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Sensitivity of climate effects of black carbon in China to its size distributions

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ABSTRACT

The climate effects of black carbon (BC) aerosols are sensitive to BC size distributions and this sensitivity over China is studied using a regional climate model, namely RIEMS2.0. A new size-resolved scheme is developed based on observational data. The simulated BC concentrations with the new scheme are better compared with the observation than the previous uniform scheme, which is likely to overestimate BC concentrations, radiative forcings, and warming effects in many regions of China due to its simple assumption on BC size. The simulation with the size-resolved scheme suggests a reduction of the all-sky radiative forcing of BC at the top of atmosphere (TOA) by 0–0.25 W m^{-2} over the most study domain. Correspondingly, the warming effect of BC is weakened by −0.04 to −0.16 K over most parts of South China and North China. The difference in BC-induced precipitation between the two schemes varies irregularly from region to region, ranging from −2.8 to 2.8 mm d^{-1}. With the size-resolved scheme, the BC radiative properties and the climate effects are reassessed and the means (ranges) over the study domain are summarized as follows. The annual mean surface concentration of BC is 0.88 μg/m^3, ranging from 1 to 8 μg/m^3 over North China and Central China. The all-sky and clear-sky radiative forcings of BC at the TOA are 0.43 and 0.39 W/m^2, respectively. Over most parts of Southwest China, Central China, and North China, the BC warming effect prevails, with enhanced temperature of 0.04–0.28 K. BC aerosols usually enhance precipitation in South China and North China, ranging from 0.40 to 2.8 mm d^{-1}.

1. Introduction

Atmospheric aerosols can alter the earth's radiation directly and indirectly. The direct effect refers to the fact that aerosols can intercept, scatter, and absorb radiation, while the indirect effect changes cloud microphysical properties such as cloud albedo, cloud droplet number, size and lifetime, as aerosols can act as cloud condensation nuclei (CCN) (Ramanathan et al., 2001; Oshima et al., 2013). For this reason, aerosols, especially anthropogenic aerosols, have been a topic of active research in environmental sciences, including atmospheric science.

According to IPCC (2013), radiation forcing by anthropogenic aerosols is estimated to be −0.45 W/m^2 and the uncertainty on this estimate is substantially large. Chang et al. (2015) studied the uncertainties in anthropogenic aerosol concentrations and direct radiative facings by emission over China. Direct radiative forcing of anthropogenic aerosols with different mixing states over China has been studied by Zhuang et al. (2013a, 2013b) In recent years, Ji et al. have made a series of studies on regional anthropogenic aerosols. Radiative effects of mineral aerosols over High Mountain Asia were investigated over a long time by them (Ji et al., 2016). They also made a simulation on carbonaceous aerosols for studying their chemical progress and climate effects (Ji et al., 2015). In addition, they (Ji et al., 2011) used the regional climate model to analyze the effects of anthropogenic aerosols over South Asia. Among all aerosols, BC aerosols can effectively absorb solar radiation at visible and infrared wavelengths and thus heat atmosphere, influencing both climate and air quality (Ramanathan and Carmichael, 2008; Bond et al., 2013). BC aerosols are generated from incomplete combustion of carbonaceous matters including fossil fuel, biomass and biofuels (Streets et al., 2003; Bond et al., 2004). On global average, BC aerosols are estimated to have a direct radiative forcing (DRF) of 0.4 W/m^2 (IPCC, 2013). The regional radiative forcing (RF) of BC has much larger uncertainty due to the surface albedo effect (Vignati et al., 2010). BC aerosols have a high degree of inhomogeneity in time and space so that their distributions vary largely from region to region, making it difficult to assess their impacts on regional climate.

Jacobson (2002) found that control of fossil-fuel induced black carbon and organic matter can be the most effective way to slow down the global warming. As is known, coal consumption is one of the major anthropogenic sources of black carbon (Penner et al., 1993). As coal industry is one of dominant energy sectors in China, it is not surprising that one-fourth of the global anthropogenic BC is emitted there (Cooke et al., 1999; Streets et al., 2001). Recent economic

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development in China further leads to an increase in BC emission, bringing pervasive attention to BC in China internationally.

Although fresh BC aerosols are hydrophobic, they are usually coated by water-soluble material after emission (Moteki and Kondo, 2007) and thus act as CCNs. The shift of BC from a hydrophobic state to a more hydrophilic state is caused by the aging process, in which BC aerosols undergo chemical and physical transformations that alter their absorption properties and radiative forcings (Seinfeld and Pandis, 2006; Kuwata et al., 2009). The aging process depends greatly on the size distribution and mass concentrations of BC aerosols, both of which can alter the climate effect of BC aerosols (Matsui, 2016). Some studies have shown that ignoring the size distributions of carbonaceous aerosols can cause high uncertainty in predicting CCN concentrations (Pierce et al., 2007; Pierce and Adams, 2008; Spracklen et al., 2011). The climate forcing of carbonaceous aerosols varies greatly with their size distribution (Bauer et al., 2010; Spracklen et al., 2011). Spracklen et al. (2011) found the mean indirect effect of carbonaceous aerosols at the top of atmosphere varied from -0.34 to -1.08 W/m^2, depending on the assumed size distributions of the aerosols in the simulation model. Although these studies have suggested that different size distributions of BC may impact BC radiative properties, the magnitudes and variations of such an effect remains uncertain, especially on the regional scale. These studies are mostly focused on the global scale and the regional modelings are few. Considering that BC in China is a large concern to the global radiation forcing and the climate effects of BC is more significant on the regional scale than on the global scale (Wang, 2004), it is necessary to study such an effect using a regional climate model over China. Several previous studies with RIEMS2.0 still used a uniform size distribution scheme (Zhang et al., 2011). In the simulation, the size setting of BC emission plays a critical role in assessing the climate effect of BC. Therefore, we can modify the size distribution of BC in the model according to the measurement data, which have become available taking advantage of recent new techniques that can measure BC particles in different sizes.

