Development of a Regional Chemical Transport Model with Size-Resolved Aerosol Microphysics and Its Application on Aerosol Number Concentration Simulation over China

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Abstract

An advanced particle microphysics (APM) model has been incorporated into a regional chemical transport model, NAQPMS, to simulate the seasonal variation of particle number concentrations over China in 2007 for the first time. The NAQPMS+APM can reproduce the particles number concentrations at remote, suburban sites and urban sites reasonably. Most of modeled values were within a factor of two of observations. The simulation indicated that particles number concentration was significantly higher in southeastern China than that in northwestern China. Monthly mean number concentration can be over 20000 cm\textsuperscript{-3} in most polluted regions in southeastern China while the value is generally below 7000 cm\textsuperscript{-3} in northwestern parts of China. Higher number concentration occurred in January while lower value occurred in April and July. In heavily polluted regions, like Sichuan Basin and central-eastern China, primary particles dominated particles number, while secondary particles formed via the nucleation process account for most of particles number over relative clean areas. The area over which secondary particles dominated showed distinct seasonal variation and its spatial pattern was coupled with primary particles distribution which was strongly influenced by the meteorological conditions, e.g., East Asia Monsoon.

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1. Introduction

Over the past few years, there has been increased research into aerosol physics and chemistry due to their important climate and environment effects (Charlson et al. 1992; Albrecht 1989; Twomey 1974, 1977; Delfino et al. 2005). The climate and environment effects of aerosols are controlled by several physical and chemical properties but among them particle size distribution is a key parameter. More and more studies point to the importance of particle size distribution in evaluating the effect of aerosols on climate and environment (Du et al. 2006; Baumer et al. 2007; Han et al. 2012). As a result, there have been many studies considering size resolved microphysics in regional (Chang et al. 2009; Fountoukis et al. 2012) and global models (Adams et al. 2003; Spracklen et al. 2006; Yu and Luo 2009). However, large uncertainties still exist in quantifying aerosol number size distribution over multiple scales in time and space (Pierce et al. 2007; Zhang et al. 2010; Lee et al. 2011).

Recently aerosol is becoming a more and more serious environmental problem in China due to the enhanced emissions of aerosol particles and their precursors into the atmosphere (Laakso et al. 2006; Chan et al. 2008). More importantly, aerosol properties showed a high spatial variability due to the inhomogeneous distribution of population and industry. While there were many studies focused on single aerosol components or aerosol mass, modeling on aerosol number concentration in China was scarce. Though several global models have been developed and evaluated against some observations (Spracklen et al. 2010; Yu and Luo 2009), their ability in simulating aerosols over China is not fully assessed. On the other hand, due to the complexity of aerosol microphysics, simplifications and parameterizations are adopted in many widely used models (Yan et al. 2004; Luo and Yu 2011). Application of these models is not enough to understand haze formation and evaluate aerosol effects over China.

In this study, we incorporated an advanced particle microphysics module (APM) into NAQPMS (Nested Air Quality Prediction Modeling System) and conducted simulation and preliminary evaluation in terms of aerosol number concentration. APM employs state-of-the-science, ion-mediated nucleation theory and high size resolution and it has exhibited good performance in simulating aerosol microphysics and the associated properties (Yu and Luo 2009; Yu et al. 2012). It can be expected that incorporation of APM into NAQPMS (Wang et al. 2006; Li et al. 2012) can be a significant step toward fully characterizing the physical and chemical features of aerosols over China. This paper may provide a new perspective of understanding aerosol behavior in China and supplement the existing research.

2. Model description and application

The NAQPMS is a three-dimensional system with full physical and chemical processes to describe regional and urban scale atmospheric pollution. Since 1995, NAQPMS has been widely applied in a number of scientific studies and air quality forecast practice (Wang et al. 2006). In the original NAQPMS, aerosol size distribution is represented by a static two-mode coarse/fine scheme. Primary species can be modeled as fine and/or coarse particles, while all secondary (chemically-formed) species are modeled as fine particles only. An aerosol thermodynamic equilibrium partition model (ISORROPiA1.7) was applied to calculate the composition, phase state and aerosol liquid water content of an ammonia-sulfate-nitrate-chloride-sodium-water inorganic system (Nenes et al. 1998). A bulk yield scheme is used to deal with the formation of secondary organic aerosols (SOA) (Li et al. 2011).

The APM model is a sectional microphysics aerosol model developed for a wide range of applications (Yu and Luo 2009; Luo and Yu 2011). The components involved in the model are black carbon (BC), organic carbon (OC), dust, sea salt, sulfate, nitrate and ammonium. Mixing state is assumed to be semi-extemal, i.e. nucleated secondary particle is internally mixed while primary particles, like BC, OC, dust and sea salt are assumed to be composed of a seeding core and coating species. The basic microphysical processes treated include nucleation, condensation/evaporation, coagulation, thermodynamic equilibrium with local humidity. In APM, 40 sectional bins are used to represent sulfate.
particles covering dry diameters ranging from 0.0012 μm to 12 μm. Nucleation scheme is the ion-mediated nucleation (IMN) which is physically-based and constrained by recently available laboratory data (Yu 2010). More details about APM can be found in Yu and Luo (2009) and reference therein.

