%) and the domain mean total precipitation by 0.03 to 0.25 mm day⁻¹ (or by 3.0% to 5.5%). Reduced precipitation slows down wet scavenging of air pollutants, also worsening air pollution. As indicated by Zhang et al. (2010), the small aerosol effect on precipitation in January is due to the omission of ice nuclei and its impact on ice cloud formation. The effects of anthropogenic aerosols in 2005 are comparable to those in 2001 reported by Cai et al. (2015) and those in 2008 reported by X.-Y. Liu et al. (2015), due to similar amounts of anthropogenic emissions of primary aerosols and the precursor gases of secondary aerosol in 2005 relative to 2001 and 2008. Such effects in East Asia are more significant than those of the total aerosols in continental U.S. and Europe reported by Zhang et al. (2010) due to higher PM concentrations resulted from its severe pollution in East Asia. As a consequence of changes in meteorological, radiative, and cloud variables, chemical predictions are affected to some extent. For example, as shown in Figure 10, the simulated max 8-hr O₃ mixing ratios are lower over most of the domain, because of lower SNSR and T₂, with domain mean decreases of 0.5-1.2 ppb and maximum decreases of 4.6-8.4 ppb. By contrast, the simulated CO mixing ratios are higher over most of the domain

with domain mean increases of 3.3-4.9 ppb and maximum increases of 82.2-294.3 ppb, because of reduced OH resulted from reduced atmospheric oxidation capacity.

5. Conclusions

This Part II paper addresses several problems and limitations identified in Part I paper based on the comprehensive evaluation of the initial application of WRF/Chem with default model treatments and original model inputs from a previous application of CMAQ over East Asia. To improve the model performance, several modifications are made in model inputs, treatments, and configurations, including the use of reinitialization of meteorological variables, the adjusted anthropogenic emission and vertical distributions, revised BCs of O₃, a revised surface drag parameterization, modified or new dust emission schemes, and a simple SOA formation module. The sensitivity simulation, ADJEMIS1, includes the reinitialization of meteorological variables, adjusted anthropogenic emissions, and the modified Shaw (2008) dust emission scheme. The sensitivity simulation, ADJEMIS2, includes all the aforementioned modifications and the new dust emission scheme of Zender et al. (2003).

Comparing to ORIGEMIS, the use of reinitialization in ADJEMIS1 and ADJEMIS2 moderately to significantly reduces the biases and increases the spatial correlations in simulated T2, Q2, WS10, and Precip. The use of the revised surface drag parameterization in ADJEMIS2 further reduces the biases in simulated WS10. The use of adjusted anthropogenic emissions in ADJEMIS1 based on the first method reduces the degree of underpredictions of surface CO, NO, NO₂, and SO₂ at all sites but worsens the performance in the simulated column abundances of CO, NO₂, SO₂, and O₃ as well as AOD in terms of domain mean statistics and spatial correlations in most months. Compared to the default Shaw (2008) scheme that generates

unrealistically large dust emissions over non-dust source regions in ORIGEMIS, the modified Shaw (2008) scheme used in ADJEMIS1 generates dust emissions over known dust resource regions. However, it does not improve the performance of PM₁₀ at most sites in most months.

Through the use of adjusted anthropogenic emissions and their vertical distributions based on the method 2, the Zender et al. (2003) dust emission scheme, and the updated BCs for O₃, ADJEMIS2 shows moderate to large improvement in simulated mixing ratios of all species with only a few exceptions and column abundances of NO, NO₂, SO₂, and O₃ in terms of domain mean performance statistics, hourly concentrations of CO, NO, NO₂, SO₂, O₃, and PM₁₀ in Hong Kong, monthly mean concentrations of O₃ in the six cities in all months and PM₁₀ concentrations in the six cities in April, July, and October, and in four out of six cities in January in Japan, the vertical profile of SO₂ concentrations over northern China, and the concentrations of PM_{2.5} and its composition (i.e., SO₄²⁻, NO₃⁻, NH₄⁺, and OC) at the THU site and those of SO₄²⁻ and NH₄⁺ at the Miyun site in Beijing, China. The use of the simple SOA module further improves OC performance at the THU site, and the performance of PM_{2.5} and PM₁₀ at all other sites except for Hong Kong. ADJEMIS2 also improves the predictions of AOD, GSW, GLW, LWP, and CF in terms of both performance statistics and spatial correlations in all months except for October. Among the three sensitivity simulations, ADJEMIS2 has the best overall performance for all major meteorological variables, surface concentrations, and column mass abundances.

