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## Investigation of direct radiative effects of aerosols in dust storm season over East Asia with an online coupled regional climate-chemistry-aerosol model

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#### ABSTRACT

A new online coupled regional climate-chemistry-aerosol model (RIEMS-Chemaero) has been developed and applied to investigate direct radiative effects of dust aerosol and mixed aerosols over East Asia in March 2010, when an extremely intense dust storm on 19-22 March swept across almost the entire east China including the Pearl River Delta of south China, where little dust storm was observed before. The model results are evaluated against ground observation of PM<sub>10</sub> concentration, aerosol optical depth (AOD) from both AERONET measurement and satellite retrieval (MODIS). The comparison demonstrates a good ability of RIEMS-Chemaero in reproducing major features of aerosol spatial distribution and temporal variation, as well as dust evolution during the dust storm period. However, the model tends to generally underpredict AOD at AERONET sites, with larger biases at urban sites than that at rural sites. Dust aerosols exerted a significant impact on radiation energy budget during the dust storm period, with the 4-day mean values of shortwave and longwave radiative forcings at the surface up to  $-90 \text{ W m}^{-2}$ and +40 W m<sup>-2</sup>, respectively, over the Gobi desert. The monthly mean net dust radiative forcings at the surface ranged from -9 to -24 W m<sup>-2</sup> over the dust source regions, and from -6 to -21 W m<sup>-2</sup> over wide downwind areas including the middle and lower reaches of the Yellow River and the Yangtze River and the Yellow Sea. The net dust radiative forcing at TOA varied from near zero to  $+6.0 \text{ W m}^{-2}$  in large areas of the continent. The monthly mean values of the net direct radiative forcings due to dust, non-dust aerosols and all aerosols (dust + sea salt + anthropogenic aerosols) averaged over the whole domain are estimated to be  $-3.9 \text{ W m}^{-2}$ ,  $-5.6 \text{ W m}^{-2}$  and  $-9.3 \text{ W m}^{-2}$ , respectively, at the surface, and to be  $+0.9 \text{ W m}^{-2}$ ,  $-3.0 \text{ W m}^{-2}$  and  $-2.0 \text{ W m}^{-2}$ , respectively, at TOA, indicating a light dust warming effect and an overall aerosol cooling effect in the springtime over East Asia. In east China, the net radiative forcings due to dust, non-dust aerosols and all aerosols at the surface are enhanced to  $-8.4 \text{ W m}^{-2}$ . -10.2 W m<sup>-2</sup> and -18.0 W m<sup>-2</sup>, respectively, due to both the frequent dust influence and the intensive anthropogenic emissions in this region. The dust forcing accounts for about 42% of the total aerosol forcing at the surface in the domain, implying a potentially important role of mineral dust in radiation budget and regional climate. The semi-direct effect of dust aerosol tends to reduce cloud cover throughout the domain and it is partly responsible for the direct radiative forcings because of the feedbacks among aerosol, radiation, cloud and dynamics.

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#### 1. Introduction

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Atmospheric aerosols affect radiation and climate by scattering or absorbing solar and infrared radiation (direct effect), by acting as cloud condensation nuclei and modifying cloud property and lifetime (indirect effect), and by absorbing radiation and thus heating the atmosphere, altering atmospheric stability and cloud formation (semi-direct effect). Aerosol climatic effect is so far one of the key issues in climate research, but the mechanism through which aerosol impacts radiation budget and climate remains largely unknown due to the complexity of aerosol properties, behaviors and interaction between aerosol and climate dynamics. The fourth assessment report from the Intergovernmental Panel on Climate Change (IPCC, 2007) indicates that the direct radiative forcing (RF) due to increase in anthropogenic aerosols is estimated to be -0.5 W m<sup>-2</sup>, with uncertainty ranging from -0.1 to -0.9 W m<sup>-2</sup>, with medium-low level of scientific understanding. The direct RF of individual aerosol species is even less certain than the total direct aerosol RF, such as mineral dust

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aerosol, its global annual mean RF is estimated as  $-0.1 \text{ W m}^{-2}$ , with an uncertainty in a range of  $-0.3 \sim +0.1 \text{ W m}^{-2}$ , which means the dust RF estimated from current methods can be either positive or negative. Therefore, more efforts are required to provide further insight into aerosol optical property and radiative effect.