In this study, we have coupled the APM with a recently developed version of NAQPMS in order to study the spatial and seasonal variations of aerosol number concentration over China. In the model, size-resolved microphysics for sulfate and sea salt aerosols is explicitly described. The formation of new particles by nucleation is calculated with IMN mechanism. Particle growth due to uptake of nitrate, ammonium, SOAs and condensation of sulfuric acid is considered. The coagulation of sea salt, secondary sulfate particles and coagulation scavenging of secondary particles by other types of particles (sea salt, BC, POC, and dust) is calculated. The microphysics of carbonaceous aerosols is not explicitly resolved. We assume a two lognormal size distribution for them and their number concentration is derived from mass concentration. In the NAQPMS+APM model, the tracers added to the original NAQPMS include 40 bins of sulfate, 20 bins of sea salt, eight category of carbonaceous aerosols, four bins of dust, sulfuric acid gas and four secondary species coated on sea salt, dust, BC, POC, respectively.

The meteorological fields input to NAQPMS+APM were produced by WRF. Emission data input to NAQPMS+APM is from REAS version 2 (Kurokawa et al. 2013). The NAQPMS+APM modeling domain in this study covers East Asia with a resolution of 81 km in a 79 × 75 horizontal grid system. Number of vertical layers is 20 with higher resolution in the surface layer. The model top is 20 km. In this study, we assume that the 4 months of January, April, July, and October can represent the four typical seasons of winter, spring, summer, and autumn, respectively. Seasonal variation for each variable is characterized by analyzing their variations in four different months. The annual mean value of variable is obtained by averaging values over the 4 months.

3. Results

3.1 Model evaluation

Long term measurements on aerosol number concentration in China are scarce, so we collected data from published papers to evaluate the performance of NAQPMS+APM preliminarily. We only chose observations with more than ten days so that they have better representation though the sampling periods between observation and model were not strictly same. The detailed information for these samples can be found in supplementary material.

Figure 1 shows the comparison of simulated number concentration of condensation nuclei with diameter larger than 10 nm (CN10nm) with observations. NAQPMS+APM not only captured the spatial distribution of CN10nm in different environmental conditions including remote mountain sites, polluted mountain sites, urban sites but also reproduced the seasonal variation of CN10nm at two urban sites (sample 7–10 at Beijing and 11–14 at Jinan). Generally, the number concentration was highest at urban sites, lower in suburban sites and lowest in mountain sites. At PKU (sample 6) and Taicang (sample 15), number concentration can be larger than 30000 cm⁻³ with modeled values being 28048 cm⁻³ and 41411 cm⁻³, respectively. At suburban sites (sample 3, 4, 5, 21), most of observed number concentrations are in the range from 10000 cm⁻³ to 20000 cm⁻³ with modeled ones being a little higher but still within a factor of two of observations. Among the mountain sites, Mount Tai (sample 20) showed the highest number concentration in Spring. Due to the higher altitude, Mount Huang (samples 18, 19) suffered less effect from the surrounding pollution and showed the lower number concentration than that at Mount Tai. At Mount Waliguan (sample 17), the number concentration showed a relative lower annual mean value with the observed being 2030 cm⁻³ and modeled being 1753 cm⁻³, respectively. The scatter plot (shown in supplementary material) showed that most of the modeled values were within a factor of two of observations. Over China adjacent seas, the simulation showed a decreased pattern from north to south (Yellow Sea > East China Sea > South China Sea) in spring. This was consistent with observations by Lin et al. (2007). The spatial pattern and magnitude of CN10nm in our modeling results were similar with the results of a global aerosol model driven by assimilated meteorology reported by Trivitayuranuk et al. (2008) and other global modeling studies (Yu and Luo 2009; Spracklen et al. 2010). Considering the different emission inventory and transport, our modeling results were comparable to these studies over China. The NAQPMS+APM model can also simulate the mass concentration of sulfate and black carbon at CAWNET sites (Zhang et al. 2012) reasonably (shown in supplementary material). These comparisons demonstrated that the NAQPMS+APM model has the ability to reasonably simulate spatial distribution and seasonal variation of aerosol number concentration.

3.2 Seasonal cycle of CN10nm over China

Figure 2 shows the simulated spatial distribution of CN10nm in January, April, July, and October in 2007. It is clear from Fig. 2 that number of CN10nm varied among different parts of China to southeastern China, i.e., CN10nm had significant higher values in southeastern China than in northwestern China. Monthly mean CN10nm can be over 20000 cm⁻³ in certain area in southeastern China while the value is generally below 7000 cm⁻³ in northwestern parts. Though observations showed the high aerosol concentration centers over the deserts in Northwest China, the modeled spatial pattern of CN10nm did not show such obvious behavior. There were two reasons for this phenomenon. Firstly, the larger size of dust particles resulted in less contribution to particles' number. Secondly, due to the coarse size bin structure, number concentration of dust particles may be underestimated, especially the ultrafine particles, under the condition of mass conservation. An improved, higher resolution bin scheme should be adopted in the future work. In central-eastern China, due to higher emission of primary particles and precursor gases, larger number of carbonaceous and secondary particles with smaller size led to high centers of particles’ number. In addition, it can be seen that there were some differences in spatial pattern and absolute value of CN10nm among different seasons. Higher CN10nm value occurred in January while lowest value occurred in April and July, which was mainly influenced by meteorological conditions with a certain effect from emissions.

