Incorporation of New Particle Formation and Early Growth Treatments into WRF/Chem: Model Improvement, Evaluation, and Impacts of Anthropogenic Aerosols over East Asia

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Abstract

New particle formation (NPF) provides an important source of aerosol particles and cloud condensation nuclei, which may result in enhanced cloud droplet number concentration (CDNC) and cloud shortwave albedo. In this work, several nucleation parameterizations and one particle early growth parameterization are implemented into the online-coupled Weather Research and Forecasting model coupled with chemistry (WRF/Chem) to improve the model's capability in simulating NPF and early growth of ultrafine particles over East Asia. The default 8-bin over the size range of 39 nm - 10 μ m used in the Model for Simulating Aerosol Interactions and Chemistry aerosol module is expanded to the 12-bin over 1 nm - 10 μ m to explicitly track the formation and evolution of new particles. Although model biases remain in simulating H₂SO₄, condensation sink, growth rate, and formation rate, the evaluation of July 2008 simulation identifies a combination of three nucleation parameterizations (i.e., COMB) that can best represent the atmospheric nucleation processes in terms of both surface nucleation events and the resulting vertical distribution of ultrafine particle concentrations. COMB consists of a power law of Wang et al. (2011) based on activation theory for urban areas in planetary boundary layer (PBL), a power law of Boy et al. (2008) based on activation theory for non-urban areas in

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PBL, and the ion-mediated nucleation parameterization of YU10 for above PBL. The application and evaluation of the improved model with 12-bin and the COMB nucleation parameterization in East Asia during January, April, July, and October in 2001 show that the model has an overall reasonably good skill in reproducing most observed meteorological variables and surface and column chemical concentrations. Relatively large biases in simulated precipitation and wind speeds are due to inaccurate surface roughness and limitations in model treatments of cloud formation and aerosol-cloud-precipitation interactions. Large biases in the simulated surface concentrations of PM₁₀, NO_x, CO, SO₂, and VOCs at some sites are due in part to possible underestimations of emissions and in part to inaccurate meteorological predictions. The simulations of 2001 show that anthropogenic aerosols can increase aerosol optical depth by 64.0 - 228.3%, CDNC by 40.2 - 76.4%, and cloud optical thickness by 14.3 - 25.3%; they can reduce surface net shortwave radiation by up to 42.5 – 52.8 W m⁻², 2-meter temperature by up to 0.34 - 0.83 °C, and PBL height by up to 76.8 – 125.9 m. Such effects are more significant than those previously reported for the U.S. and Europe.

Key words: New particle formation, Particle early growth, WRF/Chem, Aerosol direct effects, Aerosol indirect effects, East Asia

1. Introduction

Atmospheric ultrafine particles in the nanometer size range have adverse impacts on visibility and human health due to their small size. They can also impact climate by directly absorbing and scattering of radiation, and indirectly modulating cloud and precipitation formation after further growth to cloud condensation nuclei (CCN). New particle formation (NPF) processes via nucleation provide an important source of ultrafine particles and CCN on global and regional scales (Yu, 2000; Yu et al., 2008, 2012; Y. Zhang et al., 2010a, b, 2012; R. Zhang et al., 2012a). An increased aerosol number concentration resulting from NPF will enhance cloud droplet number concentration (CDNC) and cloud shortwave albedo (Twomey, 1977; Y. Zhang et al., 2012a). A number of nucleation mechanisms have been reported in the literature based on laboratory and field studies. They can be generally grouped into five classes based on the

compounds participated in the nucleation process: (1) $\text{H}_2\text{O}+\text{H}_2\text{SO}_4$ binary nucleation (Kulmala et al., 1998); (2) $\text{H}_2\text{O}+\text{H}_2\text{SO}_4+\text{NH}_3$ ternary nucleation (Merikanto et al., 2007, 2009); (3) $\text{H}_2\text{O}+\text{H}_2\text{SO}_4+\text{Ions}$ ion-mediated nucleation (Yu et al., 2008; Yu, 2010); (4) $\text{H}_2\text{O}+\text{H}_2\text{SO}_4+\text{IO}_x$ Iodine-induced nucleation (Jimenez et al., 2003); (5) nucleation involving organic compounds (Zhang et al., 2004; Fan et al., 2006; Kulmala et al., 2013). Some of those nucleation parameterizations have been implemented and tested in both 0-D and 3-D atmospheric models (e.g., Pierce and Adams, 2007; Yu et al., 2008, 2012; Wang and Penner, 2009; Y. Zhang et al., 2010a, b, 2012).

A major challenge in accurately simulating ultrafine particles lies in the model treatment of the growth of newly formed nuclei from 1 nm to measurable size ranges of 3-10 nm (i.e., the so-called early growth processes). The nucleation of gas molecules forms clusters; some of which are lost via coagulation with preexisting particles before they can grow and the rest can further grow into the measurable sizes. The early growth processes include activation of inorganic clusters by soluble organic molecules for condensational growth, heterogeneous nucleation of insoluble organic vapors on inorganic clusters, and chemical reactions of organic molecules at surfaces of small clusters (Kulmala, 2003). While nucleation rate (also called the “real” nucleation rate) refers to the rate at which stable nuclei are formed and cannot currently be measured, new particle formation rate (FR) (also called the “apparent” nucleation rate) refers to the rate at which detectable particles are formed, and must be smaller than the nucleation rate. Nucleation rates can be inferred indirectly by measuring the FR rate of particles with a diameter of 3 nm and by extrapolating this down to the stable nuclei size. By analyzing the competition between stable nuclei growth and sink by background aerosols, Kerminen and Kulmala (2002) derived a simple expression (so-called K-K equation, KK02) to relate the nucleation rate and FR at larger size. Lehtinen et al. (2007) (LE07) improved

KK02 by providing a more accurate dependence of the coagulation sink (CoagS) on particle size and eliminating prior knowledge of the condensing vapors (i.e., replacing condensation sink (CS) by CoagS in calculating FR values).

Although significant progress has achieved over the past several decades, there remain large uncertainties in the nucleation mechanisms and parameterizations used in 3-D atmospheric models. In particular, various nucleation parameterizations used in these models give different NPF rates (Zhang et al., 2010a, b, 2012a). Aerosol indirect radiative forcing calculated from various treatments of nucleation for aerosols can range from -1.22 to -2.03 W m^{-2} (Wang and Penner, 2009). No generalized mechanism or parameterization has been identified to be appropriate in representing NPF under various atmospheric conditions. This is because those nucleation parameterizations were typically derived based on limited measurements under specific conditions but large differences exist in the NPF rates between urban and forest/mountain sites under different atmospheric conditions. Further, most models do not treat the early growth of ultrafine particles, introducing large uncertainties in simulated concentrations of aerosol and CCN, as well as their impact on air quality and climate.

In this work, several nucleation parameterizations and one algorithm for particle early growth are implemented into an online-coupled meteorology-chemistry model, the Weather Research and Forecasting model with chemistry (referred to as WRF/Chem) to simulate NPF and their impacts on air quality and climate. The objectives of this work are to improve the model's representation of the formation and growth of the ultrafine particles with diameters less than 20-nm (also referred to as the nucleation mode particles) and evaluate the improved WRF/Chem's capability in simulating meteorology, air quality, and their interactions, in particular, impacts of anthropogenic aerosols on climate. East Asia is selected as a testbed for several reasons. First, East Asia has

experienced continuous rapid economic and population growth, industrialization, and urbanization during the past three decades, which led to large emissions of air pollutants thus significant degradation of air quality on regional and global scales. Second, the high concentrations of air pollutants such as particulate matters and their gaseous precursors permit a rigorous test of the robustness of model's representations of atmospheric processes under the extreme conditions; they also generate strong chemistry-climate feedbacks that affect cloud and precipitation formation. Further, East Asia has special topography and geographical locations and distinct climatic conditions. Thus, East Asia provides a supreme testbed for improving and testing the performance of WRF/Chem.

2. Model system, evaluation protocols, and observational datasets

2.1 Default Treatments in WRF/Chem

WRF/Chem version 3.3.1 is applied in this study. WRF/Chem offers multiple physics and chemistry options to simulate a variety of atmospheric processes. The major physics and chemistry options used in this work are summarized in Table 1. The horizontal grid resolution used is 36-km, and the vertical resolution is 23 layer from 1000 mb to 50 mb. The major physics options used include the modified Purdue Lin microphysics module (Lin et al., 1983), the Rapid Radiative Transfer Model (RRTM) long-wave radiation scheme (Mlawer et al., 1997), the Goddard short-wave radiation scheme (Chou and Max, 1994), the National Center for Environmental Prediction, Oregon State University, Air Force, and Hydrologic Research Lab's (NOAH) land-surface module (Tewari et al., 2004), the Yonsei University (YSU) Planetary Boundary Layer (PBL) scheme (Hong et al., 2006), the Grell-Devenyi cumulus parameterization (Grell and Devenyi, 2002), the Fast-J photolysis rate scheme (Wild et al., 2000). The gas-phase chemistry used the Carbon-Bond Mechanism version Z (CBMZ, Zaveri and Paters, 1999). The aqueous-phase chemistry is based on the Carnegie Mellon University (CMU)

mechanism (Fahey and Pandis, 2001). The aerosol module is based on the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008). MOSAIC in WRF/Chem v3.3.1 treats all major aerosol processes except for the formation of secondary organic aerosols (SOA) when CBMZ is selected as the gas-phase chemical mechanism. Although the volatility basis set (VBS) approach is available in WRF/Chem 3.3.1 for simulation of SOA when SAPRC99 is selected as the gas-phase chemical mechanism and MOSIAC is selected as an aerosol module, the SAPRC99-MOSAIC/VBS option in this version of WRF/Chem does not include aqueous-phase chemistry and cloud-aerosol interactions (Shrivastava et al., 2011). While other aerosol modules such as MADE/SORGAM treat SOA formation, they do not permit the simulation of both aerosol direct and indirect effects, which is one of the main foci of this work. The combination of CBMZ-MOSAIC-CMU is selected in this work, because this is the only combination available in WRF/Chem version 3.3.1 to simulate both aerosol direct and indirect effects. Activation of aerosol to form CCN is based on the parameterization of Abdul-Razzak and Ghan (Abdul-Razzak and Ghan, 2002). The particle size distribution in MOSAIC is simulated using eight size bins between 39-nm and 10- μm . More details about the chemistry, aerosol, and cloud treatments can be found in several studies (e.g., Grell et al., 2005; Chapman et al., 2009; and Zhang et al., 2010c).