Different from radiative forcing of green house gases, which is homogeneous across the globe, due to relatively short lifetime. aerosol radiative forcing is considered to be particularly important at regional scale. In the past 30 years, East Asia, especially China, has been the most economically active region in the world. At the same time, emissions of anthropogenic aerosols and their precursors increase dramatically in this region (Streets et al., 2003; Zhang et al., 2009b). In addition, East Asia is also one of the major dust source regions in the world, dust events often occur in springtime and exert significant influence on air quality, human health, as well as climate system (Zhang et al., 2003; Shao and Dong, 2006). Li et al. (2011) revealed that in dust storm season, dust aerosol may account for 40–60% of total PM<sub>10</sub> mass over wide areas of east China and the west Pacific rim. The coexistence and abundance of both anthropogenic and natural aerosols in East Asia make aerosol effect more complex and more difficult to be well understood.

Numerous model simulations were conducted to investigate the source, transport, transformation, deposition and air quality impact of Asian dust aerosol in the past decades (Song and Carmichael, 2001; Uno et al., 2001; Shao et al., 2003; Gong et al., 2003; Liu et al., 2003; Han et al., 2004; Tang et al., 2004; Darmenova et al., 2009) by using CTM or meteorological model coupled with their developed dust models, but only a few modeling studies focused on radiative effects of dust or anthropogenic aerosols over East Asia with radiative transfer model (RTM) or climate model (Conant et al., 2003; Takemura et al., 2003; Wang et al., 2004; Park et al., 2005; Zhang et al., 2009a). The above works have promoted our understanding on the characteristic, behavior and budget of dust aerosol in this region, however, many uncertainties remain, and our knowledge on aerosol radiative and climatic effect is still limited for East Asia.

The above studies generally used offline CTM (or RTM) driven by meteorological model or reanalysis data, which can not explore aerosol-radiation—climate interactions, but such feedback is crucial for understanding climate change and climate prediction.

In recent years, increasing attentions and efforts have been paid to online coupled models because the feedbacks among climate, chemistry, aerosol, radiation and cloud represent more realistically the atmospheric processes and improve the predictions of air quality, weather and climate. (Zhang, 2008; Grell and Baklanov, 2011). Since the first fully-coupled online model GATORM was established by Jacobson (1997), online models have been actively developed with various degrees of coupling and complexity at both global, regional and urban scales. To date, there have been several online models at meso-to-regional scales developed in the past decade in Europe, like COSMA-ART (Vogel et al., 2009), ENVIRO-HIRLAM (Baklanov et al., 2008), Meso-NH (Tulet et al., 2005) and RegCM3 (Giorgi et al., 2002), and in US, like GATORM (Jacobson, 1997), and WRF/Chem (Grell et al., 2005), which is developed based on MM5/Chem and is now used worldwide to simulate and predict meteorology and air quality. There are also some online coupled models on global scale, like GATORG (Jacobson, 2001), MIRAGE2 (Ghan and Easter, 2006), CAM3 (Collins et al., 2006) and Caltech GCM (Liao and Seinfeld, 2005). However, there is so far, no online coupled regional climate model in Asia.