Understanding the aerosol number budget and quantifying the contributions from different sources in different parts of China is very important to understand aerosol distribution and formulate effective control measures to reduce particles level. Both nucleation and primary emission can contribute the aerosol number. In this paper, secondary particles refers to the particles from nucleation processes while primary particles refers to the particles directly emitted into the atmosphere from natural and anthropogenic sources even though secondary species coated on them may contribute a large fraction to their mass. Figure 3 presents CN10nm number fraction of secondary particles. It can be clearly seen that there was a boundary line between regions dominated by secondary particles and regions dominated by primary particles. Secondary particles dominate particle number concentration over the northwestern part of China while primary particles dominate particle number concentration over the southeastern part of China.
Comparing Fig. 3 with Fig. 4 which shows the primary particles, it can be found that regions dominated by secondary particles were also regions with less primary particles. Because of the high abundance of primary particles, the resulting high condensation sink for condensable gases in central-eastern China can limit the nucleation and growth of newly formed particle. In addition, secondary particles can be more efficiently scavenged by primary particles through coagulation over regions with larger number of primary particles. As a result, over more polluted regions, like central-eastern China, particles number was dominated by primary particles with secondary particle number fraction below 50%.

Fig. 3 shows the seasonal variation of areas where secondary particles were dominant. The location of the boundary line dividing regions dominated by secondary particles and primary particles shifted in north-south direction in different seasons. This reveals the combined effects of several factors. Due to the lower temperature and strong dependency of nucleation on temperature (Yu 2010), nucleation rate was relative higher in cold seasons, which was a prerequisite for larger contribution of secondary particles. Although the area with higher nucleation in January was largest, the area dominated by secondary particles was smaller than that in April because of the larger abundance of pre-existing particles in January which can scavenge secondary particles by coagulation. In July, lower nucleation rate (not shown) due to higher temperature limited the contribution of secondary particles to particles’ number though warm-moist climate in summer can...
lead to higher mass of secondary species due to favorable conditions for secondary aerosol formation. In other words, warm-moist climate in summer is more favorable in producing aerosol mass than in producing new particles. Another important factor was the transport of primary particles by East Asia Monsoon. Regions with high pollution levels move southward under the influence of winter monsoon while move northward under the influence of summer monsoon (Zhu et al. 2004; Zhao et al. 2010; Zhang et al. 2010). Such effect was also revealed in Fig. 4. The position of regions with high primary particles abundance was located to north in summer and autumn months while it was located to south in winter and spring months, which lead to the seasonal variation of spatial distribution of coagulation sink intensity to secondary particles. As a result, regions dominated by secondary particles move southward in winter months while move northward in summer months with the arrival of summer monsoon.

4. Summary and discussion

In this study, we have coupled a size-resolved aerosol microphysics model to a regional chemical transport model and have presented the performance of the model and the modeled features of aerosol number concentration. The model has been evaluated against mass concentration of aerosol components at CAWNET sites and number concentration in different environments. The simulated mass and number concentration shows good agreement with the observations over China, which gives us confidence in our simulation in different regions with different particle pollution level. Nonetheless, further evaluation and model constrains are needed. The model’s ability in simulating aerosol size distribution and capturing new particles formation event should be assessed. The uncertainties associated with nucleation scheme, emission inventory and size distribution of emitted primary particles should be further analyzed and quantified.

The particle number concentration over China had significant seasonal variation and large spatial difference. In general, largest CN10nm number occurs in January due to the disadvantage meteorological condition for pollutant diffusion and stronger emission. Particles number concentration on the southeastern part of China can be over 20000 cm\(^{-3}\) in certain area while the value was generally below 7000 cm\(^{-3}\) on the northwestern part. Our simulations also showed that the contribution of secondary particles to particle number concentration had significant spatial and seasonal variations. The area dominated by secondary particles was largest in April. Primary particles were always dominant in central-eastern China in four months where primary emission was largest among areas over China. It is important to separate these two sources because their contributions respond in different ways to gas and particle emission and environmental changes. We can draw a conclusion that secondary particles and primary particles are strongly coupled through microphysics. There exist complex interactions among precursor gases, secondary particles and primary particles. It is important to consider microphysical processes in regional chemical transport models for understanding the physics and chemistry of aerosols and quantifying aerosol climate and environment effects.

Supplement

Supplement 1 presents the tables and figures mentioned in this manuscript.

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