Application of Online-Coupled WRF/Chem-MADRID in East Asia: Model Evaluation and Climatic Effects of Anthropogenic Aerosols

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Abstract

The online-coupled Weather Research and Forecasting model with Chemistry with the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (referred to as WRF/Chem-MADRID) is applied to simulate meteorological fields, air quality, and the direct and indirect effects of anthropogenic aerosols over East Asia in four months (January, April, July, and October) in 2008. Model evaluation against available surface and satellite measurements shows that despite some model biases, WRF/Chem-MADRID is able to reproduce reasonably well the spatial and seasonal variations of most meteorological fields and chemical concentrations. Large model biases for chemical concentrations are attributed to uncertainties in emissions and their spatial and vertical allocations, simulated meteorological fields, imperfectness of model representations of aerosol formation processes, uncertainties in the observations based on air pollution index, and the use of a coarse grid resolution. The results show that anthropogenic aerosols can reduce net shortwave flux at the surface by up to $40.5\text{--}57.2\text{ W m}^{-2}$, Temperature at 2-m by up to $0.5\text{--}0.8^{\circ}\text{C}$, NO_2 photolytic rates by up to $0.06\text{--}0.1\text{ min}^{-1}$ and the planetary boundary layer height by up to $83.6\text{--}130.4\text{ m}$. Anthropogenic aerosols contribute to the number concentrations of aerosols by up to $6.2\text{--}8.6\times 10^4\text{ cm}^{-3}$ and the surface cloud concentration nuclei at a supersaturation of 0.5% by up to $1.0\text{--}1.6\times 10^4\text{ cm}^{-3}$. They increase the column cloud droplet number concentrations by up to $3.6\text{--}11.7\times 10^8\text{ cm}^{-2}$ and cloud optical thickness by up to $19.8\text{--}33.2$. However, anthropogenic aerosols decrease daily precipitation in most areas by up to $3.9\text{--}18.6\text{ mm}$ during the 4 months. These results indicate the importance of anthropogenic aerosols in modulating regional climate changes in East Asia through aerosol direct and indirect effects, as well as the need to further improve the performance of online-coupled models.

Keywords: Online-Coupled Model, WRF/Chem-MADRID, Model Evaluation, Aerosol Direct Effects, Aerosol Indirect Effects, East Asia

1. Introduction

Aerosols are known to affect air quality, health, and ecosystems, but the extent to which they affect climate change is unclear and remains a cause for concern (IPCC, 2013). The uncertainty regarding the effects of aerosols on radiative forcing is much higher than that of greenhouse gases (IPCC, 2013). The direct effects of aerosols involve the absorption and scattering of solar radiation, whereas indirect effects involve interactions between aerosols and clouds. Aerosol indirect effects through acting as cloud condensation nuclei (CCN) or ice nuclei (IN) affect cloud formation and optical properties. Twomey (1974) indicated that a decrease in cloud droplet effective radius results in increases of both cloud optical thickness (COT) and cloud albedo, which is referred to as the first indirect effect of aerosols. A decrease in precipitation efficiency can prolong the cloud lifetime, associated with an increase of cloud droplet number concentrations (CDNC) and a decrease of the cloud droplet effective radius, is referred to as the second indirect effect (Albercht, 1989). The direct and indirect effects of aerosols have been studied using satellites (Sekiguchi et al., 2003), field observations (Coakley and Walsh, 2002), and numerical models such as general circulation models (GCMs) (Quaas et al., 2006) and online-coupled regional meteorology/climate-chemistry models (Fast et al., 2006; Zhang et al., 2010a, b, 2012a, b). GCMs have more advantages than satellite-derived and field observations (e.g., they can simulate historical and future scenarios and multi-scale regions). However, the spatial resolutions of GCMs are too coarse to represent the effects of regional changes caused by local aerosol emissions and meteorology, which can be well represented by the regional online-coupled models.

The Weather Research and Forecasting Model with chemistry (WRF/Chem) of Grell et al. (2005) is an online model that has recently been developed to simulate the meteorology and chemistry interactions. Since its initial release in 2002, WRF/Chem has been widely applied for air quality and meteorology research (Zhang et al., 2010a, b; Zhang et al., 2012b, 2013a, b; Wang et al., 2014). In particular, several studies have

83 applied WRF/Chem to simulate aerosol direct and indirect effects over regional
84 domains such as the U.S. and Europe, and over the global domain (Zhang et al.,
85 2010b, 2012a, b, 2013a). WRF/Chem has also been applied in China or East Asia in a
86 number of studies. Among these studies, only a few of them simulate the climatic
87 impacts of anthropogenic aerosols (e.g., Gao et al., 2014; Cai et al., 2014; Zhang et al.,
88 2014a). In addition, the other online coupled regional climate-chemistry-aerosol
89 model namely RIEMS-Chemaero had been developed by Han et al. (2010, 2011, 2012)
90 and namely RegCCMS (Zhuang et al., 2013b) also had been used to simulate the
91 aerosol effects over East Asia besides WRF/Chem, the former model only addresses
92 aerosol direct effects, whereas both aerosol direct and indirect effects could be
93 addressed from the latter model (Zhuang et al., 2013a). East Asia, especially China,
94 has become a major source region of anthropogenic aerosols and their precursors with
95 dramatically developing during the past 30 years (Zhang et al., 2009). Such as
96 continuous increase of anthropogenic emissions over this region will affect both
97 regional and global climate change, especially the occurrence of the widespread,
98 persistent, and severe haze in January 2013 over northern and eastern China (Jiang et
99 al., 2014) and in December 2013 over the Yangtze River delta (YRD) (Wang et al.,
100 2015). Although many studies give the climatic effect of one aerosol or multi aerosols
101 or all aerosols over this region (Han et al., 2010, 2011, 2012; Zhuang et al., 2013a, b),
102 few studies focus on seasonality of both direct and indirect climatic effects of
103 anthropogenic aerosols. In addition, aerosol direct effects were estimated over East
104 Asia (China) by many papers, however, few studied involved in aerosol indirect effects
105 so far. So the understanding between climate change and anthropogenic aerosols is not
106 enough to make strategies and policies of reducing pollutants emissions and slowing
107 climate change.

