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Influence of aerosol hygroscopic growth parameterization on aerosol optical depth and direct radiative forcing over East Asia

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ABSTRACT

The influence of aerosol hygroscopic growth parameterization on aerosol optical depth (AOD) and aerosol direct radiative forcing (ADRF) over East Asia is investigated by using an online coupled regional climate-chemistry/aerosol model (RIEMS-Chemaero) focusing on the period of summer 2006. Three aerosol hygroscopic growth schemes are tested in this study. Model performances are evaluated with ground observations and satellite retrievals. Comparison with observations of aerosol concentration demonstrates that the model is able to reproduce the spatial and temporal variations of aerosol components over East Asia. Model comparison with AOD measurements shows that AOD is best predicted by the aerosol hygroscopic growth scheme developed based on observations in China (Case B), and the aerosol hygroscopic growth affects AOD simulation significantly. In this study, the domain and seasonal mean AOD, ADRF at the top of the atmosphere, and ADRF at the surface over East Asia are estimated to be 0.31, -9 W/m^2, and -29 W/m^2 by Case B, respectively. Compared with Case B, the estimations from Case A (scheme from CCM3 radiation package) differ by +71%, +100%, and +17%, respectively, while those from Case C (κ parameterization) differ by −16%, +11%, and −17%. The large differences in AOD and ADRF among cases suggest the necessity to develop appropriate hygroscopic growth parameterization with geographical characteristics in climate model for estimating regional aerosol optical properties and radiative effects.

1. Introduction

Atmospheric aerosols affect atmospheric radiation transfer and climate by scattering or absorbing solar and infrared radiation (direct effect), by acting as cloud condensation nuclei and modifying cloud properties (indirect effect), and by altering atmospheric stability and cloud formation (semi-direct effect) (Ramanathan et al., 2001; Forster et al., 2007). Aerosol radiative effects depend strongly on aerosol optical properties (such as extinction coefficient, single scattering albedo, and asymmetry factor) (Chylek and Coakley, 1974), which are mainly determined by aerosol chemical components, sizes, and shapes on condition of humidity. The hygroscopic growth of aerosols with increasing relative humidity (RH) affects aerosol direct radiative effects by changing aerosol optical properties due to increasing water uptake of hydrophilic compositions such as sulfate, nitrate, and some organic matters. The aerosol hygroscopicity varies by space and time because of different aerosol sources, types, and chemical components (Day and Malm, 2001; Kim et al., 2006; Liu et al., 2008, 2013; Yan et al., 2009).

Because of the increasing anthropogenic emissions resulting from rapid economy growth, atmospheric aerosols have become major pollutants in East Asia, especially China. The environmental impacts and radiative effects of aerosols thus become an important issue for this region. To precisely evaluate the aerosol radiative effects (such as direct radiative forcing) in this region, light scattering coefficient of aerosol and its dependency on relative humidity should be investigated. In recent years, a number of observational studies on aerosol hygroscopic growth were conducted in China (e.g. Achtert et al., 2009; Eichler et al.,

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2008; Liu et al., 2008, 2009, 2013; Pan et al., 2009; Xu et al., 2002; Yan et al., 2009; Ye et al., 2011) and in East Asia (e.g. Jung and Kim, 2011; Kim et al., 2006, 2011; Mochida et al., 2010). Numerical model is another effective method in quantitatively studying aerosol hygroscopicity and its impacts on aerosol radiative effects. However, to our knowledge, only a few numerical studies were conducted to investigate aerosol hygroscopic growth and its impacts within East Asia. For instance, Cheng et al. (2008) studied the relative humidity dependence of aerosol optical properties and direct radiative forcing in south China by using a box model based on observations. Liu et al. (2011) combined observations and the regional model WRF to investigate hygroscopic properties of aerosols at high relative humidity conditions in north China.

In this study, an online coupled regional climate-chemistry/aerosol model (RIEMS-Chemaero) is applied to investigate the influence of aerosol hygroscopic growth parameterization on simulations of aerosol optical depth (AOD) and aerosol direct radiative forcing (ADRF) in East Asia. Three parameterizations of aerosol hygroscopic growth are tested and model results are validated with various observations and compared with each other. This work aims to reveal the characteristics of aerosol hygroscopicity in China and the importance of hygroscopic growth parameterization in prediction of aerosol direct radiative forcing at regional scale.

2. Model description

RIEMS-Chemaero is an online-coupled regional climate-chemistry/aerosol model developed on the framework of a regional climate model RIEMS, which applies the dynamic structure of the fifth-generation Pennsylvania State University-NCAR Mesoscale Model (MM5; Grell et al., 1995), with a nonhydrostatic, sigma vertical and Lambert horizontal coordinates. RIEMS has been evaluated extensively in previous studies, and it participated in the Regional climate Model Intercomparison Project (RMIP) for Asia, demonstrating a good ability in predicting East Asian climate, for more details refer to Fu et al. (2005).