Compared to MM5 results described in Part I paper, the meteorological predictions from ADJEMIS1 and ADJEMIS2 either outperform or perform similarly. Compared to CMAQ results described in Part I paper, ADJEMIS2 shows moderate to significant improvements for most surface concentrations and column mass abundances. Additional evaluation shows that

both CMAQ and improved WRF/Chem largely underpredict the wet deposition fluxes of SO_4^{2-} , NO_3^- , and NH_4^+ at all EANET sites, due to underpredictions in Precip and/or the concentrations of SO_4^{2-} , NO_3^- , and NH_4^+ . Both models significantly underpredict the concentrations of SO_4^{2-} but overpredict those of NO_3^- at most EANET sites. They predict significantly different spatial distributions, magnitudes, and domain mean for wet deposition fluxes of SO_4^{2-} , because of different gaseous precursor concentrations and Precip predictions, and the use of different wet deposition parameterizations. They also give different dry deposition predictions with higher dry deposition fluxes of SO_2 and O_3 but lower values for NO_2 and NH_3 by WRF/Chem because of different gas-phase chemistry and dry deposition treatments.

Anthropogenic aerosols increase $\text{CCN}_{0.5}$ by 59.7-70.3%, AOD by 162.9-307.3%, CDNC by 143.8% to 183.3%, and COT by 47.6-63.3%. They decrease SNSR by up to 18.7-38.6%, T2 by up to 0.5 to 1.1 °C, WS10 by up to 56.8-88.6%, PBLH by up to 13.0-28.2%, and precipitation by up to 94.1% to 100% through direct and indirect effects. The reductions in WS10, PBLH, and precipitation may further worsen the air pollution by weakening the ventilation and dispersion and decreasing the wet scavenging rates of air pollutants. The changes in meteorological, radiative, and cloud variables lead to changes in chemical predictions such as domain mean decreases of 0.5-1.2 ppb for max 8-hr O_3 mixing ratios and increases of 3.3-4.9 ppb for CO.

There are several limitations of this work. First, the simple SOA module is based on the simple aerosol yield method, and does not include gas-phase oxidation and aerosol equilibrium partitioning. While the use of such a simple SOA module does improve the performance of OC, $\text{PM}_{2.5}$, and PM_{10} , it cannot provide the insights into the formation mechanisms of the SOA. More

advanced SOA methods (e.g., the volatile basis set) should be used in the future. Second, the site-specific results at the THU and Miyun sites show difficulty of the model in capturing observed hourly, diurnal, or weekly concentrations of $\text{PM}_{2.5}$, indicating a need to improve the model's capability to better capture the characteristics of in-situ $\text{PM}_{2.5}$ and its composition at small temporal scales. Additional diagnostic analyses and sensitivity simulations are needed to pin-point likely causes for such difficulties, and explore possible methods for improvement. The evaluation of wet deposition and the intercomparisons of dry and wet depositions prediction from CMAQ and WRF/Chem indicate large uncertainties in dry and wet deposition treatments used in current 3-D atmospheric models, which should be further investigated in the future. In addition, the use of a horizontal grid resolution of 36-km may be too coarse to resolve subgrid scale processes such as cumulus cloud formation and precipitation, as well as emissions and chemistry at smaller spatial scales. Finer grid resolutions (e.g., 1-4 km) should be used when computer resources and emissions become available.

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Figure 10. Spatial distributions of absolute differences in the concentrations of PM_{10} and $\text{CCN}_{0.5}$, radiative, cloud, and meteorological variables, and the mixing ratios of O_3 and CO due to anthropogenic aerosols during January, April, July, and October in 2005.

Table 1. Baseline and sensitivity simulations using WRF/Chem

Sequence	Time Period	Simulation ID	Differences in Configuration		
			Meteorology	Model Treatment	Model Inputs
Baseline	Jan., Apr., Jul., Oct., 2005	ORIGEMIS	Free run with nudging winds in the PBL; Original PBL scheme	Shaw (2008) online dust scheme; No SOA treatment	Original anthropogenic emissions; Original O ₃ BCs
Sensitivity 1	Jan., Apr., Jul., Oct., 2005	ADJEMIS1	Daily reinitialization of meteorological variables; Original PBL scheme	Modified Shaw (2008); No SOA treatment	Adjusted anthropogenic emissions based on method 1 (see Table S1); Original O ₃ BCs
Sensitivity 2	Jan., Apr., Jul., Oct., 2005	ADJEMIS2	Daily reinitialization of meteorological variables; Modified PBL scheme with wind speed bias correction	Zender et al. (2003) online dust scheme; Simple SOA treatment	Adjusted anthropogenic emissions with re-distribution of vertical profile based on method 2 (Table S2); New O ₃ BCs (Figure S1)
Sensitivity 3	Jan., 2005	WCoff	Same with ADJEMIS2, but wind speed bias correction turned off	Same with ADJEMIS2	Same with ADJEMIS2
Sensitivity 4	Jul., 2005	SOAoff	Same with ADJEMIS2	Same with ADJEMIS2, but with SOA formation turned off	Same with ADJEMIS2
Sensitivity 5	Jan., Apr., Jul., Oct., 2005	PMoff	Same with ADJEMIS2	Same with ADJEMIS2 but turning off secondary PM formation through aerosol and cloud chemistry	Same with ADJEMIS2 but turning off anthropogenic PM emissions