108 Comparing to previous studies, this work is unique in several aspects. First, this
109 study performs a comprehensive evaluation using surface and satellite data, in
110 particular, the observations-derived Air pollution indexes (API), which were

published by national and local governments, used in this study cover almost the entire China. Second, this study estimates the direct and indirect effects of anthropogenic aerosols over East Asia in 2008 which is similar to Cai et al. (2014) in which such effects are estimated for 2001. Compared to Cai et al. (2014) that used the released version of WRF/Chem, this work uses WRF/Chem with the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (i.e., WRF/Chem-MADRID, Zhang et al., 2010a) with a different gas-phase chemical mechanism and aerosol module from those used in Cai et al. (2014). Third, WRF/Chem-MADRID was firstly used in East Asia to evaluation its performance and estimate seasonality of anthropogenic aerosols direct and indirect effects.

The aims of this study are to evaluate the online-coupled WRF/Chem-MADRID with various feedback mechanisms and to estimate the direct and indirect effects of anthropogenic aerosols over East Asia. In this paper, Section 2 describes the model configuration, emissions, and observational data used. In Section 3, the model performance is evaluated against observed meteorological parameters, chemical concentrations, and satellite data. Section 4 presents results of the direct and indirect effects of anthropogenic aerosols over East Asia in the four selected months of 2008, and Section 5 summarizes the major findings and limitations of this work.

2. Model setup and evaluation protocols

2.1. Model domain and setup

WRF/Chem-MADRID is applied to East Asia at a vertical resolution of 23 layers (from the surface to the tropopause) and a horizontal resolution of 36-km. WRF/Chem-MADRID has been applied for retrospective simulations of air quality and its interactions with meteorology in the U.S. and Europe as well as real-time air quality forecasting of O₃ and PM_{2.5} in the U.S. (Zhang et al., 2010a, 2012a, 2013a, b; Chuang et al., 2011; Yahya et al., 2014; Zhang et al., 2014b), and showed promising skill in

reproducing both meteorological and chemical observations and forecasting short-term air quality. The WRF/Chem configuration options used in this study are listed in Table S-1.

MADRID uses a sectional representation of aerosol size distribution (8 size section, 0.0215-10 μ m) to simulate both the mass and the numbers in each aerosol size bin, which is similar with the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) and different with Modal Aerosol Dynamics Model for Europe coupled with Secondary Organic Aerosol Model (MADE/SORGAM). In present study, internal mixing is assumed to within each aerosol size bin, while external mixing is treated between different aerosol size bins. ISORROPIA is used to simulate inorganic aerosol thermodynamic equilibrium and a mechanistic representation that simulates both hydrophilic and hydrophobic particles is used to simulate SOA formation (Pun et al., 2002). And binary nucleation of sulfuric acid and water vapor is used to simulate the formation of new particles through homogeneous nucleation, and the moving-center scheme approach (Jacobson, 1997) is used to simulate the growth of particles over sections with fixed size boundaries. WRF/Chem-MADRID uses the same scheme for aerosol hygroscopic growth as does WRF/Chem, i.e., the scheme of Leaitch et al. (1986), which is included as part of the aerosol activation/droplet nucleation parameterization of Abdul-Razzak and Ghan (2000) used in WRF/Chem. The aerosol species comprised sulfate, nitrate, ammonium, EC (Element carbon), OC (Organic carbon), Cl⁻ (Chloride ion), Na⁺ (sodium), the other inorganics, and aerosol water. The calculations of the optical and microphysical properties are performed using several physical schemes separately from the aerosol modules in WRF/Chem. Such calculations are therefore the same for MADRID and MOSIAC, although they depend on aerosol concentrations and properties simulated by MADRID and MOSIAC (which may be somewhat different, because MADRID and MOSIAC differ in terms of their treatments for aerosol dynamics, thermodynamics, and size distribution). A brief description is provided below. The Detailed treatments can be found in Fast et al. (2006)

and Chapman et al. (2009). Aerosol optical properties as a function of wavelength are determined by aerosol chemical properties and sizes. First, the refractive index of each size bin is calculated by volume averaging, and Mie theory is used to calculate the extinction efficiency, the scattering efficiency, and the intermediate asymmetry factor. And then all above parameters are summarized from all size bins to determine the aerosol optical properties. The effect of aerosols on incoming solar radiation is determined based on the Goddard shortwave radiation scheme once the aerosol optical properties are determined. Hygroscopic properties in this study depend on particulate composition. Aerosol particles are activated to form cloud droplets within each size bin following Abdul-Razzak and Ghan (2002). Cloud droplets evaporate and aerosols are resuspended when cloud dissipates in a grid cell. And aerosol activation and resuspension are calculated simultaneously using turbulent vertical mixing in WRF/Chem-MADRID. The Lin microphysics scheme (Lin et al., 1983) which treats water vapor and other hydrometeors: cloud water, rain, snow, graupel and cloud ice includes a prognostic treatment of cloud droplet number (Ghan et al., 1997) and ice clouds through prescribing ice nuclei distribution, and making the autoconversion of cloud droplets to rain droplets was added to WRF/Chem following Liu et al. (2005). The interactions of clouds and radiations are connected by cloud droplet number. More details on other physics options and aerosol-cloud interaction treatments have been described elsewhere (Grell et al., 2005; Fast et al., 2006; Chapman et al., 2009).

WRF/Chem-MADRID is applied to the four months in 2008: January, April, July, and October, each representing one season. Note that July is the month before the 2008 Summer Olympic Games that took place in Beijing, China, from August 8 to 24, 2008. The meteorological initial and boundary conditions are derived from the $1.0 \times 1.0^\circ$ and 6-hour resolution National Centers for Environmental Prediction global Final Analysis reanalysis data (<http://rda.ucar.edu/datasets/ds083.2/>). Initial chemical conditions and boundary conditions for the gas and aerosol species are obtained from a global chemical transport model (i.e., GEOS-Chem)

(<http://acmg.seas.harvard.edu/geos/index.html>). All simulations on subsequent days use the chemical predictions at the last hour of the previous day as the initial chemical conditions for the next day. The anthropogenic emission inventories for gaseous and PM species are taken from INTEx-B 2006 (Zhang et al., 2009) and different monthly anthropogenic emission inventories are used in this work to simulate seasonal variation of gaseous and aerosols. The 2008 emissions over China are adjusted according to the evaluation results from MM5-CMAQ using surface and satellite observations (Liu et al., 2010). Emissions of sea salt and dust are generated online using Gong et al. (2002) and a modified Shaw (2008), respectively. A 7- or 8-day spin-up (at the beginning from the 24th day of the previous month of every run month) is used to reduce the influence of initial conditions on the modeled results.