In recent years, various aerosol-related processes including emission, transport, diffusion, deposition, and chemistry have been implemented into RIEMS, creating an online RIEMS-Chemaero model (Han, 2010; Han et al., 2012). Multi-phase chemistry is represented by CB-IV gas mechanism (Gery et al., 1989), ISORROPIA thermodynamics model (Nenes et al., 1998), heterogeneous reactions (Li and Han, 2010), and aqueous reactions (Chang et al., 1987). Major aerosol types, namely sulfate, nitrate, ammonium, black carbon (BC), organic carbon (OC), anthropogenic particulate matters other than BC and OC (AnthPM), dust, and sea salt are considered in this model. Sulfate is oxidized by reactions of SO_2 with OH, H_2O_2, and O_3 in both gas phase chemistry and aqueous phase chemistry. Nitrate and ammonium aerosols are formed via the thermodynamic equilibrium process among sulfate–nitrate–ammonia–water system. BC, primary OC, and AnthPM are emitted directly into the atmosphere, while formation of secondary OC is parameterized by a bulk yield method (Lack et al., 2004). A lognormal size distribution is assumed for sulfate, nitrate, ammonium, BC, and OC with median radius of 0.07 μm, 0.07 μm, 0.07 μm, 0.01 μm, and 0.02 μm, and geometric standard deviations of 2.0, 2.0, 2.0, 2.0, and 2.2, respectively (D'Almeida et al., 1991). Dust deflation and sea salt generation are calculated using the same schemes as those in Han et al. (2004), but 5 size bins (0.1–1.0, 1.0–2.0, 2.0–4.0, 4.0–8.0, 8.0–20.0 μm) are used in this study.

Dry deposition velocities of gases are parameterized using the scheme of Walmsley and Wesely (1996), and subgrid cloud mixing and wet deposition are treated by a scavenging submodel as that in Chang et al. (1987). Dry deposition velocities of aerosols are calculated as the inverse of total resistances plus the gravitational settling term for different particle components and sizes. Below-cloud scavenging of aerosols is parameterized based on an expression scavenging rate, which is a function of precipitation rate and collision efficiency of particle by hydrometeor (Han et al., 2004).

A modified radiation package based on the radiation package of the NCAR Community Climate Model, version 3 (CCM3; Kiehl et al., 1996) is used to calculate aerosol perturbation to radiation transfer. RIEMS-Chemaero has ever been used to investigate the direct radiative and climatic effects of anthropogenic and natural aerosols (Han, 2010; Han et al., 2012, 2013) and the long-term trends of aerosol concentrations and direct radiative forcing over East Asia (Li et al., 2013).

In this study, an external mixture between aerosols is assumed, which is thought to be more appropriate for regions close to aerosol sources (Giorgi et al., 2002) and has been observed in eastern China (e.g. Cheng et al., 2009; Meier et al., 2009; Rose et al., 2010). Three cases with different aerosol hygroscopic growth parameterizations are conducted and their results are compared with each other to investigate the impact of hygroscopic growth treatment on simulation of aerosol optical properties. The first case applies the parameterization in the CCM3 package, in which two schemes from Kiehl et al. (2000) and Im et al. (2001) are used for sulfate and other aerosol types, respectively. The above two schemes are derived based on laboratory measurements and field observations in the United States. The hygroscopic growth factor of sulfate optical properties is expressed as a function of relative humidity (RH) in an exponential way:

$$f(RH) = \exp[c_1 + c_2/(RH + c_3) + c_4/(RH + c_5)], \tag{1}$$

where $f(RH)$ denotes the hygroscopic growth factor of aerosol scattering coefficient, c_1 to c_5 are five wavelength-dependent fitting coefficients taken directly from the CCM3 radiation package. The values of these coefficients are 11.24, −0.304, −1.088, −177.6, and 15.37, respectively, at a wavelength of 550 nm. The scheme of Kiehl et al. (2000) is specifically for sulfate, whereas the scheme of Im et al. (2001) applies to other aerosol types (such as carbonaceous aerosols, etc.), which is in a form of power law:

$$f(RH) = (1 - RH)^{-g}, \tag{2}$$

where g is an empirical fitting value, setting to 0.38. In this case, aerosols are assumed to be mixed externally. The simulation with Eqs. (1) and (2) refers to Case A.

The second case is based on observational studies conducted in China during summer. Several field observations concerning the hygroscopic growth of aerosol scattering coefficient were conducted in China (Xu et al., 2002; Yan et al., 2009; Liu et al., 2008, 2009; Pan et al., 2009). Among those