Table 2. Performance statistics in terms of normalized mean bias (%) for meteorological variables from WRF/Chem

Variable	ORIGEMIS				ADJEMIS1				ADJEMIS2				WCoff	SOAoff
	Jan.	Apr.	Jul.	Oct.	Jan.	Apr.	Jul.	Oct.	Jan.	Apr.	Jul.	Oct.	Jan.	Jul.
T2	75.6	-9.8	-6.0	1.5	33.1	-6.0	-3.3	-0.8	23.8	-6.1	-3.4	-1.4	28.9	-3.3
Q2	23.4	1.5	0.4	13.1	15.4	7.8	3.5	6.6	13.8	6.4	2.7	5.3	15.3	2.7
WS10	101.0	66.5	70.9	84.5	84.3	58.8	56.4	72.7	47.6	24.7	22.5	37.3	84.1	22.6
Precip	92.7	13.6	11.4	89.8	24.6	10.4	-4.6	1.1	15.6	3.8	-9.1	-8.4	24.5	-9.2

Table 3. Performance statistics in terms of normalized mean bias (%) for chemical species and AOD from WRF/Chem

(a) January, 2005

Variable	ORIGEMIS				ADJEMIS1				ADJEMIS2				WCoff			
	China/ E. Asia ^b	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Tai- wan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan
CO	-	-64.0	-62.4	-69.7	-	-54.0	-51.4	-62.1	-	-62.6	-60.1	-66.9	-	-65.6	-62.5	-67.5
NO	-	-99.4	-96.8	-96.9	-	-99.0	-92.1	-91.8	-	-97.8	-86.3	-90.4	-	-97.8	-87.9	-91.2
NO ₂	-	-88.1	-75.5	-80.2	-	-83.5	-57.2	-67.8	-	-73.8	-51.4	-68.5	-	-54.5	-49.0	-70.2
SO ₂ ^a	-71.6	-79.6	-65.5	-81.6	-48.6	-68.1	-45.9	-65.9	-48.1	-57.9	-37.7	-68.2	-53.3	-67.8	-37.7	-72.5
O ₃	-	160.3	83.8	33.3	-	185.5	64.8	26.0	-	128.7	32.1	4.2	-	120.5	31.5	4.8
PM _{2.5}	-	-	-61.0	-	-	-	-68.1	-	-	-	-56.8	-	-	-	-61.4	-
PM ₁₀ ^a	-37.8	-15.8	-60.7	-46.0	-37.5	-49.4	-71.5	-68.1	-38.6	-30.4	-64.0	-60.4	-15.0	-38.4	-66.1	-54.0
Column CO	4.0	-	-	-	17.1	-	-	-	7.2	-	-	-	6.8	-	-	-
Column NO ₂	-30.6	-	-	-	3.7	-	-	-	2.4	-	-	-	1.3	-	-	-
Column SO ₂	21.4	-	-	-	132.2	-	-	-	72.1	-	-	-	59.4	-	-	-
TOR (TOMS)	45.4				47.6				28.5				28.4			
TOR (OMI)	27.2	-	-	-	29.1	-	-	-	12.4	-	-	-	12.3	-	-	-
AOD	-16.2	-	-	-	-21.1	-	-	-	11.4	-	-	-	11.6	-	-	-

(b) April, 2005

Variable	ORIGEMIS				ADJEMIS1				ADJEMIS2			
	China/ E. Asia ^b	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Tai- wan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan
CO	-	-50.6	-67.2	-61.4	-	-61.9	-57.7	-53.7	-	-58.7	-56.2	-52.7
NO	-	-99.2	-92.2	-92.9	-	-98.6	-85.7	-89.9	-	-97.4	-66.1	-84.1
NO ₂	-	-86.1	-67.5	-76.4	-	-79.2	-50.9	-66.1	-	-35.6	-38.3	-60.7
SO ₂ ^a	-80.4	-84.2	-44.5	-79.5	-62.0	-68.7	-11.8	-65.5	-57.5	-3.7	-13.7	-66.2
O ₃	-	88.0	13.6	-11.0	-	122.3	19.8	-4.5	-	36.7	-0.1	-15.7
PM _{2.5}	-	-	-41.1	-	-	-	-56.6	-	-	-	-37.2	-
PM ₁₀ ^a	-17.2	-47.4	-49.7	33.3	-54.5	-30.5	-67.1	-37.7	-12.2	-2.7	-51.0	-5.5
Column CO	-24.2	-	-	-	-18.3	-	-	-	-18.6	-	-	-
Column NO ₂	-19.1	-	-	-	11.1	-	-	-	16.7	-	-	-
Column SO ₂	-31.7	-	-	-	32.1	-	-	-	-2.2	-	-	-
TOR (TOMS)	-0.5				2.0				-6.5			
TOR (OMI)	14.2	-	-	-	17.1	-	-	-	7.3	-	-	-
AOD	-38.7	-	-	-	-49.1	-	-	-	-18.9	-	-	-