In the baseline simulations, all emissions are included and all chemical and meteorological processes as well as the feedback between the meteorological processes and chemical species are treated. To estimate the impact of anthropogenic aerosol direct and indirect effects, a sensitivity simulation is performed by turning off anthropogenic PM emissions, and secondary PM formation through aerosol and cloud chemistry, but use the same model configuration and initial and boundary conditions as the baseline simulation. Therefore, the differences between the two sets of simulations represent the direct and indirect effects of anthropogenic aerosols. This approach is similar to that of Zhang et al. (2010b) except that Zhang et al. (2010b) focused on the effects of both anthropogenic and natural aerosols whereas this work focuses only the effects of anthropogenic aerosols. The spatial distributions of observations and predictions are analyzed. Several sites are selected for detailed temporal analyses. The details of evaluation protocols can be seen in supplement materials.

2.2. Observational datasets and evaluation protocols

The variables and observational data used in this study are summarized in Tables

S-2 and S-3. Surface chemical concentrations of SO₂, NO₂, and PM₁₀ are derived from API reported by the Ministry of Environmental Protection of the People's Republic of China and the Local Environmental Protection Bureau. Further details about the conversion between API and SO₂ or NO₂ or PM₁₀ concentration can be found elsewhere (Qu et al., 2010). The hourly O₃ mixing ratios were obtained at three sites (Ryori, Tsukuba, and Yonagunijima) in Japan. The geographical distributions of the surface meteorological and chemical sites are shown in Figure S-1. PM_{2.5} data used for evaluation are based on three different measurement methods (TEOM, half-hourly resolution; medium flow, daily resolution; small flow, weekly resolution). The daily average mass concentrations of EC, OC, SO₄²⁻, NO₃⁻, and NH₄⁺ in PM_{2.5} were observed at Tsinghua University (THU) and Miyun (MY) sites only in July. The simulated aerosol optical depth (AOD), cloud water path (CWP), cloud fraction (CF), and cloud optical thickness (COT) are compared with satellite observations from the Moderate Resolution Imaging Spectroradiometer (MODIS, <http://ladsweb.nascom.nasa.gov/data/search.html>), and Cloud droplet number concentrations (CDNCs) are calculated by Bennartz (2007) using MODIS data, and the column abundances of CO, NO₂ and the tropospheric ozone residual (TOR) are compared with satellite observations from the Measurements of Pollution In The Troposphere (MOPITT, http://eosweb.larc.nasa.gov/PRODOCS/mopitt/table_mopitt.html), Ozone Monitoring Instrument (OMI, <http://www.temis.nl/airpollution/no2.html>), OMI (http://acdb-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html), respectively. In this work, model predictions from 10:00 A.M. to 12:00 A.M. local standard time (LST) are extracted and averaged to compare with the MODIS due to Terra orbits cross the equator at 10:30 A.M.LST. And the frequency, spatial resolution and other information of satellite data are listed in Table S-3.

3. Model evaluation

3.1. Gaseous species and particulate matter

Figure 1 shows the overlay plots of simulated and observed monthly-mean daily-averaged surface concentrations of SO₂, NO₂, the maximum 1-hr O₃, and PM₁₀ concentrations. Tables 1 show performance statistics for gaseous and PM species, respectively. The highest simulated SO₂ concentrations occur in January, which is consistent with observations and is caused by high SO₂ emissions and other reasons (e.g., low temperature, low planetary boundary layer that prevents dispersion of air pollution and dry weather condition that reduces the rate of removal for pollutants) as reported by Liu et al. (2010). WRF/Chem underpredicts the concentrations of SO₂ in all 4 months with MBs of -42.6 ppb to -11.8ppb and NMBs of -71.5% to -45.6%. The highest bias occurs in January due mainly to underestimated emissions of SO₂ and overpredicted wind speed (Table S-4). Such underpredictions have also been reported in other studies (e.g., CMAQ, Liu et al. (2010)). Similar to SO₂, NO₂ concentrations vary seasonally, with the maximum concentrations in January, followed by October, July, and April. Unlike the observations that show higher concentrations of SO₂ and NO₂ in April than in July, the simulated concentrations of SO₂ and NO₂ in July are higher than those in April, because of the fact that the model more significantly overestimates the wind speed in April than in July, which permits greater accumulation of pollution in July than in April. The maximum 1-hr O₃ concentrations are moderately underpredicted in April and slightly-to-moderately overpredicted in other months. The highest value of simulated surface maximum 1-hr O₃ occurs in July (up to 111.5 ppb, Figure 1) over Sichuan, YRD, and the North China Plain; especially the North China Plain because of the relatively higher O₃ precursor emissions and stronger photochemical reactions. The low concentrations for most species in July result from a stringent emission control prior (Liu et al., 2011) to the 2008 Beijing Summer Olympic Games and a heavy precipitation that can effectively remove air pollutants from the atmosphere.

The highest concentrations of simulated PM₁₀ occur in January with maximum

concentrations up to $441.8 \mu\text{g m}^{-3}$ over the Taklimakan Desert in Xinjiang province, the North China Plain, Sichuan Basin, the east of Hubei province, and north of Anhui and Jiangsu provinces. The simulated levels of PM_{10} show the lowest concentrations in July because of heavy precipitation (see Table S-4). Compared with the observations of PM_{10} , the simulated values are moderately-to-largely underpredicted for all 4 months with NMBs of -59.3% to -31.0% . Several factors may contribute to such biases. First, the wind speed is overpredicted significantly (Table S-4), which would enhance the diffusion of pollutants. Second, the emissions of primary PM species such as BC, OC, and mineral dust and the precursors for secondary PM species such as SO_2 , NO_2 , and volatile organic compounds (VOCs) may have been largely underestimated. The performance statistics of PM_{10} are similar to those of CMAQ (Liu et al., 2010).