2.2 Nucleation and early growth parameterizations tested in WRF/Chem

Table 2 summarizes the parameterizations of the default nucleation parameterization used in WRF/Chem, and several new nucleation parameterizations and one particle early growth algorithm (i.e., LE07) to be examined in this study. Wexler et al. (1994) (WE94) is the default module used in WRF/Chem. Unlike other parameterizations that explicitly calculate the nucleation rate, WE94 (i.e.,) calculates the critical concentration of H_2SO_4 in $\mu\text{g m}^{-3}$ based on temperature and relative humidity and does

not explicitly calculate nucleation rates, J. The number of particles produced by WE94 is thus somewhat arbitrary, because it depends on the smallest size bin of the model. One can estimate nucleation rates from WE94, but it also depends on the assumed smallest size bin of the model. To explicitly simulate nucleation, several sets of nucleation parameterizations are implemented into WRF/Chem in this work. The first set is the power law of Wang et al. (2011) (WA11), which was derived based on the measurement data at an urban site in Beijing, China. WA11 contains three power laws corresponding to the activation, kinetic, and thermodynamic theories which depend on the power orders of 1, 2, and 3 of the concentrations of sulfuric acid (H_2SO_4) and are referred to as WA11_ACTI, WA11_KINE, and WA11_THEM, respectively. The second set is the parameterization of Boy et al. (2008) (BO08), which was derived based on measurement data obtained in mountain and forest areas. BO08 contains two power laws corresponding to the activation and kinetic theories which depend on the power orders of 1 and 2, and are referred to as BO08_ACTI and BO08_KINE, respectively. The third set is based on the ion-mediated nucleation parameterization of Yu (2010) (YU10), which contains lookup tables derived from a kinetic model that explicitly solves the interactions among ions, neutral and charged clusters, vapor molecules, and pre-existing particles (e.g., Yu, 2006, 2010). YU10 is suitable for the whole troposphere and has been shown to replicate new particle formation rates in both upper and lower troposphere (Yu et al., 2010).

MOSAIC does not simulate all the early growth processes of nano particles from ≤ 1 nm to detectable size ranges (≥ 3 nm) such as activation of inorganic clusters for condensational growth, heterogeneous nucleation of insoluble organic vapors, and chemical reactions of organic molecules. The semi-empirical formulation of LE07 is thus implemented in WRF/Chem to avoid the simulation of detailed nucleation and early growth of stable nuclei and to compare the simulated formation rates directly with the

192 observed data from the field experiments. LE07 was derived by estimating the FR,
193 nucleation mode growth rate (GR) and scavenging rate onto background particles (i.e.,
194 CoagS). The derived formulation provides a means to account for the aforementioned
195 initial growth processes that are not treated in the aerosol modules in WRF/Chem yet
196 avoid detailed nucleation and initial growth models. It also allows a comparison of the
197 simulated NPF formation rates directly with observational data. LE07 is coupled with
198 various nucleation parameterizations implemented in WRF/Chem to explicitly track the
199 evolution of new particles resulted from NPF events. The new particle formation rates for
200 the smallest four bins (i.e., Bins 1-4) are adjusted with the calculated GR from LE07,
201 following the approach used in the Community Atmospheric Model version 5 (Liu et al.,
202 2012)) (although CAM5 uses KK02, which is an older version of the growth scheme).
203 The lowest size bin for the default 8-bin structure in MOSAIC has diameter ranges of 39-
204 78 nm, which are much larger than the reported size (around 1-2 nm) of newly formed
205 particles (Kulmala et al., 2004, 2013). This 8-bin structure is thus insufficient to the
206 accurate representation of the early growth of nuclei to larger particles in the nucleation
207 mode (1-20 nm). In this study, the original 8 size structure bin is thus first modified, then
208 extended to 12 size structure bin with the first 4 bins for ultrafine particles with the
209 diameters of 1-20 nm to accurately simulate the formation and early growth of the
210 ultrafine particles. In order to ensure consistent bin boundaries for overlapped bins
211 between 8-bin and 12-bin structures (i.e., bins 1-8 in 8-bin structure and bins 5-12 in 12-
212 bin structure), the default 8 bins are modified (referred to as modified 8-bin thereafter).
213 The extension of 8-bin to 12-bin increases the computational cost by about 70%. While
214 such an extension adds 4 bins to represent particles with diameters of 1-20 nm that can be
215 afforded by 3-D atmospheric models, they may still be too coarse to accurately simulate
216 the nucleation and initial growth of nuclei and detectable nano particles in the lower end

of the size spectrum. The use of LE07 would provide a compromise between the numerical accuracy needed for an accurate simulation of the nuclei formation and growth processes, and the computational efficiency required for large scale 3-D atmospheric models. Table 3 summarizes the size ranges of each bin in the 12-bin structure along with the original and updated 8-bin structures. The new 12-bin structure is used in all WRF/Chem simulations.

2.3 Model configurations and simulation setup

Two simulation periods have been selected with different purposes (1) four representative months in 2001 (i.e., January, April, July, and October, one for each season) and (2) July in 2008. The 2001 episode is selected because it represents a heavy-polluted episode (K. Wang et al., 2009) and the aerosol-meteorology interactions are potentially significant. In addition, according to previous studies, several dust storms have been reported in 2001 (Zhang et al., 2003), and also NPF in anthropogenic plumes advecting from East Asia has been observed in 2001 during the National Atmospheric and Space Administration (NASA) Transport and Chemical Evolution over Pacific (TRACE-P) mission (Weber et al., 2003). However, no nucleation measurements were performed in China in 2001. Limited nucleation observations were made in China after 2004. In particular, nucleation relevant parameters such as FR, CS, and particle number size distribution (N-PSD) of 3-900 nm were collected during July-September 2008 during the Campaigns of Air Quality Research in Beijing and Surrounding Region 2008 (CAREBeijing2008) (Wang et al., 2011; Wang, 2012). To evaluate appropriateness of various nucleation parameterizations and identify the most suitable one for 2001 simulations, July 2008, one month before 2008 Olympics in Beijing China, is therefore selected. Several WRF/Chem simulations are carried out using individual nucleation parameterizations BO08_ACTI, WA11_ACTI, and YU10 and a combination of them for

July 2008 to provide an initial testing of various nucleation parameterizations to access the model's capability in reproducing the NPF event and early growth processes. Based on the July 2008 simulation results, an optimal set of nucleation parameterizations among the four parameterizations tested is identified and then applied to a multi-month baseline simulation in 2001. In addition to the 2001 baseline simulations, sensitivity simulations without anthropogenic aerosols are carried out to quantify the impacts of anthropogenic aerosols on simulated air quality and climate. In the sensitivity simulations, the emissions of primary aerosols and the formation pathways of secondary aerosols are turned off following Zhang et al. (2010c).

For all the simulations, the meteorological initial and boundary conditions are based on the National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data. The chemical initial and boundary conditions are based on Liu et al. (2010) over East Asia. The anthropogenic emissions are based on an updated version of TRACE-P over China for 2001 simulations (Carmichael et al., 2003), and the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) emission for 2008 simulations (Zhang et al., 2009). The emitted primary PM species include primary elemental carbon (EC), organic carbon (OC), sulfate (SO_4^{2-}), nitrate (NO_3^-), other unknown inorganic $\text{PM}_{2.5}$, and coarse PM. The original PM emissions are based on 3-mode, which has been mapped into the bins 5-12 in the 12-bin sectional size structure using the internal conversion code in WRF/Chem. The primary PM emissions in bins 1-4 are assumed to be zero. The emissions of biogenic VOCs are simulated online based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 (Guenther et al., 2006). Dust emissions are simulated online based on the modified dust entrainment and deposition (DEAD) scheme of Zender et al. (2003) by Wang et al. (2012). Sea-salt emissions are simulated based on Gong et al. (2002).

2.4 Evaluation protocols and observational database

Table 4 summarizes observational databases used in model evaluation. The surface network datasets include the National Climatic Data Center (NCDC), the Air Pollution Index (API) in mainland China, data from Japan, Taiwan, and Hong Kong, PM_{2.5} and composition data from Tsinghua, and other data reported in the literatures. Satellite datasets include Scanning Imaging Absorption spectrometer (SCIAMACHY), the Total Ozone Mapping Spectrometer (TOMS), the Ozone Monitoring Instrument (OMI), the Measurements of Pollution in the Troposphere (MOPITT), and Terra Moderate-resolution Imaging Spectroradiometer (MODIS). The meteorological variables evaluated include daily precipitation (Precip), temperature at 2-m (T2), pressure (P), specific humidity (Q2), wind speed (WS10), and wind direction (WD10) that are most relevant to air pollutions. The chemical variables evaluated include the concentrations of H₂SO₄, NO, VOCs, PM₁₀, PM_{2.5}, and PM composition (NH₄⁺, SO₄²⁻, NO₃⁻, Na⁺, and Cl⁻); the column abundance of SO₂, O₃, NO₂, and CO, aerosol optical depth (AOD), and cloud variables (cloud optical thickness (COT), cloud water path (CWP), and precipitating water vapor (PWV)). Additional variables for nucleation evaluation include FR, growth rate (GR), CS, and N-PSD. The performance evaluation of WRF/Chem predictions is conducted in terms of statistical, spatial, and temporal comparisons following the protocols of Zhang et al. (2010b, 2013). Observed and simulated values of nearly all meteorological and chemical variables are compared through the calculation of statistical measures including the normalized mean bias (NMB), the normalized mean error (NME), the mean bias (MB), the root mean square error (RMSE), and correlation coefficients (R).

3. Testing of nucleation and early growth parameterizations for July 2008

Table 5 summarizes observed NPF rates at various sites in China during 2004-2012. The observed NPF rates of 3-nm particles range from 1.1 to 81.4 cm⁻³ s⁻¹ in urban

areas (Wu et al., 2007; Yue et al., 2009; Gao et al., 2011; Wang et al., 2013; Herrmann et
 al., 2013), $0.7\text{--}72.7\text{ cm}^{-3}\text{ s}^{-1}$ in rural areas (Gao et al., 2009; Shen et al., 2011; Wang et al.,
 2013), and $0.5\text{--}6.9\text{ cm}^{-3}\text{ s}^{-1}$ in coastal areas (Liu et al., 2008a, b; Guo et al., 2012). The
 initial testing of the default nucleation parameterization (i.e., WE94) shows a domainwide
 average NPF rate of $2 \times 10^{-5}\text{ cm}^{-3}\text{ s}^{-1}$ assuming arbitrarily a minimal diameter of 1.5 nm,
 which is too small comparing with observations. Because of this and also the reasons
 stated in Section 2.2, the WE94 parameterization is therefore not included in the final
 simulations for July 2008. The initial testing of the three power laws of WA11 and two
 power laws of BO08 shows that WA11_KINE, WA11_THEM, and BO08_KINE give
 NPF rates that are much greater than the above observed ranges. Therefore, only
 WA11_ACTI and BO08_ACTI are further examined from the first two sets of
 parameterizations in the final simulations for July 2008. In addition, given distinct
 differences in FR values in urban, rural, and coastal areas (see Table 5), using a
 combination of several parameterizations may provide the most realistic representation of
 the NPF events and FR values under various atmospheric conditions. Therefore, based on
 various land surfaces, a combination of three parameterizations is used to simulate NPF
 under different atmospheric conditions in the final simulations, i.e., WA11_ACTI for
 urban areas in PBL, BO08_ACTI for non-urban areas in PBL, and YU10 for above PBL
 (referred to as COMB). The fraction of land index 1 (urban and built-up land) based on
 the surface land index of the U. S. Geological Survey is used to determine the use of
 WA11_ACTI over urban areas.