In this study, we will examine how BC concentrations and associated radiative properties and climate effects change with different size distributions of BC aerosols using a climate model RIEMS2.0. A size-resolved scheme based on observational data will be developed (Section 3) and compared with the previous uniform size scheme (Section 4). Finally, the size-resolved scheme will be used to analyze the climate effects of BC (Section 5). The climate model RIEMS2.0 will be described in Section 2 and the conclusions will be provided in Section 6.

2. Model descriptions and experiment design

2.1. Mesoscale numerical model RIEMS2.0

The climate model, namely the Regional Integrated Environmental Model System (RIEMS2.0), was developed by the Key Laboratory of Regional Climate-Environment for Temperate East Asia (RCE-TEA), Chinese Academy of Sciences, based on RIEMS version 1.0 (Fu et al., 2000). RIEMS2.0 includes detailed description of chemical, hydrological, and biological processes related to the climate system, originally adopted from the fifth-generation Penn-sylvania State University/National Center for Atmospheric Research mesoscale model (MM5v3) (Fu et al., 2000).

The chemical transport module and the aerosol module developed by Liu et al. (2010) are coupled with the RIEMS2.0 online (Liu et al., 2001). The aerosol module in this version involves sulfate, nitrate, BC, organic carbon, primary and secondary OC, dust and sea salt aerosols, among which, BC, dust sea salt aerosols and primary OC are primary aerosols. The chemical transport model considers both physical and chemical processes, including photochemistry, gas and aqueous phase chemistry, dry and wet deposition, transport and diffusion. For the primary aerosols, emission, transport, diffusion and dry (wet) deposition are considered but the chemical processes are excluded. However, for secondary aerosols, the chemical processes are considered because of their importance. The chemistry transport model involves the CBM4 chemical mechanism (Carbon Bond Mechanism) (Gery et al., 1989) with 36 species and 83 chemical reactions. In order to compute sulfate, nitrate and secondary organic aerosols, the aerosol model adopted the organic and inorganic aerosol models: the thermodynamics equilibrium model ISORROPIA (CMAXS) (Hildemann et al., 1984) and the secondary organic model, which is from the air-quality model REMSAD (Regional Model System for Aerosols and Deposition) developed by U.S. Environment Protection Agency (Odum et al., 1996; Griffin et al., 1999). RIEMS has participated in the Regional Climate Model Intercomparison Project for Asia (Fu et al., 2005) and has been used to simulate regional climates in East Asia (Zhang et al., 2011; Han et al., 2012). RIEMS2.0 can simulate the climate over East Asia very well. Dong et al. (2014) used RIEMS2.0 to simulate the temperature in China and found that this model can show the spatial temperature distribution of temperature by comparison with observations. Zhao et al. (2013) also found the simulation of RIEMS2.0 could show similar distributions of SAT as observed data albeit a little cold bias over East Asia. In addition, RIEMS2.0 was used to analyze the Multi-Year mean precipitation and temperature in China (Zhao et al., 2009).

Three mixture states of BC were considered in the model, including external mixture, partly internal mixture and completely internal mixture. An external mixture state between several types of aerosols is assumed. The distribution of aerosol concentrations is computed by the Atmosphere chemical-aerosol model and then the results are input to the radiation module CCM3 for assessing the direct effect of aerosols. When computing the control case without the anthropogenic aerosols, only the background aerosols are considered. Here we have not taken the indirect effects of aerosols into consideration in this study.

The simulation domain is centered at 32°N and 107°E, with 55 (south to north) × 56 (west to east) grids at 60 km resolution. There are 16 vertical layers from the surface to 100 hPa.

2.2. Meteorological and emission data

The NCEP/NCAR reanalysis data for 2010 at six-hour intervals were used as boundary conditions and the initial fields for the climate variables simulations. The emission inventory is for 2010, with a resolution of 0.25° by 0.25°, which is acquired from the Multi-resolution Emission Inventory for China (MEIC) of Tsinghua University, China (http://www.meicmodel.org). The inventory includes thirty-two chemical species, such as SO_2, BC, OC, and VOCs.

2.3. Numerical experiments

After setting the size scheme for black carbon in the model, the radiative effects of BC were assessed by running the radiation scheme twice in the model. The first run included all types of the aerosols while the second run excluded BC. Therefore, the radiative effects of BC are the difference between the two runs. By altering the size of BC in the model, the sensitivity of the climate impacts of BC aerosols to their sizes can be quantified over the entire China and by region. The regional delineations in this study are shown in Fig. 1, which is similar to Xiong (2001). Totally, twenty sensitivity experiments in different sizes and two experiments with different size schemes were run in this study.

The boundary conditions were set up by the hourly simulations from a global chemical transport model, GEOS-Chem (http://acmg.seas.harvard.edu/geos/), with both hydroscopic BC and non hydroscopic BC. The optical parameters of aerosols including single albedo, asymmetry and extinction coefficient were referred the database at UCAR. (http://www.ccsm.ucar.edu/models/atm-cam/download/). All simulations are integrated for the period of 1 January to 31 December of year 2010 after a 3-day spin up of the model.