WRF/Chem underpredicts $\text{PM}_{2.5}$ concentrations slightly-to-moderately with NMBs of -18.5% to -7.0% in July but significantly with NMBs of -71.7% to -47.2% against TEOM measurements and -64.0% to -50.9% against small flow measurements in the other three months, and -63.3% against medium flow data in January. For SO_4^{2-} and OC concentrations, WRF/Chem underpredicts in July with the NMBs of -45.9% and -41.1% , respectively. The large underpredictions in SO_4^{2-} concentrations may be attributed to several factors such as underestimation in the emissions of SO_2 as reported by Liu et al. (2010), missing formation pathways through heterogeneous chemistry as indicated by Wang et al. (2012), and overpredictions in wind speeds (Table S-4). NH_4^+ , NO_3^- , and BC concentrations in July are slightly overpredicted with NMBs of 1.0, 23.2, and 6.8%, respectively. The results indicate possibly large underestimations in the emissions of SO_2 and primary OM and underpredictions of secondary organic aerosol (SOA) due to its missing precursors (e.g., primary OM, glyoxal) and pathways (e.g., aqueous oxidation in clouds) that are not treated in WRF/Chem-MADRID.

Figure 2 shows the time series of observed and predicted SO_2 , NO_2 and PM_{10} at

four representative sites (Beijing, Shanghai, Guangzhou, and Xi'an are urban sites) in
 China and PM_{2.5} concentrations at four representative sites (THU located in Beijing is
 a urban site, MY located in Beijing is a suburban site, Baiyangdian (BYD) located in
 Hebei province is a rural site, Shangdu (SD) located in Inner Mongolia is a rural site).
 WRF/Chem-MADRID reproduces SO₂ concentration during all the four months at
 Xi'an and April at Beijing, but overpredicts those in all months at Shanghai and July
 and October at Beijing, underpredicts those in all months at Guangzhou and January
 at Beijing. Similar to SO₂, it overpredicts NO₂ concentrations at Shanghai and
 underpredits those nearly in all months at Guangzhou and Xi'an.
 WRF/Chem-MADRID reproduces NO₂ concentration in all months at Beijing. PM₁₀
 concentrations are significantly underpredicted at Beijing, Guangzhou, and Xi'an,
 particularly at Beijing, whereas good performance is found at Shanghai for PM₁₀. The
 model can capture well the peak concentration in January at Shanghai, but shows
 difficulties in capturing similar peaks at other sites during the same time period. For
 PM_{2.5}, better performance is found in July than in January at the four sites and the
 model can reproduce the peak concentration well in July, but cannot do so in January.
 Although MY and BYD are not urban sites, the observed PM_{2.5} concentrations at both
 sites are almost the same magnitude as those at Beijing in January, suggesting that
 PM_{2.5} pollution has become a regional problem spreading from urban centers to
 suburban and rural areas, and more attentions should be given to the PM_{2.5}
 measurements and modeling over those areas. The large biases in the simulated
 temporal variations are most likely caused by several reasons. First, high episodic
 emissions are not accurately represented in the emissions used in this work, this is
 particularly true in January when the contributions of primary PM emissions are
 relatively large, as well as the uncertainties in the emissions of horizontal and vertical
 allocations can largely affect the model accuracy, as reported by Wang et al. (2014).
 Second, the overpredictions in wind speeds may contribute in part the large
 underpredictions at some sites during some times. Finally, the use of a coarse grid

resolution of 36-km may not be sufficient to capture the observed temporal variations which require the use of a finer grid resolution.

3.2. Performance of Column Predictions and properties of clouds

The simulated spatial distribution of monthly-mean AOD, CF, COT, CWP, CDNC and the column abundance of CO, NO₂ and TOR are compared with satellite observations. Figure 3 and Figure S-4 show the spatial distributions of the observed and simulated monthly mean column abundances, and Table 2 presents the corresponding performance statistics. Overall, WRF/Chem reproduces the observed spatial distributions of those column masses to a large extent and their seasonal variations to some extent. These results are generally consistent with those from CMAQ (Liu et al., 2010). The model performs the best in July with an NMB of -0.6%, but it overpredicts the column concentrations of CO in January and October with NMBs of 10.2% and 11.0%, and underpredicts them in April with an NMB of -23.0%. While MOPITT observed very high CO column mass in the northeastern China, N. and S. Korea, Japan that extends further into the oceanic areas off the east coast of China, the model fails to reproduce them because the emissions used in the simulation did not represent accurately the large open biomass burning emissions over those land areas.

The NMBs of NO₂ are 146.2, 36.1, -0.7, and 80.0% in January, April, July, and October. While the model reproduces well the spatial distribution of the column concentrations of NO₂, it overpredicts them in all months except for July. The largest overpredictions occur over Sichuan, Hubei, Hunan, Anhui, Zhejiang, and Jiangxi provinces of China in January. The overpredictions in column NO₂ but underpredictions in surface NO₂ concentrations indicate inaccuracies in the vertical allocation of the emissions, e.g., surface level emissions may have been underestimated but elevated emissions may have been overestimated.

The TOR in January and April are overpredicted with NMBs of 34.5% and

39.5%, respectively, but underpredicted in July and October with NMBs of -15.8% and -2.8%, respectively. The overpredictions in January and April and underpredictions in July occur throughout most of the domain. The high summer values are likely the result of stronger photochemical reactions. The observed values of TOR are the highest in July, followed by April, October, and January; the simulated values of TOR are the highest in April, followed by January, July, and October. The discrepancies between the observed and simulated values of TOR may be caused by uncertainties in the emissions of O₃ precursors such as NO₂ and VOCs, upper-layer boundary concentrations of O₃, and uncertainties associated with the satellite retrieval algorithms (Liu et al., 2010).

The observed and simulated AOD values are higher over the North China Plain, YRD, PRD (Pearl River Delta), and Sichuan Basin in all four months, which are consistent with the higher PM emissions and higher PM concentrations over these areas than remaining areas. Underpredictions occur in all four months with MBs of -0.3 to -0.03, NMBs of -57.4% to -13.5%, and NMEs of 38.5%-58.1%, especially the north and south of China, owing to the underpredictions of surface PM concentrations especially SO₄²⁻ over those areas as shown in Table 1, the underpredictions of relative humidity (Figure S-2) over those areas will lead to the underpredictions of particle growth factor and further underestimate the optical properties

(http://ruc.noaa.gov/wrf/WG11/wrf_tutorial_2014/WRF_CHEM_feedbacks.pdf), uncertainties in the boundary conditions used, and possible biases in processing the MODIS AODs using some retrieval algorithm (Kaufman et al., 2005; Remer et al., 2005) and uncertainties due to different wavelength (MODIS at 550nm, and WRF/Chem at 600nm). In particular, the largest underpredictions occur for both surface PM_{2.5} and AOD in April.