Table 6 summarizes performance statistics for meteorological variables and
 chemical concentrations from the simulation COMB (the results from other simulations
 are similar, thus not shown). COMB simulates T2, P, Q2, and WD10 well with NMBs
 within $\pm 5\%$. WS10 is overpredicted by 49.1%, due mainly to an inaccurate

representation of the surface roughness (Mass and Owens, 2010). Daily precipitation is underpredicted by 21% domainwide, due in part to underpredictions in cloud fraction (by 34.4%). The overpredictions in simulated WS10 may contribute in part to the underpredictions of SO₂, NO₂, VOCs, and PM₁₀, therefore the accuracy of the simulated NPF event. The underpredictions in precipitation may contribute in part to the underpredictions of the wet scavenging and deposition of newly-formed particles, which also affect the accuracy of the simulated NPF event, as discussed later.

Figure 1 shows the spatial distributions of the surface FR of bin-1 from WRF/Chem simulations using various nucleation parameterizations. The maximum FR value predicted by YU10 near the surface over the whole domain is $\sim 8.2 \text{ cm}^{-3} \text{ s}^{-1}$, which may underestimate FR values. On the other hand, WA11_ACTI gives the maximum value of around $251.6 \text{ cm}^{-3} \text{ s}^{-1}$, which may overpredict FR values. BO08_ACTI (range from 0 - $42.7 \text{ cm}^{-3} \text{ s}^{-1}$) and COMB (range from 0 - $42.9 \text{ cm}^{-3} \text{ s}^{-1}$) give similar FR values on the surface, which are within the range of the observations. Figure 2 compares the zonal mean values of FR of bin-1 using various parameterizations. Since WA11_ACTI was developed based on measurements at a heavily-polluted urban site in Beijing, it gives very high values ($\sim 10.6 \text{ cm}^{-3} \text{ s}^{-1}$) over the whole zonal areas (see Figure 2b). BO08_ACTI predicts FR values of 1.0 to $4.8 \text{ cm}^{-3} \text{ s}^{-1}$ in the lower portion of the atmosphere (1000-500 mb), but much smaller values ($< 1 \text{ cm}^{-3} \text{ s}^{-1}$) in upper atmosphere ($< 500 \text{ mb}$). Therefore, it is unlikely for BO08_ACTI to produce the high measured ultrafine particle number concentrations in upper atmosphere reported by Yu et al. (2008) (see Figure 2(f)). The ionization rate is high in upper atmosphere (see Figure 2e) due to strong galactic cosmic rays, leading to high FR values (Figure (c)) predicted by YU10 thus high measured ultrafine particle number concentrations. However, YU10 gives low FR values in lower atmosphere due to the low ionization rate. Among the four simulations, COMB is the

only one that can capture well the vertical spatial patterns of the observed ultrafine particle number concentrations shown in Figure 2 (f).

The observed N-PSD are quite different during polluted ($\text{PM}_{10} > 150 \mu\text{g m}^{-3}$) and non-polluted days ($\text{PM}_{10} \leq 150 \mu\text{g m}^{-3}$) at an urban site of Beijing (Yue et al., 2009). This is because NPF events can occur more easily at a lower background PM level (non-polluted), which would lead to higher number concentrations of ultrafine particles ($< 0.1 \mu\text{m}$) compared to polluted days (Wu et al., 2007; Yue et al., 2009). The simulated N-PSD is thus analyzed for polluted ($\text{PM}_{10} > 150 \mu\text{g m}^{-3}$) and non-polluted days ($\text{PM}_{10} \leq 150 \mu\text{g m}^{-3}$). Figure 3 compares predicted monthly-averaged N-PSD during polluted and non-polluted days at an urban site, the campus of Peking University (PKU) (39.99°N , 116.31°E), using WRF/Chem without and with various nucleation parameterizations including BO08_ACT1, WA11_ACT1, YU10, and COMB. Since no N-PSD observations during polluted and non-polluted days were readily available in the literature during July 2008, lognormal fits of observed N-PSD of summer 2006 of Yue et al. (2009) at the same site are used for a qualitative comparison in Figure 3(f). The observed average N-PSD in summer 2006 showed significant shift to larger sizes on polluted days than non-polluted days. Such shifts in simulated N-PSD are captured well only by simulations with nucleation treatments. Comparing to the N-PSD predicted by WRF/Chem without simulating nucleation, WRF/Chem with various nucleation parameterizations can explicitly track the PSD for particles with diameter less than $0.01 \mu\text{m}$ (i.e., nucleation mode) resulted from NPF and early growth processes. While the simulation with WA11_ACT1 tends to overpredict the number concentrations of nucleation mode under non-polluted conditions, the simulations with the other three parameterizations can replicate to some degrees the differences in the PSD for nucleation mode particles under polluted and non-polluted conditions with the closest agreement by Yu (2010).

367 Gaseous sulfuric acid, H_2SO_4 , is a key component for nucleation, and it is
368 predominantly produced through the oxidation of SO_2 by hydroxyl radical (OH) in the
369 presence of oxygen (O_2) and water vapor (H_2O). In the presence of solar radiation and
370 H_2O , OH radicals are produced from ozone (O_3) which is controlled by its major
371 precursors such as nitrogen oxides (NO_2) and volatile organic compounds (VOCs).
372 Therefore, those gas-phase species and their chemistry are closely linked to the
373 occurrence of the nucleation events. In addition, the background PM concentration that
374 reflects the level of pollution affects the occurrence of NPF events and the FR values.
375 Other important parameters describing NPF include the FR and GR of new particles, as
376 well as the CS, which directly determine the number concentrations of new particles.
377 Figure 4 shows the time series for H_2SO_4 , CS, FR, PM_{10} , SO_2 , O_3 , and NO_2 at the surface
378 simulated using various nucleation parameterizations along with observations at the same
379 urban site, PKU, in Beijing in July 2008. Table 6 summarizes the performance statistics
380 for concentrations of SO_2 , O_3 , NO_2 , several VOCs species (i.e., toluene (TOL), xylene
381 (XYL), Isoprene), and PM_{10} from the simulation with COMB (the results for all other
382 parameterizations are similar). Table 7 summarizes the corresponding performance
383 statistics for H_2SO_4 , CS, GR, and FR by the four simulations with various nucleation
384 parameterizations.

385 As shown in Figure 4 and Table 7, the number concentrations of H_2SO_4 are
386 significantly overpredicted with NMBs of 739.3-816% from the simulations with various
387 nucleation parameterizations using all observed data during July 2008, likely due to an
388 overestimate of the gas-phase oxidation rates of SO_2 to H_2SO_4 as a result of
389 overestimated atmospheric oxidation capacities. On July 12, 17, and 30, the reported peak
390 observed number concentrations of H_2SO_4 are 7.1×10^6 , 11.2×10^6 , and $8.0 \times 10^6 \text{ cm}^{-3}$,
391 respectively (Wang, 2012), which are closer to the simulated number concentrations of

392 H_2SO_4 of $(3.6\text{-}3.9) \times 10^7 \text{ cm}^{-3}$ by various nucleation parameterizations. The limited
 393 observed data itself may also introduce some uncertainties in the model evaluation. Wang
 394 (2012) also reported some uncertainties associated with the measurements of number
 395 concentrations of H_2SO_4 due to a short atmospheric lifetime of H_2SO_4 . Zheng et al.
 396 (2011) reported this uncertainty could be as high as 36%. The mixing ratios of key
 397 precursor SO_2 are underpredicted at this site (see Figure 4) and also domainwide with
 398 NMB of -44.7% (see Table 6). The mixing ratios of O_3 are overpredicted by 18.6% at this
 399 site (see Figure 4 and Table 6). Those model biases are closely related. For example, the
 400 overprediction of O_3 may have produced too much OH radicals, which may lead to
 401 unrealistically high conversion of SO_2 to H_2SO_4 , leading to underpredictions of SO_2 but
 402 overpredictions of H_2SO_4 . The atmospheric oxidation capability in the ambient
 403 environment in Beijing during July 2008 may not be as strong as what WRF/Chem
 404 predicts. The mixing ratios of both VOCs and NO_x control O_3 formation in megacities
 405 (such as Beijing and Shanghai) in China (Wang et al., 2006; Duan et al., 2008; Ran et al.,
 406 2009; Cai et al., 2010). The statistics in Table 6 indicate that, in both Beijing and
 407 Shanghai, the mixing ratios of anthropogenic VOCs (e.g., TOL and XYL) are
 408 significantly underpredicted with NMBs of -80.9 to -54.2% and -70.0% to -48.9%,
 409 respectively, but those of the typical biogenic VOCs (e.g., isoprene) are overpredicted by
 410 MEGAN2 (22.1-115.4%), which explains in part the overpredictions of O_3 . Possible
 411 reasons for the underprediction of the mixing ratios of anthropogenic VOCs include the
 412 underestimations of their emissions, the use of the coarse resolution that cannot resolve
 413 the high emissions in urban areas, and the overpredictions of WS10. The mixing ratios of
 414 NO_2 are generally underpredicted at the PKU site (see Figure 4) and also domainwide
 415 with an NMB of -36% (see Table 6). Thus, in addition to the overpredictions of biogenic
 416 VOCs, the overpredictions of O_3 in both cities may be due to insufficient titration as a

result of the underprediction of NO₂ mixing ratios. Large uncertainties exist in emissions of NO, which may help explain in part the underpredictions of NO₂. In addition to the possible overpredictions of the conversion rate of SO₂ to H₂SO₄, the large underprediction of CS with NMBs of -71.1% to -69.1% may be another reason for the overprediction of H₂SO₄ at this site. The large underprediction of CS is mainly caused by the underpredictions of preexisting PM concentrations (as reflected in NMBs of PM_{2.5} and PM₁₀ in Table 6). As shown in Table 7, the overall GR is largely underpredicted with NMBs of -77.1% to -60.6% for the NPF events at this site. Several reasons may contribute to the underpredictions of GR at this site, e.g., the underpredictions of CS, missing additional condensable vapors (e.g., organics), and the use of a coarse grid resolution. In particular, the concentrations of organic condensable organics are high in Beijing areas as indicated by the observed very high organic aerosol concentrations reported by Zhang et al. (2007) and Jimenez et al. (2009). Wang (2012) reported that the condensation of H₂SO₄ can only explain 2-16% of the observed total growth rate of fine particles during July –September 2008, and most of the GR may be caused by the condensable organic compounds. Uncertainties in the observation-derived GR values (Wang et al., 2011) may also contribute to the uncertainties in the model evaluation. While WA11_ACTI significantly overpredicts the FR values with an NMB of 526.4%, YU10 largely underpredicts them with an NMB of -63%. BO08_ACTI and COMB show a closer agreement with observed FR values with NMBs of -12.4% and 56.5%, respectively. Although BO08_ACTI gives the smallest NMB of -12.4% at the PKU site, the BO08_ACTI gives much lower FR values in upper layers than YU10 that are inconsistent with observed vertical distributions of ultrafine particle number concentrations (see Figure 2).