(c) July, 2005

Variable	ORIGEMIS				ADJEMIS1				ADJEMIS2				SOAoff			
	China/ E. Asia ^b	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Tai- wan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan
CO	-	-72.0	-62.6	-59.5	-	-65.01	-50.7	-46.6	-	-64.8	-48.5	-46.5	-	-64.8	-48.5	-46.2
NO	-	-98.7	-91.8	-93.2	-	-97.3	-80.2	-88.4	-	-84.8	-52.7	-79.2	-	-85.1	-53.8	-79.7
NO ₂	-	-83.7	-61.7	-64.4	-	-71.0	-37.0	-46.6	-	-31.9	-20.6	-38.3	-	-31.9	-20.4	-38.3
SO ₂ ^a	-80.1	-81.6	-49.6	-78.0	-61.4	-65.0	-8.9	-62.3	-57.3	-40.1	-6.3	-61.1	-57.2	-40.3	-5.9	-61.0
O ₃	-	125.0	14.5	16.3	-	134.3	20.3	38.7	-	26.8	2.3	25.3	-	28.6	4.0	27.3
PM _{2.5}	-	-	-63.9	-	-	-	-58.0	-	-	-	-26.5	-	-	-	-32.9	-
PM ₁₀ ^a	-35.5	-31.8	-68.9	-51.2	-36.6	-25.2	-66.5	-53.1	-7.2	7.2	-47.4	-24.8	-12.3	2.7	-51.1	-31.8
Column CO	-3.5	-	-	-	5.2	-	-	-	4.1	-	-	-	4.4	-	-	-
Column NO ₂	-11.3	-	-	-	26.6	-	-	-	31.4	-	-	-	31.0	-	-	-
Column SO ₂	-40.0	-	-	-	23.3	-	-	-	-17.2	-	-	-	-17.1	-	-	-
TOR (TOMS)	-32.3				-27.3				-17.0				-16.9			
TOR (OMI)	-19.4	-	-	-	-13.4	-	-	-	-1.1	-	-	-	-1.0	-	-	-
AOD	-28.0	-	-	-	-19.5	-	-	-	19.6	-	-	-	14.6	-	-	-

(d) October, 2005

Variable	ORIGEMIS				ADJEMIS1				ADJEMIS2			
	China/ E. Asia ^b	Hong Kong	Taiwan	Japan	China/ E. Asia	Hong Kong	Tai- wan	Japan	China/ E. Asia	Hong Kong	Taiwan	Japan
CO	-	-55.3	-53.8	-65.4	-	-45.0	-47.5	-52.5	-	-54.9	-56.2	-59.7
NO	-	-98.7	-93.8	-97.4	-	-97.8	-83.9	-93.0	-	-95.2	-73.6	-90.7
NO ₂	-	-44.0	-68.0	-74.9	-	-40.5	-45.7	-56.5	-	-32.1	-42.3	-56.3
SO ₂ ^a	-78.0	-59.1	-45.9	-79.0	-59.6	-39.0	-11.9	-58.2	-56.2	-21.6	-14.9	-59.2
O ₃	-	94.4	23.7	25.8	-	113.7	7.1	32.6	-	83.9	-8.8	12.8
PM _{2.5}	-	-	-25.2	-	-	-	-60.5	-	-	-	-48.2	-
PM ₁₀ ^a	-2.9	-6.9	-49.3	-15.5	-28.0	-28.6	-70.2	-41.7	-38.6	1.1	-58.6	-10.2
Column CO	0.2	-	-	-	11.4	-	-	-	2.2	-	-	-
Column NO ₂	-0.01	-	-	-	42.9	-	-	-	34.2	-	-	-
Column SO ₂	26.1	-	-	-	172.6	-	-	-	89.5	-	-	-
TOR (TOMS)	5.6				8.9				1.1			
TOR (OMI)	26.7	-	-	-	13.7	-	-	-	-19.6	-	-	-
AOD	5.6	-	-	-	11.7	-	-	-	56.1	-	-	-

^a The SO₂ and PM₁₀ concentrations in mainland China are derived from Chinese Air Pollution Index data.

^b The domain is mainland China for surface concentrations and East Asia for column variables

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Figure 2. Continued.

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