Underpredictions of CF are found over the north of China with NMBs of -33.4, -33.2, -14.5, and -28.3% in January, April, July, and October, respectively. CWP

is significantly underestimated over the entire domain with NMBs of -86.3% to -78.4% in four months due to the contribution of convective clouds to cloud water content is not yet included in Lin et al.(1983). COT is significantly underestimated over most of the domain with NMBs of -90.2% to -83.2% due to underpredicted CWP. Both magnitude and spatial distributions of CF are more consistent with the MODIS observations than COT and CWP (Table 2 and Figure S-4). CDNC are underpredicted in four months with NMBs of -2.7, -4.0, -3.8, and -3.9%. The highest CDNC occur in April, followed by January, October and July. The spatial distributions of simulated and observed CDNC are quite different though the statistical results of CDNC between model and satellite show the good performance.

4. Direct and indirect effects of aerosols

Figure 4 shows the direct and indirect effects of anthropogenic aerosols on T2, net shortwave flux at the surface (referred to as GSW), NO₂ photolysis rates, and planetary boundary layer (PBL) height (PBLH) in January, April, July, and October. GSW is not only resulted from the scattering and absorption of anthropogenic aerosols to solar radiation but also contributed by the reflection and absorption of clouds which are affected by the changes of anthropogenic aerosols through changing cloud fraction and cloud optical properties and prolonging cloud lifetime. The reductions in GSW and T2 are found over most areas with domain-wide mean values of -6.5 W m^{-2} and $-0.04 \text{ }^{\circ}\text{C}$ in January, -8.3 W m^{-2} and $-0.06 \text{ }^{\circ}\text{C}$ in April, -12.1 W m^{-2} and $-0.08 \text{ }^{\circ}\text{C}$ in July, -8.5 W m^{-2} and $-0.07 \text{ }^{\circ}\text{C}$ in October. Comparing the results from Han et al. (2012) with the monthly mean SW (Shortwave) at the surface due to all aerosol and Non-dust aerosols of -11.40 W m^{-2} and -5.31 W m^{-2} in March 2010 over the whole domain, the value of GSW in April is somewhat larger than it because indirect effect is considered in this study. GSW is caused about 65% due to the direct effects of anthropogenic aerosol, and about 35% contributed by the indirect effects of anthropogenic in spring using the results of Han et al. (2012) and this study if we

neglect the effect of sea salt. The larger changes of GSW caused by anthropogenic aerosols occur in eastern China and the Sichuan Basin where anthropogenic aerosol concentrations are high, as well as near coastal ocean areas that are influenced by transport of anthropogenic aerosols through sea-land-air circulation and interaction, and it includes sea breeze and land breeze, and so on. Due to the large heat capacity of ocean, the ocean is relatively insensitivity to the reductions in the shortwave radiation, leading to a smaller reductions in GSW over oceanic areas. Decreased solar radiation has various impacts on the atmosphere and ecosystem. For example, it reduces near surface temperature and photochemical activities in the atmosphere as shown in Figure 4. It also reduces the photosynthetic activity of plants living on land and in the ocean, which will influence the entire ecosystems. The largest domain-wide mean decreases of T2 occur in July, particularly over the Sichuan Basin, followed by October, April, and January. The changes of T2 with maxima of 0.5-0.8 °C can be found over Sichuan Basin, which is consist with Han et al. (2011), whereas the scope and degree of the T2 in this study less than Han et al. (2011) with maxima of -0.8-1.6 °C due to the fact that the natural aerosols (dust and sea salt) aren't included when simulating the aerosol effects in this study. T2 decreases more than 0.3 °C in July over Beijing, Tianjin and southern of Hebei province, and it is larger than the results from Ma et al. (2012) (0.17 °C at Beijing, 0.18 °C at Tianjin, and 0.23 °C at Shijiazhuang). The biases of two results are due to the fact of the differences of model configurations, simulated year, and simulated domain and so on. The changes in T2 are mainly caused by the changes in GSW but are also affected by other factors including surface properties (e.g., soil moisture and soil temperatures) (Zhang et al., 2010b). The changes in T2 are controlled by cooling effects and warming effects. It is note that, in present study, T2 shows a small increase over ocean in four months, due to the fact that water vapor mixing ratio increases by up to $1.0\text{-}1.8\times10^{-4}\text{ kg kg}^{-1}$ (Figure not shown) and longwave radiation (LR) slightly increases by up to $1.8\text{-}4\text{ W m}^{-2}$ (Figure not shown) over ocean in four months. Such that increases of water vapor mixing ratio and LR are

abundant for offsetting the cooling effect from GSW. So the reduction in GSW over the ocean may not necessarily lead to the reduction in T2. Photolysis rates of NO₂, which are directly affected by reductions in incoming solar radiation and temperature, reduce by up to 0.06-0.1 min⁻¹. The PBLH decreases by up to 105.0, 83.6, 130.4, and 127.8 m in January, April, July, and October, respectively, indicating a more stable atmosphere that is caused by warming resulting from BC absorption of solar radiation within the PBL and cooling at surface resulting from the reduced levels of solar radiation. Small increases occur in the PBLH over some oceanic areas, as a response to the small increases in temperature over those areas. The PBLH decreases more than 40 m in July over Beijing-Tianjin-Hebei region, and it more than the results from Ma et al. (2012) with PBLH decreases 34.42 m in the same region.