441 As shown in Figure 4, at this urban site, the PM_{10} concentrations are
442 underpredicted on July 3 (185), 13 (195), and 19-22 (201-204), which may lead to the
443 underpredictions of the coagulation scavenging rates due to fewer particles in the
444 atmosphere. Since the newly-formed nuclei undergoes two competitive processes:
445 coagulation scavenging and condensational growth (Kerminen et al., 2001), the
446 inaccurate prediction of atmospheric PM concentrations may be a source of uncertainties
447 in predicting NPF events. Although the PM_{10} concentrations are well predicted on July 7
448 (189) and 16 (198), the NPF events are still overpredicted during the two days by COMB
449 and on July 16 by all parameterizations except for YU(10) (see Figure 7), which implies
450 that other factors such as precipitation may play an important role in dictating the
451 nucleation event. Heavy precipitation could lead to strong wet scavenging and deposition,
452 removing newly-formed particles before they can grow into larger particles. The observed
453 precipitation reported by Wang (2012) during the two days is not captured by the
454 simulation, which may explain to a large extent the overpredicted NPF on both days.
455 Despite inaccurate predictions in the NPF events on July 7 and 16, PM_{10} mass
456 concentrations are well predicted because the contributions of newly-formed particles to
457 PM_{10} mass concentrations are negligible.

458 In addition to precipitation, other meteorological variables such as T2, P, Q2,
459 WS10, and WD10 may affect the occurrence of the NPF event and predicted ultrafine
460 number concentrations. To understand the favorable meteorological and chemical
461 conditions for NPF events in Beijing, the time series of T2, relative humidity at 2-m
462 (RH2), and overlay of N-PSD, H_2SO_4 , PM_{10} , and CS predicted from COMB at the PKU
463 site are shown in Figure 5. These plots suggest that NPF events occur under the certain
464 conditions with low RH, high H_2SO_4 concentrations, low PM_{10} concentrations, and low
465 CS. Figure 6 compares the diurnal variations of RH2, H_2SO_4 , PM_{10} , and CS at the PKU

site from the simulation with COMB during NPF and Non NPF events in July, 2008. The results show that RH, PM₁₀, and CS are 23%, 54%, and 54% lower during NPF days than those on Non NPF days; however, H₂SO₄ concentration is about 139% higher on NPF days than on Non NPF days. These results from COMB are consistent with the observational discrepancies of RH, H₂SO₄, PM₁₀, and CS during NPF and Non NPF events analyzed by Wu et al. (2007) and Wang (2012) in Beijing.

Figure 7 compares the simulated time series of N-PSD with those observed by Wang (2012) during the CAREBeijing2008 campaign and the simulation results without nucleation (NO_NUCL) and with various nucleation parameterizations. The criterion for discerning NPF events is the burst of the nucleation mode particles with usually maximum number concentrations $> 10^4 \text{ cm}^{-3}$ for 3 nm – 10 nm particles (Birmili and Wiedensohler, 2000; Wu et al., 2007). The observations show the occurrence of nucleation events on July 7, 12, 17, and 30, corresponding to Julian days 189, 194, 199, and 212, respectively. Comparing to the N-PSD without accounting for nucleation, the N-PSDs predicted with nucleation parameterizations show varying degrees of improvement for nucleation and Aitken modes by capturing the reported NPF events on July 7, 12, 17, and 30. BO08_ACT1 underpredicts both nucleation and Aitken mode particle concentrations during all days. WA11_ACT1 tends to overpredict nucleation mode and underpredict Aitken mode particle concentrations on both nucleation and non-nucleation days. YU10 underpredicts both nucleation and Aitken mode particle concentrations on most days but overpredicts them during July 20-22 (Julian days of 202-204) when observations showed no occurrence of nucleation. COMB underpredicts nucleation and Aitken mode particle concentrations on observed nucleation days but overpredicts nucleation mode particle concentrations on non NPF days such as on July 3 (Julian day

185), 13 (195), 16 (198), and 19-22 (201-204). Overall, the simulations with BO08_ACT1 and COMB show better agreement than WA11_ACT1 and YU10. As shown in Figure 8, comparing to the simulation without a nucleation parameterization, the simulation COMB gives significantly higher PM number concentrations at the surface layer and the column CCN concentrations at a supersaturation of 1.0%, indicating the potentially important role of the nucleation process on cloud formation.

4. Application and comprehensive evaluation of WRF/Chem with COMB for 2001

The aforementioned analyses show that among the four parameterizations, COMB gives the best overall performance in capturing surface NPF events and the vertical distribution of ultrafine particles. In this section, WRF/Chem with COMB and LE07 is applied to four representative months (i.e., January, April, July, and October) in 2001. The comprehensive model evaluation of the improved WRF/Chem is performed to examine its capability in reproducing available meteorological and chemical observations. Sensitivity simulations are then conducted to estimate the impacts of anthropogenic aerosols on aerosol direct and indirect effects.

4.1 Meteorological and chemical evaluation

Table 8 summarizes the performance statistics for meteorological variables using NCDC data for the 2001 simulations. T2 is simulated reasonably well during the four months (Jan., Apr., Jul., and Oct.) in 2001, with MBs of -1.4 to 1.4 °C, NMBs of -10% to 40%, and R of 0.55~0.89. Q2 is well simulated with MBs of 0.3-0.5 g kg⁻¹, NMBs of 2.4-14.6%, and R of 0.55~0.92. WD10 is also well predicted with MBs of -8.6 to -2.7°. However, WS10 is overpredicted for all four months with NMBs of 48.7-82.3%, which is similar to previous studies (Cheng and Steenburgh, 2005; Zhang et al., 2010c, 2012b), for the reason mentioned previously. Daily total precipitation is moderately underpredicted

with NMBs of -46% to -37% and R values of 0.04~0.17, which may be due to the large uncertainties in cloud formation and aerosol-cloud-precipitation interactions.

4.2 Chemical predictions

Table 9 summarizes performance statistics for surface concentrations in the four months. Figure 9 shows the spatial distributions of simulated monthly-mean PM_{10} overlaid with observations during the four months in 2001. The northwestern China, northern China, and southern Mongolian are the three regions with the largest dust aerosol sources in East Asia, leading to high PM_{10} concentrations. The API-derived PM_{10} concentrations are overall well reproduced with NMBs of -25% to -11.3%, and R values of 0.53 - 0.81. The model reproduces well the seasonality of PM_{10} concentrations over mainland China, with the highest concentrations in winter, followed by spring, fall, and summer. However, over Taiwan in all months and Japan in January and July 2001, PM_{10} concentrations are moderately-to-significantly underpredicted with NMBs from -56% to -35%. Possible reasons include the underestimation of the dust transport from desert, the emissions of local natural and anthropogenic sources, and the underestimations of $PM_{2.5}$. Over Hong Kong, PM_{10} concentrations are well predicted with NMBs of -5.5% to 15.0%. The surface concentrations of CO , NO_2 , and SO_2 are underpredicted in nearly all months (except for SO_2 in July over Taiwan) (with NMBs of -84% to -27% for CO , -89% to -26% for NO_2 , and -84% to -17% for SO_2) over Taiwan, Hong Kong, and Japan, likely due to the underestimation of anthropogenic emissions or limited capabilities of the model in simulating some meteorological variables which significantly affect air quality (e.g., the planetary boundary layer height (PBLH) and WS10/WD10). For instance, high wind speeds favor the air pollutant dispersion. Thus, the overprediction of WS10 may lead to the underprediction of concentrations of some chemical species. O_3 surface mixing ratios

are overpredicted in nearly all months over Japan and Taiwan with NMBs of 9.1-164.5% (except for April over Japan with an NMB of -15.5%), likely due to inaccurate predictions of its precursors (such as NO_x and VOCs) and underpredicted NO_x titration effects as discussed previously. Figure 10 compares simulated and observed monthly average PM composition at an urban site (Tsinghua) in Beijing, indicating that the concentrations of $\text{PM}_{2.5}$, SO_4^{2-} , NO_3^- , and NH_4^+ are largely underpredicted in January, due in part to the underestimation of the emissions of precursors such as SO_2 and NO_x and in part to the inaccurate meteorological predictions (e.g., the significant overpredictions of WS10 in January). The overpredicted T2 indicates a potential overprediction of PBLH, which would favor pollutant dispersion, leading to PM underpredictions. The underpredictions of SO_4^{2-} may also be due to the underestimation of CS.

Figures 11 and 12 compare simulated and observed column O_3 , NO_2 , and CO concentrations, as well as aerosol and cloud variables, respectively. Table 10a summarizes the corresponding performance statistics for those variables. All column variables in all months are simulated reasonably well with NMBs of -18.9% to 24.6% and R values of 0.31 to 0.79 except for column O_3 in January, and column CO in January and October that show a poor agreement in terms of spatial distribution. As shown in Figure 12 and Table 10b, AOD is reasonably well predicted with NMBs of -39.8% to 33.9% and R values of 0.53-0.72. CF, COT, and CWP are significantly underpredicted, with NMBs of -72.9% to -38.4% and R values of 0.16-0.74 in the four months, especially in the northern part of East Asia, which may be due to the poor performance on simulating ice clouds by the current model (Zhang et al., 2013) and the underestimations of the subgrid convective clouds and the related aerosol effects (Yu et al., 2013).

4.3 Direct and Indirect feedbacks of anthropogenic aerosols

Figure 13 shows the absolute differences of PM number concentrations, AOD, surface net shortwave radiation (SNSR), T2, PBLH, CDNC, COT, and total precipitation (TP, which includes convective and non-convective precipitation) between the baseline simulations and those without anthropogenic aerosols. Anthropogenic aerosols contribute significantly to the mass and number concentrations of PM in East Asia in all months in 2001. The total aerosol number concentrations over all size bins over East Asia are about 12 times higher compared with those simulated over the U.S. by Zhang et al. (2010c) in January and July, 2001. Due to the increase of anthropogenic aerosols, AOD increases by about 0.09 to 0.20 (or by 73-228%) during the four months in 2001. The largest increases occur in eastern China in all months and in both eastern and northeastern China in July and October. Anthropogenic aerosols can reduce surface net shortwave radiation (SNSR) by up to 42.5 W m^{-2} in January, 52.8 W m^{-2} in April, 48.2 W m^{-2} in July, and 51.1 W m^{-2} in October (or by up to 24.1-36.6%) due to direct scattering (cooling effect) that dominates over the absorption (warming effect). The largest decreases in SNSR occur over areas where AOD is high. As direct responses to the reductions in SNSR and latent heat, T2 reduces by up to $0.74 \text{ }^{\circ}\text{C}$ in January, $0.54 \text{ }^{\circ}\text{C}$ in April, $0.34 \text{ }^{\circ}\text{C}$ in July, and $0.83 \text{ }^{\circ}\text{C}$ in October. 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 86.5-125.9 m (or by up to 11.2-22.9%), indicating an increased atmospheric stability that can further worsen the air pollution in East Asia. Through acting as CCN, anthropogenic aerosols can increase column CDNC by $4.3 \times 10^5 - 9.6 \times 10^5 \text{ cm}^{-2}$ (or by 40.2% - 76.4%), COT by 0.52 - 1.36 (or by 14.3% - 25.3%) during the four months in 2001. Large increases in CDNC occur in central China and spread eastward in all months particularly in January. Anthropogenic aerosols can suppress the formation of warm clouds, reducing the domainwide mean total precipitation by 0.01 to 0.11 mm day^{-1} (or by 0.7% to 2.7%). As

indicated by Zhang et al. (2010c), the small aerosol effect on precipitation in January is due to the omission of ice nuclei and its impact on ice cloud formation.