This paper describes an online coupled regional climatechemistry-aerosol model (namely RIEMS-Chemaero) newly developed in RCE-TEA, IAP, CAS, which includes most types of anthropogenic and natural aerosols, relevant gas species, as well as a series of physical and chemical processes. The paper is organized as follows: we first describe the major structure and components of RIEMS-Chemaero, and then examine model performance by comparing with ground observations and satellite retrievals. Next we describe the model simulation of dust aerosol evolution and perturbation of radiation during a severe dust storm, and discuss the aerosol induced shortwave, longwave and net direct radiative forcings and the potential semi-direct effect of dust aerosol associated with cloud cover change. Finally, we present the relative magnitude of the direct radiative forcings due to dust aerosol, nondust aerosols and total aerosols for various regions. This work represents an important effort in developing regional climate model and in advancing our understanding of aerosol radiative impact for East Asia.

#### 2. Model and parameters

#### 2.1. Model description

RIEMS (Regional Integrated Environmental Model System) was developed based on the dynamic structure of the fifth-generation Pennsylvania State University – NCAR Mesoscale Model (MM5; Grell et al., 1995), applying a nonhydrostatic, sigma vertical and lambert horizontal coordinate. It includes a number of physical parameterizations, such as the Biosphere-Atmosphere Transfer Scheme (BATS; Dickinson et al., 1993) for land surface process, the Medium-Range Forecasts (MRF) scheme for planetary boundary layer process (Hong and Pan, 1996), the Grell cumulus convective parameterization scheme (Grell, 1993), and a modified radiation package based on the radiation package of the NCAR Community Climate Model, version CCM3 (Kiehl et al., 1996). RIEMS has ever been used to simulate present climate over East Asia (Xiong et al., 2006) and to investigate the impact of human-induced land cover change on East Asia monsoon (Fu, 2003). RIEMS is also one of the key models in the Regional Climate Model Intercomparison Project (RMIP) for Asia (Fu et al., 2004).

Recently, gas chemistry and aerosol modules have been incorporated into RIEMS by Han (2010). Gas-phase chemistry is represented by CB-IV mechanism (Gery et al., 1989), which includes 37 gas species and 91 reactions. In this study, ISORROPIA (Nenes et al., 1998) is coupled with RIEMS to account for thermodynamic equilibrium process and to yield nitrate and ammonium. Totally 27 gas species and 16 aerosol components (sulfate, nitrate, ammonium, black carbon (BC), organic carbon (OC) and secondary organic carbon (SOA), 5 bins of soil dust and 5 bins of sea salt) are transported, with advection and diffusion algorithms identical to that for water vapor.

Heterogeneous reactions of gaseous species (SO<sub>2</sub>, NO<sub>2</sub>, HNO<sub>3</sub>, O<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, NO<sub>2</sub>, NO<sub>3</sub>, OH, HO<sub>2</sub>) on dust surface are also parameterized and included in RIEMS by Li and Han (2010) as well. SOA production from primary anthropogenic and biogenic VOCs is calculated using a bulk aerosol yield method according to Lack et al. (2004). Sulfate is formed by the reaction of SO<sub>2</sub> with OH, which is derived from real-time CB-IV calculation, and by conversion of SO<sub>2</sub> to sulfate through aqueous phase chemistry (dissolution of SO<sub>2</sub> in cloud water, followed by oxidation of ions by  $H_2O_2$  and  $O_3$ ), which is similar to that in RADM (the Regional Acid Deposition Model) (Chang et al., 1987).

A lognormal size distribution is assumed for sulfate, nitrate, ammonium, BC and OC (D'Almeida et al., 1991), with median radius of 0.07  $\mu$ m, 0.07  $\mu$ m, 0.07  $\mu$ m, 0.01  $\mu$ m and 0.02  $\mu$ m, and geometric standard deviations of 2.0, 2.0, 2.0, 2.0 and 2.2, respectively. Soil dust deflation and sea salt generation are calculated using the same schemes as that in Han et al. (2004), but instead of the 10 size bins for dust and sea salt, 5 size bins (0.1–1.0, 1.0–2.0, 2.0–4.0, 4.0–8.0, 8.0–20.0  $\mu$ m) are used in this study to save CPU time. Dust deflation is calculated with an empirical model, depending on a series of controlling parameters, like threshold friction velocity, threshold

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