Figure 5 shows the monthly-mean vertical profile of BC, PM_{2.5} and the changes in potential temperature (T) at four sites: Beijing, Shanghai, Guangzhou, and Xi'an. Monthly-Mean T values reduce at surface by up to 0.08, 0.10, 0.27, 0.18 °C at Beijing, 0.08, 0.06, 0.06, 0.08 °C at Shanghai, 0.13, 0.09, 0.03, 0.10 °C at Guangzhou, and 0.25, 0.17, 0.33, 0.26 °C at Xi'an in four months, respectively, due to the reductions of solar radiation which are backscattered by anthropogenic aerosols, whereas they increase at higher altitudes attributed to the increase infrared radiation by absorbing aerosols such as black carbon between 1000-4000 m at Beijing, 100-4000 m at Shanghai, 800-3500 m at Guangzhou, 1000-4000 m at Xi'an. And the changes of T between two run cases justify a more stable atmosphere that is caused by anthropogenic aerosols. The layer of stable atmosphere in January (Figure 5, red line) are much thicker than in other three month at all sites, indicating that the pollutants are accumulated easier in January.

Figure 6 shows the changes of aerosol number concentration, CCN at a supersaturation (s) of 0.5% at the surface, CDNC in cloud, COT, and precipitation in 2008 caused by indirect effects of anthropogenic aerosols. Similar to anthropogenic PM mass concentrations, PM number concentrations are higher over eastern China than over western China, with the domain-wide mean and maximum values in the

469 range of $5.1-7.4 \times 10^3 \text{ cm}^{-3}$ and $6.2-8.6 \times 10^4 \text{ cm}^{-3}$, respectively. The domain-wide mean
 470 aerosol number concentration is the highest in January and the lowest in July.
 471 Anthropogenic PM serves as effective CCN, leading to an increase in CCN, CDNC,
 472 and COT. CCN concentrations at $s=0.5\%$ at the surface are higher over eastern and
 473 southern parts of China, particularly in the Sichuan Basin with domain-wide mean
 474 concentrations of $0.6-1.4 \times 10^3 \text{ cm}^{-3}$. The column CDNCs in cloud increase by
 475 $0.4-0.7 \times 10^8 \text{ cm}^{-2}$ domain-wide mean and by up to $3.6-11.7 \times 10^8 \text{ cm}^{-2}$ domain-wide
 476 maximum, with higher values over areas where PM number and CCN concentrations
 477 are high. The regions in which CDNCs increase tend to extend eastward from January
 478 to October, reflecting the long range transport of PM, particularly in upper layers.
 479 Comparing to the domain-wide mean CDNC concentrations of $0.8-1.7 \times 10^6 \text{ cm}^{-2}$
 480 resulted from total aerosol concentrations in the U.S. reported by Zhang et al. (2010b),
 481 the CDNCs resulted from anthropogenic aerosols in East Asia are higher by factors of
 482 43-47, resulting from much higher anthropogenic PM concentrations in East Asia than
 483 in the U.S. Rather, the Monthly-Mean $\text{PM}_{2.5}$ concentrations are higher 2-10 times in
 484 China than in U.S. by comparing the values of $\text{PM}_{2.5}$ concentrations from Fig.6 in
 485 Zhang et al.(2010b) and from Figure 5 in this study. As a consequence of aerosol
 486 indirect effects, COT increases and total daily precipitation decreases over the areas
 487 where CDNC increases, as shown in Figure 6. Larger increases in COT occur over
 488 southern China, particularly in the Yunnan–Guizhou Plateau, Sichuan Basin, and
 489 Hunan Province, and oceanic areas in all months except for July. In July, higher COT
 490 mainly occurs over land areas in southern and northeastern China. Daily precipitation
 491 decreases in most areas during the four months. The values of daily precipitation
 492 decrease by up to 18.6 mm over most of the land areas of China with the largest
 493 reductions in July, which is caused by increasing levels of CCN and CDNCs. The
 494 change of precipitation is the least pronounced in January and most pronounced in
 495 July, which is similar to the findings over the U.S. by (Zhang et al., 2010b). As
 496 explained in Zhang et al. (2010b), since this version of WRF/Chem only considers the

aerosol indirect effects on warm cloud formation and does not treat such effects on ice cloud formation, the impact of precipitation via ice nuclei in January cannot be simulated in this study. For other months, anthropogenic aerosols can mainly suppress the formation of warm rain, leading to appreciable reduction in precipitation, particularly in July. Those results are consistent with reported aerosol impacts on precipitation (e.g., Rosenfeld et al., 2007, 2008). Atmospheric PM can be removed through wet deposition via precipitation scavenging. Reduced precipitation will reduce the sink of pollutants, thus enhance the pollutant concentrations in the atmosphere (Jacobson et al., 2007). The uncertainty of precipitation change is seem to be bigger than other parameters, due to several factors as follows. First, temperature, wind and PBLH are affected by anthropogenic aerosols through both aerosol-radiation interaction and aerosol-cloud process, influencing the evaporation of water vapor and the hydrologic circulation between atmosphere and surface. Second, a series of complex interactions among aerosol, radiation, the cloud-rain conversion rate, cloud properties, cloud lifetime as well as circulation adjustment and water vapor redistribution attributes to the inhomogeneous change in precipitation. Third, the mechanism related to precipitation in this study is imperfect.

5. Conclusions

WRF/Chem-MADRID is applied to simulate the meteorological fields, gaseous and PM species concentrations, and the direct and indirect effects of aerosols over East Asia in January, April, July, and October 2008. Model evaluation against available meteorological and chemical observations from surface networks, and satellites shows that despite some biases WRF/Chem-MADRID can reproduce the spatial distributions and seasonal variations of most meteorological fields, chemical concentrations, and column variables reasonably well.