Zhang et al. (2010c) studied the feedbacks of total aerosols (including both of anthropogenic and natural aerosols) in the continental U.S. and reported that total aerosols can reduce SNSR by up to 11.4 and 39.5 W m⁻², T2 by up to 0.16 and 0.37 °C PBLH by up to 22.4 and 92.4 m, and COT by up to 2.1 and 10.3 in January and July, respectively. Zhang et al. (2013) performed a similar study in Europe and reported that total aerosols can reduce SNSR by up to 16.5 W m⁻², T2 by up to 0.17 °C PBLH by up to 34.4 m, and COT by up to 1.1 in July, 2001. The direct, semi-direct, and indirect effects of anthropogenic aerosols in East Asia are either comparable or even more significant than those of the total aerosol in continental U.S. and Europe due to high PM concentrations resulted from its severe pollution, particularly in January.

5. Conclusions

East Asia has been selected for testing various nucleation parameterizations, identifying the most appropriate nucleation parameterization, and examining the aerosol direct and indirect effects and their seasonal variations in 2001. To explicitly track the formation and growth of new particles, the default 8-bin structure used in MOSAIC in WRF/Chem has been extended to 12-bin. Three nucleation parameterizations (i.e., BO08_ACTI, WA11_ACTI, and YU10) are incorporated into WRF/Chem. WRF/Chem simulations for July 2008 are performed with each of the three parameterizations and a combination of them (i.e., COMB) which uses WA11_ACTI for urban areas and BO08_ACTI for non-urban areas in the PBL, and YU10 for all locations above the PBL. The simulation results for nucleation related parameters, chemical concentrations, and related meteorological variables are evaluated using all available observations. Among all WRF/Chem simulations, the simulation with COMB gives the best overall performance

in reproducing the reported NPF events in Beijing and the vertical distribution of ultrafine particles in the atmosphere. However, WRF/Chem with COMB still large overpredicts the concentrations of key precursor (i.e., H_2SO_4) of NPF, but underpredicts the condensation sink, growth rate, and PM concentrations. Those inaccuracies can be explained to a large extent by the uncertainties in simulated atmospheric oxidation capability and thus the gas-phase oxidation rate of SO_2 , the overprediction in wind speed, the underpredictions in the background PM concentrations, and inaccuracies in the nucleation and early growth of nuclei and nano particles (e.g., missing additional condensable organic vapors), and very limited observational data and associated uncertainties in the measurement data. In particular, the model does not treat the co-condensation of organic compounds that may be responsible for the large underprediction of the growth rates of the ultrafine particles. Additional uncertainties exist in the model inputs such as emissions used and model representations such as nucleation parameterizations and the early growth algorithm used in the simulations, all of which will affect the accuracies of the model predictions. A coarse resolution of 36-km is used in all simulations in this work, which may explain in part the biases in model predictions. For example, two different representative sites (such as urban and rural) might be located in the same grid due to the coarse resolution. However, the simulated FR is the same in each grid at both sites. Using a finer grid resolution with nesting methods will likely improve the model predictions of relevant variables during the NPF events. In addition, only two different types (urban and non-urban) of land covers are considered in this study. A total of 24 types of land covers are classified in model. Different land covers indicate different ambient conditions under which the nucleation mechanisms may be different due to different emissions of gaseous precursors (such as SO_2), thus affecting new particle formation. The results also show that low RH, high H_2SO_4 concentration,

low background PM concentration, and low CS are four favorable conditions for NPF events in Beijing, which are consistent with their impacts on NPF events based on observational data analyses.

WRF/Chem with COMB and 12-bin is applied to simulate air quality, meteorology, and aerosol direct and indirect effects for January, April, July, and October in 2001. The comprehensive model evaluation shows that the model can well predict the near surface temperature, water vapor, and pressure, but higher biases exist in the predictions of the daily precipitation and wind speeds due to inaccurate model representations of related processes such as surface roughness, cloud formation, aerosol-cloud-precipitation interactions. Comparing with the satellite data, the model predicts AOD and column mass concentrations of CO, NO₂, O₃, and SO₂ reasonably well (except for the column O₃ in January, and column CO in January and October 2001 in terms of spatial distributions). The dust storms from northwestern China, northern China and southern Mongolian are well captured with the online dust emission module of Zender et al. (2003). However, the model underpredicts the surface concentrations of PM₁₀, NO_x, CO, SO₂, and VOCs at some sites due in part to possible underestimation of emissions and in part to inaccurate meteorological predictions such as overpredictions of wind speeds. Due to high concentrations of anthropogenic aerosols over East Asia, the effects of anthropogenic aerosols are significant with overall impacts greater than those of total aerosols in the U.S. and Europe. The simulation results indicate that the anthropogenic aerosols in East Asia can reduce surface net shortwave radiation by up to 42.5 – 52.8 W m⁻² (or by 24.1 - 36.6%), T₂ by up to 0.34 - 0.83 °C and PBLH by up to 76.8 – 125.9 m (or by 11.2 - 22.9%). They can also increase AOD by 64.0 - 228.3%, CDNC by 40.2 - 76.4%, COT by 14.3 - 25.3%, and LWP by 5.4 - 44.8% during the four months in 2001. These results indicate the importance of aerosol feedbacks in East Asia, and thus a need

to continually improve model's representations of relevant atmospheric aerosol and cloud processes such as aerosol-cloud interactions and cloud and precipitation formation.

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Figure 1. Spatial distributions of the surface formation rate of bin-1 from WRF/Chem with 12-bin using various nucleation schemes, (a) BO08_ACTI, (b) WA11_ACTI, (c) YU10, and (d) COMB that uses WA11_ACTI for urban areas in PBL, BO08_ACTI for non-urban areas in PBL, and YU10 for above PBL.

Figure 2. Zonal mean values of formation rate (J) using various nucleation schemes, (a) BO08_ACTI, (b) WA11_ACTI, (c) YU10, and (d) COMB, (e) the ionization rate simulated by WRF/Chem with 12-bin and COMB scheme, and (f) ultrafine particle number concentrations (cm^{-3}) based on the measurement data (Yu et al., 2008) (sigma is the height index used in Global Chemical Transport Model (GEOS-Chem), 48 vertical sigma levels extending from the surface to approximately 0.01 hPa).

Figure 3. Monthly-averaged particle number size distributions on non-polluted ($\text{PM}_{10} \leq 150 \mu\text{g m}^{-3}$) and polluted days ($\text{PM}_{10} > 150 \mu\text{g m}^{-3}$) at Beijing urban site, the campus of Peking University (PKU) (39.99°N , 116.31°E) in Beijing based on predictions with WRF/Chem with 12-bin without (a) and with various nucleation parameterizations (b)-(e) and lognormal fits of the measured particle number size distributions in the summer of 2006 of Yue et al. (2009) (f).

Figure 4. Comparison of the time series for surface H_2SO_4 , CS, FR, PM_{10} , SO_2 , O_3 and NO_2 from WRF/Chem (12-bin) with available observations (observations are indicated as black dots for FR and as solid black line with or without dots for other parameters) at an urban site, the campus of Peking University (PKU) (39.99°N , 116.31°E) in Beijing in July 2008.

Figure 5. Times series of T_2 , RH_2 , Particle number size distribution, number concentrations of H_2SO_4 , mass concentrations of PM_{10} , and CS simulated by WRF/Chem with 12-bin, COMB, and LE07 at an urban site, the campus of Peking University (PKU) ($39.99^\circ N$, $116.31^\circ E$) during July 2008.

Figure 6. Average diurnal variations during the NPF days and no NPF event (Non_NPF Event) days simulated by WRF/Chem with 12-bin, COMB, and LE07 at an urban site, the campus of Peking University (PKU) ($39.99^\circ N$, $116.31^\circ E$) in Beijing in July 2008 for RH_2 , number concentrations of H_2SO_4 , mass concentrations of PM_{10} , and CS .

Figure 7. Comparison of the time series of particle number size distributions (3-900 nm) between observational data (Wang, 2010) and modeling results using 12-bin WRF/Chem with and without various nucleation schemes (NO_NUCL, BO08_ACTI, WA11_ACTI, YU10, and COMB) at an urban site, the campus of Peking University (PKU) (39.99°N, 116.31°E) in Beijing in July 2008. The color bar represents $dN/d\log D_p$ (cm⁻³).

Figure 8. Absolute differences in the simulated monthly-averaged total PM number concentrations at the surface layer and the column CCN concentrations at a supersaturation of 1.0% between the simulation COMB and the simulation without a nucleation parameterization.

Figure 9. Overlay of observed and simulated monthly-mean PM₁₀ during the four months (Jan. Apr. Jul. and October) in 2001. The simulation is based on WRF/Chem with 12-bin, COMB, and LE07. The observations are derived from China MEP API, Hong Kong EPD, Taiwan AQMN, and Japan NIES.

January

April

July

October

Figure 10. Observed and simulated monthly mean concentrations of $\text{PM}_{2.5}$ and its composition (NH_4^+ , SO_4^{2-} , NO_3^- , Na^+ , and Cl^-) at an urban site (Tsinghua) in Beijing during four months (January, April, July and October) in 2001. The simulation is based on WRF/Chem with 12-bin, COMB, and LE07. The observations are taken from Tsinghua University. The values over each bar indicate the mass concentrations of each species.

Figure 11. Observed and simulated spatial distributions of column mass concentrations of CO, O₃ and NO₂ during four months (January, April, July and October) in 2008. The simulation is based on WRF/Chem with 12-bin, COMB, and LE07.

Figure 12. Observed and simulated spatial distributions of AOD, CCN5, CF, COT, and CWP in four months (January, April, July and October) of 2001. The simulation is based on WRF/Chem with 12-bin, COMB, and LE07. The observations are taken from MODIS Terra data for all other variables.

Figure 13. Spatial distributions and absolute differences for (a) total PM number concentrations, (b) AOD, (c) SNSR, (d) T2, (e) PBLH, (f) CDNC, (g) COT, (h) total precipitation due to anthropogenic aerosols during the four months (January, April, July and October) in 2001. The absolute differences are taken between simulations with and without anthropogenic aerosols using WRF/Chem with 12-bin, COMB, and LE07.

Table 1. WRF/Chem model configurations and model set up used in this work.

Attribute	Model Configuration
Simulation period	July, 2008; January, April, July, and October, 2001
Domain	East Asia
Horizontal resolution	36 km (164×97)
Vertical resolution	23 layers from 1000 mb - 50 mb
Meteorological initial condition (IC) and boundary condition (BC)	The National Centers for Environmental Predictions Final Analysis (NCEP-FNL) reanalysis data; re-initialization every day
Shortwave radiation	Goddard shortwave radiation scheme (Chou and Max, 1994)
Longwave radiation	The rapid radiative transfer model (RRTM) (Mlawer et al., 1997)
Land surface	Community National Centers for Environmental Prediction (NCEP), Oregon State University, Air Force, and Hydrologic Research Lab-NWS Land Surface Model (NOAH) (Tewari et al., 2004)
Surface layer	Monin-Obukhov (Monin and Obukhov, 1954; Janjic, 2002)
Planetary boundary layer (PBL)	Yonsei University (YSU) PBL scheme (Hong et al., 2006)
Cumulus	Grell-Devenyi ensemble (Grell and Devenyi, 2002)
Microphysics	Purdue Lin (Lin et al., 1983)
Aerosol activation	Abdul-Razzak and Ghan (Abdul-Razzak and Ghan, 2002)
Gas-phase chemistry	Carbon-Bond mechanism version Z (CBMZ) (Zaveri and Peters, 1999)
Photolysis	Fast-J (Wild et al., 2000)
Aerosol module	Model for simulating aerosol interactions and chemistry (MOSAIC) (Zaveri et al., 2008)
Aqueous-phase chemistry	Carnegie Mellon University (CMU) mechanism (Fahey and Pandis, 2001)
Chemical IC	Liu et al. (2010) based on the Community Multiscale Air Quality (CMAQ) modeling system
Chemical BC	Liu et al. (2010) based on the Goddard Earth Observing System Atmospheric Chemistry Transport Model (GEOS-Chem)
Anthropogenic emissions	Adjusted version for Wang et al. (2010) following the approach of X. Zhang (2013)
Biogenic emissions	Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 (Guenther et al., 2006)
Dust emissions	Zender et al. (2003) implemented by K. Wang et al. (2012)
Sea-salt emissions	Gong et al. (2002)

Table 2. Nucleation parameterizations and particle early growth treatment to be examined in this work

(a) Nucleation parameterizations

Category	Reference	Theoretical Basis	Dependent variable	Equation:
Binary	Wexler et al., 1994 (WE94) (Default in WRF/Chem)	Classical binary (H ₂ SO ₄ -H ₂ O) nucleation theory	^a <i>T</i> , ^b <i>RH</i> , and ^c <i>C_{cir,H2SO4}</i>	$C_{cir,H2SO4} = 0.16 \exp(0.1T - 3.5RH - 27.7)$
Power Law	Wang et al., 2011 (WA11), used for urban and polluted rural areas	Activation theory	<i>N_{sulf}</i>	$^f J = ^g A \times N_{sulf}$ $A = 1.95 \times 10^{-6} \text{ s}^{-1}$
		Kinetic theory	<i>N_{sulf}</i>	$J = ^h K \times N_{sulf}^2$ $K = 3.44 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$
		Thermodynamic theory	<i>N_{sulf}</i>	$J = ^i T_c \times N_{sulf}^3$ $T_c = 5.96 \times 10^{-20} \text{ cm}^6 \text{ s}^{-1}$
	Boy et al., 2008b (BO08), used for mountain and forest areas	Activation theory	<i>N_{sulf}</i>	$J = A \times N_{sulf}$ $A = 0.28 \times 10^{-6} \text{ s}^{-1}$
		Kinetic theory	<i>N_{sulf}</i>	$J = K \times N_{sulf}^2$ $K = 0.18 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$
Ion-mediated	Yu, 2010 (YU10)	Ion-mediated nucleation (IMN) theory	<i>N_{sulf}</i> , <i>T</i> , <i>RH</i> , ^j <i>Q</i> , and ^k <i>S</i>	Look-up table

(b) Particle early growth algorithm

Category	Reference	Theoretical Basis	Dependent variable	Equation:
Particle Early Growth	Lehtinen et al., 2007 (LE07)	Based on coagulation sink	^l <i>CoagS</i> , ^m <i>GR</i> , <i>dx</i> , <i>d1</i> and <i>r</i>	$m = \frac{\log[CoagS(a_x) / CoagS(d_1)]}{\log[d_x / d_1]}$ $r = \frac{1}{m+1} \left[\left(\frac{d_x}{d_1} \right)^{m+1} - 1 \right]$ $J_x = J_1 \cdot \exp[-r \cdot d_1 \cdot \frac{CoagS(d_1)}{GR}]$

^a*T* – temperature; ^b*RH* – relative humidity; ^c*C_{cir,H2SO4}* – the critical concentration in $\mu\text{g m}^{-3}$; ^d*N_{sulf}* – number concentrations of H₂SO₄, cm^{-3} ; ^e*C_{NH3}* – volume mixing ratio of NH₃; ^f*J* – formation rate; ^g*A* – activation coefficient; ^h*K* – kinetic coefficient; ⁱ*T_c* – thermodynamic coefficient; ^j*Q* – ionization rate, ion-pairs $\text{cm}^{-3} \text{ s}^{-1}$; ^k*S* – surface area of pre-existing particles, $\mu\text{m}^2 \text{ cm}^{-3}$; ^l*CoagS* – coagulation sink; ^m*GR* – growth rate

Table 3. The default 8-bin, modified 8-bin, and new 12-bin structure in WRF/Chem

Bin #	8-bin						Bin #	12-bin		
	Default			Modified				New		
	low ^a	high ^b	dcen ^c	low	high	dcen		low	high	dcen
							01	1.0×10 ⁻³	2.15×10 ⁻³	1.47×10 ⁻³
							02	2.15×10 ⁻³	4.64×10 ⁻³	3.16×10 ⁻³
							03	4.64×10 ⁻³	1.0×10 ⁻²	6.81×10 ⁻³
							04	1.0×10 ⁻²	2.15×10 ⁻²	1.47×10 ⁻²
01	3.91×10 ⁻²	7.81×10 ⁻²	5.52×10 ⁻²	2.15×10 ⁻²	4.64×10 ⁻²	3.16×10 ⁻²	05	2.15×10 ⁻²	4.64×10 ⁻²	3.16×10 ⁻²
02	7.81×10 ⁻²	1.56×10 ⁻¹	1.10×10 ⁻¹	4.64×10 ⁻²	1.0×10 ⁻¹	6.81×10 ⁻²	06	4.64×10 ⁻²	1.0×10 ⁻¹	6.81×10 ⁻²
03	1.56×10 ⁻¹	3.13×10 ⁻¹	2.21×10 ⁻¹	1.0×10 ⁻¹	2.15×10 ⁻¹	1.47×10 ⁻¹	07	1.0×10 ⁻¹	2.15×10 ⁻¹	1.47×10 ⁻¹
04	3.13×10 ⁻¹	6.25×10 ⁻¹	4.42×10 ⁻¹	2.15×10 ⁻¹	4.64×10 ⁻¹	3.16×10 ⁻¹	08	2.15×10 ⁻¹	4.64×10 ⁻¹	3.16×10 ⁻¹
05	6.25×10 ⁻¹	1.25	8.84×10 ⁻¹	4.64×10 ⁻¹	1	6.81×10 ⁻¹	09	4.64×10 ⁻¹	1	6.81×10 ⁻¹
06	1.25	2.5	1.77	1	2.15	1.47	10	1	2.15	1.47
07	2.5	5	3.54	2.15	4.64	3.16	11	2.15	4.64	3.16
08	5	10	7.07	4.64	10	6.81	12	4.64	10	6.81

^alow – Low bound diameter of the bin in μm ; ^bhigh – High bound diameter of the bin in μm ; ^cdcen – Geometric mean diameter of the bin in μm

Table 4. Summary of observational databases used in model evaluation

Variables/Species	Dataset	Data Frequency
<i>Meteorology</i>		
Precip (mm day ⁻¹)	NCDC	Hourly
T2 (°C)	NCDC	Hourly
Q2 (kg kg ⁻¹)	NCDC	Hourly
WS10 (m s ⁻¹)	NCDC	Hourly
WD10 (°)	NCDC	Hourly
<i>Gaseous Species</i>		
H ₂ SO ₄	Wang (2012) (for Beijing)	Daily
SO ₂	SCIAMACHY (column mass for the whole domain)	Monthly
	API (for Beijing)	Daily
	NIES (for Japan)	Monthly
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
O ₃	TOMS and OMI (column mass for the whole domain)	Monthly
	TOMS (column mass for the whole domain)	Monthly
	TOMS (column mass for the whole domain)	July 25-26, 2008
	He et al. (2010)	Daily
	API (for Beijing)	Monthly
	NIES (for Japan)	Daily
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
NO ₂	GOME (column mass for the whole domain)	Monthly
	API (for Beijing)	Daily
	NIES (for Japan)	Monthly
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
NO	NIES (for Japan)	Monthly
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
CO	MOPITT (column mass for the whole domain)	Monthly
	API (for Beijing)	Daily
	NIES (for Japan)	Monthly
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
VOCs	Wang et al. (2010a) (For Beijing)	Monthly
	Cai et al. (2010) (For Shanghai)	Monthly
<i>Aerosol Species</i>		
PM ₁₀	API from MEP (for China mainland)	Daily
	NIES (for Japan)	Monthly
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
PM _{2.5}	Miyun and Tsinghua	Daily
	AQMN (for Taiwan)	Daily
	EPD (for Hong Kong)	Daily
Na ⁺	Miyun and Tsinghua	Daily
NH ₄ ⁺	Miyun and Tsinghua	Daily
Cl ⁻	Miyun and Tsinghua	Daily
NO ₃ ⁻	Miyun and Tsinghua	Daily
SO ₄ ²⁻	Miyun and Tsinghua	Daily
<i>Nucleation-Related Parameters</i>		
Formation rate	Wang (2012) (for Beijing)	Daily
Particle size distribution	Wang (2012) (for Beijing)	Monthly
Condensation sink	Wang (2012) (for Beijing)	Daily
<i>Aerosol and Cloud Column Variable</i>		
COT	MODIS	Monthly
CWP (g m ⁻²)	MODIS	Monthly
PWV (cm)	MODIS	Monthly
AOD	MODIS	Monthly

SCIAMACHY- Scanning Imaging Absorption spectrometer; TOMS-the Total Ozone Mapping Spectrometer; OMI-the Ozone Monitoring Instrument; MOPITT-the Measurements of Pollution in the Troposphere; MODIS -Terra Moderate-resolution Imaging Spectroradiometer; NIES - the National Institute of Environmental Studies in Japan (<http://www.nies.go.jp/igreen/index.html>); AQMN- the Taiwan Air Quality Monitoring Network (<http://taqm.epa.gov.tw/taqm/en/default.aspx>); EPD- the Environmental Protection Department (EPD) of Hong Kong (<http://epic.epd.gov.hk/>); MEP- the Ministry of Environmental Protection of China (MEP) (<http://datacenter.mep.gov.cn/>).

Table 5. Observed New particle formation rates in China.

Location	Lat ^a & Lon ^b	Type of location	New particle formation rate J (cm ⁻³ s ⁻¹)			Time period	References
			Mean	Min	Max		
Beijing	39°59'N, 116°18'E	Urban	19.3	3.3	81.4	Mar., 2004 – Feb., 2005	Wu et al., 2007
		Urban	-	1.1	22.4	Aug. 2004, 2006, Sept. 2005-2006	Yue et al., 2009
Nanjing	32°07'N, 118°57'E	Urban	10.1	2.2	34.5	Mar. – Nov., 2008	Wang et al., 2013
		Urban	33.2	-	-	Nov. 18, 2011 – Mar. 31, 2012	Herrmann et al., 2013
Lanzhou	36°08'N, 103°41'E	Urban	4.3	1.8	7.1	25, Jun. – 19, Jul., 2006	Gao et al., 2011
Yufa	39°31'N, 116°18'E	Rural	-	1.6	33.0	Aug. 4 – 25, 2004; Aug. 10- Sept. 10, 2005 and 2006	Yue et al., 2009
Shangdianzi	40°39'N, 117°07'E	Rural	8.0	0.7	72.7	Mar., 2008 – Aug., 2009	Shen et al., 2011
	40° 39'N, 117° 17'E	Rural	5.0	0.4	24.5	Mar. – Nov., 2008	Wang et al., 2013
Taicang	31°27'N, 121°08'E	Rural	2.2	1.2	2.5	May 5 –Jun. 2, 2005	Gao et al., 2009
Hong kong	22°15'N, 114°10'E	Coastal	2.9	1.0	6.9	Oct. 25 –Nov. 29, 2010	Guo et al., 2012
Xinken	22°37'N, 113°35'E	Coastal	3.4	0.5	5.2	Oct. 3 – Nov. 5, 2004	Liu et al., 2008a, b

Table 6. Performance statistics for meteorological and chemical predictions using updated 12-bin WRF/Chem model with COMB^a for July 2008.