The simulated concentrations of SO₂, NO₂, and maximum 1-hr O₃ in January are the highest among the four months with NMBs of -70.5, -25.3, and 5.9%,

respectively. However, their lowest concentrations occur in different months: April for SO₂ and NO₂ with NMBs of -71.5% and -45.6%, July for maximum 1-hr O₃ with NMBs of 20.6%. The simulated concentrations of PM₁₀ are the highest in October and the lowest in April, whereas the observations are the highest in January and the lowest in July. The model performs the best in July for PM_{2.5} with NMBs of -18.5% to -7.0%. Moderate to large underpredictions occur for SO₂, PM_{2.5}, and PM₁₀ in all the four months, NO₂ in all months except for July, maximum 1-h O₃ in July, due mainly to the underestimations of emissions of SO₂, NO₂, primary OM, and mineral dust and to the inaccurate vertical allocation of emissions for some species such as NO₂. The performance for surface gaseous species and PM is better in July than in January mainly because of more accurate meteorological predictions and emissions in July than January. Moderate to large underpredictions are found for AOD with NMBs of -57.4% to -13.5% in the 4 months, for TOR in July with an NMB of -15.8%, and for column CO in April with an NMB of -23.0%. By contrast, moderate-to-large overpredictions occur in January and October for CO, in January, April, and October for NO₂, and in January and April for TOR. COT, CF, CWP, and CDNC are underpredicted with NMBs of -90.2% to -83.2%, -33.4% to -14.5%, -86.3% to -78.4%, and -4.0% to -2.7% in four months, respectively. Those model biases are attributed to several reasons including inaccurate emissions or their vertical allocation, inaccurate boundary conditions, and uncertainties in the satellite retrievals. The use of a coarse grid resolution also explains in part the model biases in most variables, particularly those that are highly sensitive to horizontal/vertical grid resolution.

Anthropogenic aerosols play a key role in modulating the regional climate through affecting radiative budget and cloud formation. They can decrease GSW by 6.5-12.1 W m⁻² domain-wide mean and by up to 40.5-57.2 W m⁻² and decrease T₂ by 0.04-0.08 °C domain-wide mean and by up to 0.5-0.8 °C. The decreased GSW and T₂ lead to reductions in photolysis rates for NO₂ and PBL height by up to 0.06-0.1 min⁻¹, and 83.6-130.4 m, respectively, creating a more stable atmospheric boundary layer

that can exacerbate air pollution. For comparison, these results show the consistency with published paper.

Anthropogenic aerosols can increase in-cloud CDNCs and COT through increasing aerosol number concentrations and CCN, and reduce precipitation over most of the domain during the 4 months. The large changes in those variables occur in central and eastern China where anthropogenic aerosol concentrations are the highest and the adjacent oceanic areas off the eastern coastal areas where the impacts of the long range transported pollutants are the largest. Aerosol number concentrations from anthropogenic sources are the highest in January reaching up to $8.6 \times 10^4 \text{ cm}^{-3}$ over most areas, especially over eastern China and the Sichuan Basin. Anthropogenic-induced CCN concentrations at $s=0.5\%$ at the surface are $0.6\text{-}1.4 \times 10^3 \text{ cm}^{-3}$ domain-wide mean and up to $1.0\text{-}1.6 \times 10^4 \text{ cm}^{-3}$, with higher values over regions with higher PM mass and number concentrations. Anthropogenic aerosols increase in-cloud CDNCs over most of the domain in all the four months and such increases extend eastward except for July. As a consequence of increased CDNCs, COTs increase over regions in the four months by 1.3-1.7 domain-wide mean and by up to 19.8-33.2. In contrast to changes in COT, daily precipitation decreases in most areas in the four months with reductions by up to 3.9-18.6 mm with the largest decrease in July, because anthropogenic aerosols can suppress the formation of warm rain.

The results from this work demonstrate the importance of anthropogenic aerosol direct and indirect effects in East Asia. Comparing to simulated aerosol direct and indirect effects over the U.S. and Europe (Zhang et al., 2010b, 2013b), the magnitudes of aerosol effects are larger and more robust in East Asia due to high anthropogenic aerosol concentrations. Aerosol effects reported here may have been underestimated for several reasons. First, aerosol effects on cloud feedbacks associated with convective clouds and precipitation are currently not simulated in WRF/Chem, which may cause an underestimation in the aerosol indirect effects. Second, the aerosol activation parameterization used in this work is based on Abdul-Razzak and Ghan

(2002), which tends to underestimate aerosol activation fraction, thus CDNC and COT. As shown in Zhang (2014a) and Gantt et al. (2014), using a more advanced parameterization based on Fountoukis and Nenes (2005) and its recent updates, simulated CDNC and COT agree better with observations. Additional limitations include uncertainties in model inputs such as emissions and their vertical/spatial allocations, uncertainties and/or imperfectness in the model's representations of aerosol formation processes (e.g., SOA and new particle formation) and aerosol-radiation-cloud-precipitation interactions, relatively short month-long simulation periods that may not be sufficient to characterize long-term climate-chemistry feedbacks, the use of a coarse horizontal grid resolution for model simulations, and lack of observational data for verification of simulated aerosol direct and indirect effects. Future work should focus on the above areas that can potentially improve the model performance and its fidelity in simulating meteorology, air quality, and the climatic impacts of aerosols.

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766 **Table 1.** Performance statistics of surface gaseous and PM predictions in 2008¹

	Month	Mean obs	Mean sim	MB	R	RMSE	NMB (%)	NME (%)
SO ₂ ,ppb	January	60.5	17.8	-42.6	0.0	63.7	-70.5	74.4
	April	34.4	9.8	-24.6	-0.3	32.0	-71.5	80.7
	July	22.3	10.6	-11.8	-0.1	19.5	-52.7	69.6
	October	27.5	15.0	-12.5	0.0	24.1	-45.6	62.9
NO ₂ ,ppb	January	27.5	20.6	-7.0	0.4	16.9	-25.3	48.5
	April	20.8	11.3	-9.5	0.4	15.6	-45.6	63.3
	July	13.0	13.4	0.4	0.3	18.1	3.1	87.5
	October	23.6	14.5	-9.1	0.2	16.7	-38.4	56.6
Maximum	January	31.7	33.5	1.9	0.6	11.0	5.9	23.6
1-hr O ₃ , ppb	April	42.5	36.2	-6.3	0.2	20.5	-14.8	38.5
	July	23.8	28.7	4.9	-0.1	18.9	20.6	55.7
	October	28.8	30.9	2.0	0.3	14.7	7.0	39.8
PM ₁₀ ,µg m ⁻³	January	129.7	57.2	-72.6	0.3	105.1	-55.9	63.3
	April	103.52	42.1	-61.4	0.1	79.4	-59.3	62.6
	July	83.2	50.5	-32.8	0.2	49.9	-39.4	48.1
	October	87.5	60.4	-27.1	0.2	55.4	-31.0	51.5
PM _{2.5} ,µg m ⁻³ (TEOM)	January	52.8	27.9	-25.0	0.4	55.3	-47.2	63.3
	April	122.2	34.6	-87.6	0.2	151.0	-71.7	76.6
	July	72.4	63.6	-8.8	0.5	47.9	-12.2	49.7
	October	60.7	31.9	-28.9	0.5	62.7	-47.5	57.2
PM _{2.5} ,µg m ⁻³ (Medium flow)	January	105.4	38.7	-66.7	0.5	103.1	-63.3	66.6
	April							
	July	67.6	62.9	-4.7	0.8	27.6	-7.0	30.7
	October							
PM _{2.5} ,µg m ⁻³ (Small flow)	January	88.9	32.0	-56.9	-0.6	81.7	-64.0	72.1
	April	81.4	37.7	-43.7	0.9	49.4	-53.6	53.6
	July	70.3	57.3	-13.0	0.3	30.5	-18.5	36.5
	October	56.7	27.8	-28.9	0.3	39.9	-50.9	50.9
SO ₄ ²⁻ ,µg m ⁻³	July	25.3	13.7	-11.6	0.6	18.8	-45.9	59.7
NH ₄ ⁺ ,µg m ⁻³	July	7.0	8.7	1.6	0.7	5.1	23.2	54.8
OM,µg m ⁻³	July	8.7	5.1	-3.6	0.3	4.7	-41.1	45.5
NO ₃ ⁻ ,µg m ⁻³	July	13.0	13.1	0.1	0.6	9.7	1.0	56.9
EC,µg m ⁻³	July	3.6	3.9	0.2	0.5	1.9	6.8	43.5