Variable	Dataset	July 2008							
		Mean Obs	Mean Mod	Data #	MB	RMSE	NMB, %	NME, %	R
Meteorology									
T2 ^b , °C	NCDC ^g	24.9	23.7	317429	-1.2	25.6	-4.8	15.506	0.56
P ^c , mb	NCDC	956.6	942.5	188681	-14.1	1331.8	-1.5	2.3	0.94
Q2 ^d , g kg ⁻¹	NCDC	14.5	15.2	188542	0.7	5.6	4.6	12.8	0.87
WS10 ^e , m s ⁻¹	NCDC	2.5	3.7	316728	1.2	8.2	49.1	88.9	0.26
WD10 ^f , degree	NCDC	183.2	179.1	283878	-4.1	13389.1	-2.2	48.2	0.16
24-h rain, mm	NCDC	4.9	3.8	22321	-1.0	350.8	-21.3	1.5	0.01
Gas-phase species									
SO ₂ , ppbv	API	11.0	6.1	26	-4.9	6.3	-44.7	49.1	0.45
TOL ¹ , ppbv (Beijing)	B. Wang et al., 2010a	3.67	0.70		-2.97		-80.9		
TOL, ppbv (Shanghai)	Cai et al., 2010	7.74	2.32		-5.42		-70.0		
XYL ¹ , ppbv (Beijing)	B. Wang et al., 2010a	1.20	0.55		-0.65		-54.2		
XYL, ppbv (Shanghai)	Cai et al., 2010	2.23	1.14		-1.09		-48.9		
Isoprene, ppbv (Beijing)	B. Wang et al., 2010a	0.68	0.83		0.15		22.1		
Isoprene, ppbv (Shanghai)	Cai et al., 2010	0.13	0.28		0.15		115.4		
O ₃ , ppbv	He et al., 2010	63.8	75.6	48	11.9	53.7	18.6	70.5	0.62
NO ₂ , ppbv	API	28.2	17.9	26	-10.3	13.1	-36.4	40.0	-0.02
Aerosol species									
PM ₁₀ , µg cm ⁻³	API ^h	113.5	88.7	28	-24.8	38.5	-21.9	28.4	0.76
PM _{2.5} , µg cm ⁻³	Tsinghua site	77.8	69.6	31	-8.3	22.9	-10.6	24.9	0.91
Na ⁺ , µg cm ⁻³		0.26	0.14	31	-0.12	0.25	-46.9	84.2	0.04
NH ₄ ⁺ , µg cm ⁻³		7.9	11.1	31	3.2	6.8	39.9	68.6	0.85
Cl ⁻ , µg cm ⁻³		1.3	0.3	31	-1.1	1.3	-86.6	89.6	-0.03
NO ₃ ⁻ , µg cm ⁻³		15.4	22.0	31	6.6	15.6	42.9	80.6	0.76
SO ₄ ²⁻ , µg cm ⁻³		27.7	13.2	31	-14.6	18.7	-52.5	55.4	0.84
PM _{2.5} , µg cm ⁻³	Miyun site	65.6	63.8	31	-1.8	29.6	-2.7	34.8	0.81
Na ⁺ , µg cm ⁻³		0.26	0.14	31	-1.2	0.27	-46.9	84.2	0.04
NH ₄ ⁺ , µg cm ⁻³		7.9	11.1	31	3.2	6.4	39.9	68.6	0.85
Cl ⁻ , µg cm ⁻³		15.4	22.0	31	6.6	15.5	42.9	80.6	0.76
NO ₃ ⁻ , µg cm ⁻³		27.7	13.2	31	-14.5	15.2	-52.5	55.4	0.84
SO ₄ ²⁻ , µg cm ⁻³		65.6	63.8	31	-1.8	29.6	-2.7	34.8	0.81

^aCOMB – Combination of BO08_ACT1, WA11_ACT1 and YU10; ^bT2 – Temperature at 2 meters; ^cP – Atmospheric pressure; ^dQ2 – Water vapor mixing ratios at 2 meters; ^eWS10 – Wind speeds at 10 meters; ^fWD10 – Wind directions at 10 meters; ^gNCDC – National Climatic Data Center;

^hAPI – Air Pollution Index; ¹TOL – Benzene, Toluene, Ethylbenzene, i-Propylbenzene, n-Propylbenzene; ¹XYL – m,p,o-Xylene, Styrene, 1,3,5-Trimethylbenzene, 1,2,4-Trimethylbenzene, 1,2,3-Trimethylbenzene, p-Ethyltoluene

Table 7. Evaluation of nucleation related variables predicted with WRF/Chem 12-bin with various nucleation parameterizations.

Variable	Parameterization	July 2008							
		Mean Obs	Mean Mod	Data #	MB	RMSE	NMB, %	NME, %	R
H_2SO_4 , cm^{-3}	BO08_ACTI ^a	4.3×10^6	3.9×10^7	228	3.5×10^7	6.7×10^7	808.2	858.7	0.25
	WA11_ACTI ^b	4.3×10^6	3.6×10^7	228	3.2×10^7	5.9×10^7	739.3	789.9	0.25
	YU10 ^c	4.3×10^6	3.8×10^7	228	3.4×10^7	6.5×10^7	787.6	839.4	0.25
	COMB ^d	4.3×10^6	3.9×10^7	228	3.5×10^7	6.7×10^7	816.0	866.3	0.25
CS^e , s^{-1}	BO08_ACTI	4.1×10^{-2}	1.2×10^{-2}	420	-2.9×10^{-2}	3.2×10^{-2}	-69.9	70.8	0.51
	WA11_ACTI	4.1×10^{-2}	1.3×10^{-2}	420	-2.8×10^{-2}	3.2×10^{-2}	-69.1	70.4	0.49
	YU10	4.1×10^{-2}	1.2×10^{-2}	420	-2.9×10^{-2}	3.2×10^{-2}	-70.4	71.2	0.52
	COMB	4.1×10^{-2}	1.2×10^{-2}	420	-2.9×10^{-2}	3.2×10^{-2}	-71.1	71.7	0.55
GR^f , nm h^{-1} (3-7 nm)	BO08_ACTI	12.5	2.9		-9.6		-77.1		-
	WA11_ACTI	12.5	3.4		-9.1		-73.1		-
	YU10	12.5	4.1		-8.4		-66.8		-
	COMB	12.5	4.9		-7.6		-60.6		-
FR^g , $\text{cm}^{-3} \text{s}^{-1}$ (3-6 nm)	BO08_ACTI	7.3	6.4		-0.9		-12.4		-
	WA11_ACTI	7.3	45.9		38.6		526.4		-
	YU10	7.3	2.7		-4.6		-63.0		-
	COMB	7.3	11.5		4.2		56.5		-

^aBO08_ACTI – Activation theory of Boy et al. 2008; ^bWA11_ACTI – Activation theory of Wang et al. 2011; ^cYU10 – Ion-mediated nucleation of Yu, 2010; ^dCOMB – Combination of BO08_ACTI, WA11_ACTI, and YU10; ^eCS – Condensation sink; ^fGR – Growth rate for particles with diameters of 3-7 nm; ^gFR – Formation rate for particles with diameters of 3-6 nm. The observations are taken from Wang (2012).

Table 8. Evaluation of meteorological variables predicted with WRF/Chem (12-bin) with COMB in 2001.

Variable	Month	Mean Obs.	Mean Mod.	Data #	MB	RMSE	NMB, %	NME, %	R
T2 ^a , °C	Jan.	1.1	1.5	299396	0.4	26.9	40.0	36.4	0.89
	Apr.	13.6	12.3	297625	-1.3	40.2	-9.7	35.3	0.69
	Jul.	25.2	23.8	302284	-1.4	30.0	-5.6	16.3	0.55
	Oct.	15.9	15.5	303799	-0.4	28.0	-2.5	25.3	0.74
Q2 ^c , g kg ⁻¹	Jan.	3.4	3.9	175430	0.5	1.6	14.6	25.5	0.92
	Apr.	6.2	6.4	171193	0.3	2.8	4.2	20.0	0.90
	Jul.	14.8	15.1	177205	0.3	5.9	2.4	12.8	0.87
	Oct.	8.3	8.8	174734	0.5	3.2	5.9	16.1	0.91
WS10 ^d , m s ⁻¹	Jan.	3.2	5.8	295637	2.6	18.6	82.3	105.9	0.43
	Apr.	3.2	5.1	293157	1.8	13.3	55.8	88.8	0.31
	Jul.	2.6	3.9	306316	1.3	8.2	48.7	86.0	0.29
	Oct.	2.7	4.8	301608	2.1	12.1	79.0	102.6	0.44
WD10 ^e , degree	Jan.	210.1	207.4	248091	-2.7	17978.0	-1.3	45.1	0.25
	Apr.	190.8	183.9	264684	-6.9	16761.1	-3.6	48.9	0.21
	Jul.	183.2	175.1	267535	-8.1	14291.2	-4.4	49.2	0.14
	Oct.	181.7	173.1	254266	-8.6	20521.8	-4.7	57.6	0.23
24-h rain, mm	Jan.	1.9	1.2	21215	-0.7	45.4	-37.4	118.9	0.17
	Apr.	1.9	1.1	22142	-0.8	61.1	-40.4	128.5	0.16
	Jul.	5.0	3.0	21882	-2.0	290.3	-40.7	125.5	0.10
	Oct.	2.7	1.5	22865	-1.2	139.1	-44.5	135.6	0.04

^aT2 – Temperature at 2 meters; ^bP – Atmospheric pressure; ^cQ2 – Water vapor mixing ratios at 2 meters; ^dWS10 – Wind speeds at 10 meters;

^eWD10 – Wind directions at 10 meters

Table 9. Evaluation of surface chemical concentrations predicted with WRF/Chem (12-bin) in 2001.

Dataset	Variable	Month	Mean Obs	Mean Mod	Data #	MB	RMSE	NMB, %	NME, %	R
Mainland China	PM ₁₀ , μg cm ⁻³	Jan.	161.5	143.3	40	-18.2	126.5	-11.3	52.2	0.53
		Apr.	148.05	115.8	42	-32.2	72.5	-21.8	37.0	0.81
		Jul.	91.5	68.6	42	-22.9	45.3	-25.0	40.2	0.69
		Oct.	113.2	92.1	42	-21.0	51.1	-18.6	34.0	0.58
Taiwan	PM ₁₀ , μg cm ⁻³	Jan.	65.7	28.6	134	-17.9	28.4	-56.4	67.2	0.22
		Apr.	67.4	32.6	144	-18.8	25.5	-51.7	57.0	0.50
		Jul.	38.0	19.6	123	-10.8	16.0	-48.3	52.9	-0.12
		Oct.	64.7	32.7	145	-17.8	25.8	-49.5	57.0	0.07
	PM _{2.5} , μg cm ⁻³	Jan.	36.0	18.1	134	-17.9	28.4	-49.8	64.5	-0.08
		Apr.	39.6	20.8	144	-18.8	25.5	-47.4	53.6	0.18
		Jul.	27.4	16.6	123	-10.8	16.0	-39.3	46.1	0.17
		Oct.	39.0	21.2	145	-17.8	25.8	-45.7	51.2	0.27
	SO ₂ , ppbv	Jan.	4.6	3.5	1896	-1.0	5.0	-22.6	69.0	0.22
		Apr.	4.6	3.6	1912	-0.9	4.5	-19.8	65.5	0.24
		Jul.	3.0	3.5	1917	0.5	3.8	17.8	87.9	0.17
		Oct.	3.8	3.2	1953	-0.7	3.3	-16.9	60.4	0.30
	NO ₂ , ppbv	Jan.	24.1	7.3	1933	-16.8	19.6	-69.8	70.4	0.41
		Apr.	24.1	6.8	1881	-17.3	19.5	-71.8	72.1	0.49
		Jul.	14.4	6.2	1927	-8.2	10.5	-57.1	59.6	0.44
		Oct.	19.7	6.1	1944	-13.6	15.5	-68.9	69.2	0.43
	NO, ppbv	Jan.	11.1	0.7	1905	-10.4	15.8	-93.9	94.1	0.29
		Apr.	8.0	0.6	1857	-7.5	11.7	-93.0	93.2	0.38
		Jul.	6.3	0.6	1906	-5.6	8.6	-89.7	90.2	0.32
		Oct.	4.7	0.5	1890	-4.3	6.9	-90.3	90.7	0.18
	CO, ppmv	Jan.	0.7	0.3	1985	-0.4	0.5	-53.2	54.2	0.12
		Apr.	0.7	0.3	1914	-0.5	0.6	-61.5	61.7	0.21
		Jul.	0.5	0.2	1933	-0.3	0.4	-63.2	63.8	0.16
		Oct.	0.6	0.3	1980	-0.3	0.4	-53.0	53.3	0.36
	O ₃ , ppbv	Jan.	23.0	31.2	1953	8.2	12.1	35.8	43.6	0.36
		Apr.	27.6	33.9	1901	6.3	12.3	22.9	37.1	0.39
		Jul.	20.4	27.2	1952	6.8	11.5	33.4	47.5	0.21
		Oct.	34.3	38.1	1977	3.8	12.6	11.0	28.9	0.28
Hong Kong	PM ₁₀ , μg cm ⁻³	Jan.	57.6	66.3	31	8.6	27.4	15.0	38.4	0.48
		Apr.	51.9	49.5	30	-2.5	23.0	-4.7	34.8	0.67
		Jul.	31.9	30.1	31	-1.8	11.0	-5.5	22.4	0.61
		Oct.	62.1	64.1	31	2.1	22.1	3.3	25.1	0.22
	SO ₂ , μg m ⁻³	Jan.	14.2	6.6	31	-7.6	10.4	-53.5	53.4	0.56
		Apr.	14.4	4.8	30	-9.6	12.2	-66.9	67.0	0.12
		Jul.	19.8	3.3	31	-16.6	20.9	-83.5	83.5	0.60
		Oct.	11.0	6.1	31	-4.9	8.4	-44.2	46.6	0.34
	NO ₂ , μg m ⁻³	Jan.	33.4	10.2	31	-23.2	24.7	-69.5	69.5	0.40
		Apr.	61.7	9.2	30	-52.5	53.6	-85.1	85.1	0.56
		Jul.	48.0	3.9	31	-44.1	47.0	-91.8	91.8	0.36
		Oct.	63.2	7.0	31	-56.3	57.5	-89.0	89.0	0.13
	NO, μg m ⁻³	Jan.	73.4	0.3	31	-73.1	83.1	-99.6	99.6	0.00
		Apr.	66.1	0.4	30	-65.7	70.9	-99.4	99.4	0.00
		Jul.	77.6	0.2	31	-77.4	84.4	-99.8	99.8	-0.18
		Oct.	40.8	0.2	31	-40.6	43.7	-99.5	99.5	-0.07
	CO, μg m ⁻³	Jan.	1045.6	640.7	31	-404.9	446.0	-38.7	38.7	0.62
		Apr.	843.0	377.3	30	-465.6	503.0	-55.2	55.2	0.36
		Jul.	1277.9	207.0	31	-1070.9	1158.0	-83.8	83.8	-0.07
		Oct.	932.1	499.8	31	-432.3	481.3	-46.4	46.4	0.24
	O ₃ , μg m ⁻³	Jan.	27.6	67.2	31	39.6	41.4	143.4	143.4	0.26
		Apr.	34.4	58.5	30	24.1	28.5	69.9	69.9	0.50
		Jul.	18.5	49.0	31	30.5	32.1	164.5	164.5	0.05
		Oct.	53.9	81.3	31	27.4	31.6	50.9	50.9	0.32
Japan	PM ₁₀ , μg cm ⁻³	Jan.	22.2	12.4	1470	-9.8	13.3	-44.1	52.3	0.29
		Apr.	33.9	31.0	1537	-2.9	7.7	-8.6	18.2	0.30
		Jul.	35.1	22.6	1537	-12.5	15.6	-35.5	38.2	0.29
		Oct.	27.6	26.4	1541	-1.2	7.3	-4.3	20.6	0.44
	SO ₂ , ppbv	Jan.	3.9	2.1	1478	-1.8	3.3	-46.4	69.1	0.18
		Apr.	6.0	2.5	1490	-3.5	4.6	-57.6	65.2	0.29
		Jul.	5.3	2.8	1453	-2.6	4.6	-48.3	70.2	0.23
		Oct.	4.2	2.6	1444	-1.6	3.3	-38.8	64.2	0.36
	NO ₂ , ppbv	Jan.	17.8	6.7	1467	-11.1	12.7	-62.1	62.7	0.65
		Apr.	16.9	7.7	1465	-9.1	11.4	-54.1	57.4	0.54
		Jul.	12.6	9.3	1465	-3.3	7.7	-26.4	48.8	0.50
		Oct.	16.9	8.5	1468	-8.4	10.3	-49.6	52.2	0.69
	NO, ppbv	Jan.	15.4	0.03	1456	-15.4	19.4	-99.8	99.8	0.19
		Apr.	6.6	0.04	1448	-6.6	8.2	-99.4	99.4	0.03
		Jul.	5.5	0.05	1446	-5.5	6.7	-99.2	99.2	-0.03
		Oct.	9.9	0.05	1452	-9.9	12.2	-99.5	99.5	0.21
	CO, ppmv	Jan.	0.6	0.2	132	-0.3	0.4	-56.2	56.8	0.34
		Apr.	0.4	0.3	131	-0.2	0.2	-42.2	42.9	0.30
		Jul.	0.3	0.2	131	-0.1	0.1	-27.0	31.1	0.43
		Oct.	0.5	0.3	130	-0.2	0.2	-41.9	42.9	0.49
	O ₃ , ppbv	Jan.	26.7	29.1	202	2.4	5.9	9.1	17.7	0.38
		Apr.	43.2	36.5	200	-6.7	9.9	-15.5	19.4	0.17
		Jul.	28.7	34.6	202	5.9	9.6	20.5	27.6	0.41
		Oct.	26.0	30.4	202	4.4	7.7	17.1	23.4	0.38

Table 10. Evaluation of column variables predicted with WRF/Chem (12-bin) with COMB in 2001.

(a) Gases

Variable	Month	Mean Obs	Mean Mod	Data #	MB	RMSE	NMB, %	NME, %	R
CO, $\times 10^{17}$ molecules cm^{-2}	Jan.	18.1	22.5	15854	4.2	7.8	24.6	27.5	0.59
	Apr.	22.3	20.8	15908	-1.5	5.0	-6.7	18.5	0.70
	Jul.	No data							
	Oct.	17.6	19.6	15908	2.0	5.2	11.1	18.1	0.74
NO ₂ , $\times 10^{15}$ molecules cm^{-2}	Jan.	2.2	2.8	9845	0.6	2.8	25.1	64.1	0.74
	Apr.	1.6	1.9	14677	0.3	1.3	22.3	51.3	0.78
	Jul.	1.3	1.6	12671	0.3	1.4	19.8	54.0	0.69
	Oct.	2.0	2.2	12551	0.1	2.2	2.7	53.6	0.79
O ₃ , DU	Jan.	26.4	30.8	14850	4.4	8.4	16.6	25.0	0.02
	Apr.	37.4	36.8	15329	-0.5	6.1	-0.8	12.3	0.31
	Jul.	43.2	35.0	15908	-8.1	9.9	-18.9	20.3	0.40
	Oct.	30.7	31.5	15908	5.9	7.2	2.3	20.2	0.52

(b) Aerosol and cloud properties

Variable	Month	Mean Obs.	Mean Mod.	Data #	MB	RMSE	NMB, %	NME, %	R
AOD	Jan.	0.27	0.25	10735	-0.02	0.19	-9.1	48.4	0.63
	Apr.	0.45	0.27	13152	-0.18	0.25	-39.8	44.0	0.72
	Jul.	0.29	0.22	14907	-0.07	0.17	-23.5	40.9	0.66
	Oct.	0.22	0.30	14544	0.08	0.21	33.9	60.3	0.53
CCN5, $\times 10^8$ # cm^{-2}	Jan.	7.8	7.5	5871	-0.3	8.8	-6.6	62.3	0.72
	Apr.	9.0	9.8	5836	0.8	5.9	7.9	41.5	0.76
	Jul.	4.8	7.1	5718	2.3	4.7	45.2	71.8	0.79
	Oct.	6.3	9.8	6143	3.5	7.6	54.5	97.4	0.63
CF (%)	Jan.	0.67	0.35	15908	-0.32	0.36	-47.8	48.0	0.71
	Apr.	0.59	0.29	15908	-0.30	0.32	-50.7	50.8	0.73
	Jul.	0.66	0.38	15908	-0.28	0.32	-42.6	42.9	0.64
	Oct.	0.56	0.34	15908	-0.21	0.26	-38.4	40.4	0.62
COT	Jan.	16.8	5.2	15908	-11.6	13.7	-69.0	71.0	0.58
	Apr.	15.4	4.2	15908	-11.3	12.1	-72.9	73.1	0.72
	Jul.	17.5	7.2	15908	-10.3	13.0	-58.9	64.6	0.18
	Oct.	17.8	5.4	15908	-12.4	13.7	-61.1	65.5	0.48
CWP, g m^{-2}	Jan.	171.0	49.8	15908	-121.2	151.5	-70.9	71.3	0.34
	Apr.	164.0	50.1	15908	-113.9	127.7	-69.4	70.1	0.54
	Jul.	228.1	93.4	15908	-134.7	171.3	-59.1	66.7	0.23
	Oct.	205.3	66.7	15908	-138.6	156.4	-67.5	69.3	0.48