767 ¹MB-mean bias, R-correlation coefficient, RMSE-root mean square error, NMB- normalized mean
 768 bias, and NME-normalized mean gross error.

Table 2. Performance statistics of column predictions in 2008

	Month	Mean obs	Mean sim	MB	R	RMSE	NMB (%)	NME (%)
CO ²	January	19.6	21.6	2.0	0.5	7.3	10.2	22.1
	April	23.9	18.4	-5.5	0.7	7.5	-23.0	26.8
	July	16.8	16.7	-0.1	0.5	5.7	-0.6	26.4
	October	18.4	20.4	2.0	0.8	4.3	11.0	16.2
NO ₂ ²	January	23.4	57.5	34.1	0.8	65.9	146.2	150.2
	April	20.1	27.4	7.3	0.8	18.8	36.1	53.2
	July	17.4	17.3	-0.1	0.7	15.6	-0.7	43.6
	October	17.8	32.0	14.2	0.9	32.6	80.0	87.5
TOR, Dobson	January	30.7	41.3	10.6	0.8	11.4	34.5	34.5
	April	37.1	51.8	14.7	0.7	15.6	39.5	39.5
	July	42.1	35.4	-6.7	0.6	9.8	-15.8	20.1
	October	33.8	32.9	-0.9	0.8	3.9	-2.8	9.3
AOD	January	0.3	0.2	-0.1	0.6	0.2	-38.5	49.7
	April	0.5	0.2	-0.3	0.5	0.3	-57.4	58.1
	July	0.3	0.2	-0.1	0.5	0.2	-23.2	47.5
	October	0.3	0.2	0.0	0.7	0.1	-13.5	38.5
COT	January	36.0	4.7	-31.4	0.7	35.1	-87.1	87.1
	April	29.6	3.7	-25.8	0.7	27.6	-87.3	87.4
	July	32.9	5.5	-27.4	0.3	28.8	-83.2	83.6
	October	33.9	3.3	-30.6	0.6	32.5	-90.2	90.3
CF, %	January	68.6	45.6	-22.9	0.6	36.5	-33.4	44.2
	April	57.1	38.1	-19.0	0.7	26.2	-33.2	39.3
	July	67.8	58.0	-9.8	0.6	36.2	-14.5	40.0
	October	54.4	39.0	-15.4	0.6	28.0	-28.3	41.8
CWP, g m ⁻²	January	119.0	23.3	-95.8	0.7	107.1	-80.4	80.5
	April	99.0	16.5	-82.5	0.7	88.3	-83.4	83.4
	July	119.4	25.8	-93.6	0.4	100.7	-78.4	79.8
	October	113.1	15.5	-97.7	0.7	103.5	-86.3	86.5
CDNC, cm ⁻³	January	155.9	151.8	-4.2	0.9	36.8	-2.7	11.4
	April	160.7	154.2	-6.4	0.9	38.7	-4.0	12.0
	July	115.9	111.5	-4.4	0.9	29.8	-3.8	12.9
	October	133.1	127.9	-5.2	0.9	36.6	-3.9	14.2

²The unit for Mean obs, Mean sim, MB, and RMSE are 1×10^{-17} molecules cm⁻² for CO, 1×10^{-14} molecules cm⁻² for NO₂.

FIGURES

772 **Figure 1.** Overlay of observed and simulated surface concentrations of SO_2 , NO_2 , 1-h maximum hourly O_3 , and PM_{10} in 2008 at a 36-km-grid
773 resolution. Circles indicate the observations

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Figure 2. Time series of observed and simulations concentrations of SO₂, NO₂, and PM₁₀ at four sites (Beijing, Shanghai, Guangzhou, Xi'an) and PM_{2.5} at four sites (THU, MY, BYD, SD) in China in 2008. Note that while PM₁₀ data are available for all the four months, PM_{2.5} data are only available in January and July.

778 **Figure 3.** Spatial distributions of simulated and observed mean monthly column abundances of CO
779 and NO₂, tropospheric ozone residual (TOR), and aerosol optical depth (AOD) in 2008.
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Figure 4. Effects of anthropogenic aerosols on T2, net shortwave flux at the surface (GSW), NO₂ photolysis rates, and planetary boundary layer (PBL) height in 2008.

Figure 5. Simulated monthly-mean vertical profiles of BC, PM_{2.5} concentrations and the changes of temperature due to elevated anthropogenic aerosols in January, April, July and October 2008 at four sites: Beijing, Shanghai, Guangzhou, Xi'an.

787 **Figure 6.** Effects of anthropogenic aerosols on aerosol number concentration, cloud condensation nuclei (CCN) at $s = 0.5\%$, in-cloud cloud droplet
788 number concentrations (CDNCs), cloud optical thickness (COT), and precipitation in 2008.