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Simulation of atmospheric aerosols in East Asia using modeling system RAMS-CMAQ: Model evaluation

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Abstract

The modeling system RAMS-CMAQ is applied in this paper to East Asia to simulate the temporo-spatial concentration distributions of atmospheric aerosols. For evaluating its performances, modeled concentrations of aerosols such as sulfate, nitrate, ammonium, black carbon and organic carbon were compared with observations obtained in East Asia on board of two aircrafts in the springtime of 2001. The comparison showed generally good agreement, and, in particular, that the modeling system captured most of the important observed features, including vertical gradients of the aerosols of the Asian outflow over the western Pacific. The evaluation results provide us with much confidence for further use of the modeling system to investigate the transport and transformation processes of atmospheric aerosols over East Asia and to assess their impacts on the Earth's radiation budget.

Keywords: East Asia; CMAQ; TRACE-P; ACE-Asia

1. Introduction

East Asia is an important source region of all major tropospheric aerosol types. It has been estimated that fast economic development, large areas of desert, intensive forest and agriculture fires in the region contribute to one-fourth to one-third of total global emissions of SO_2, organic matter, soot, and dust (e.g., Chin, Rood, Lin, Muller, & Thompson, 2000). Aerosol particles influence climate by modifying the global energy balance through absorption and scattering of radiation (direct effects) and by modifying the reflectance and persistence of clouds and the development and occurrence of precipitation (indirect effects). Some studies (e.g., Menon, Hansen, Nazarenko, & Luo, 2002) indicated that increases in precipitation and floods in the southern part of China and in droughts in the northern part of China were probably associated with the increase in black carbon (BC) concentrations. Beside their important role in changing the Earth's radiation budget, aerosol particles have strong impacts on environment, for example, heavy air pollutions in many Chinese urban areas associated with particulate matters in recent years (e.g., Zhang, Wang, Sheng, Kanai, & Ohta, 2004), acidification of lakes and forests through deposition of sulfate (ASO_4) and nitrate (ANO_3), and fertilization of land and oceans through deposition of nitrate and other nutrients. In addition, heterogeneous reactions on the surface of aerosols are shown to have significant impact on tropospheric chemistry (Jacob, 2000).

Although in situ measurements and satellite- and ground-based remote sensing provide essential information regarding aerosol loading, geographical and vertical distribution, processes and influences, such measurements are necessarily limited in space and time and are limited in their ability to distinguish natural and anthropogenic aerosol components. Chemical transport models are a critical element in aerosol radiative forcing studies as they are the only means by which the atmospheric aerosol can be partitioned into natural and anthropogenic com-

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ponents. They also provide a prognostic capability of exploring the effects of increasing or decreasing aerosol precursor emissions on atmospheric aerosol concentrations.

East Asia encompasses some of the most complex gas-particle atmospheric dynamics on the Earth. Gas-phase emissions of organics, NO_x, and SO_2 from the Asian continent undergo photo-oxidation as air masses are advected eastward over the Pacific. Gas-to-particle conversion occurs as condensable species are produced in the gas phase. These continental outflows contain anthropogenically derived particles as well as wind-blown mineral dust. In the continental outflow region, primary aerosols of mineral dust and sea salt origin, and the continental anthropogenic aerosols, are transformed by gas–aerosol interactions. To accurately calculate an atmospheric aerosol distribution, chemical transport models must incorporate a quantitative parameterization of precursor gas and aerosol emissions and the processes controlling aerosol formation, transport, chemical transformation and removal (Huebert et al., 2003).

The Models-3 Community Multi-scale Air Quality modeling system (CMAQ) is an Eulerian-type model that simulates concurrently the atmospheric and land processes affecting the transport, transformation and deposition of air pollutants and their precursors on both regional and urban scales (Byun & Ching, 1999), and the Regional Atmospheric Modeling System (RAMS) is a highly versatile numerical code for simulating and forecasting meteorological phenomena (Pielke et al., 1992). We applied the modeling system RAMS-CMAQ to the Asian Pacific region in different seasons to investigate the temporo-spatial variations in concentrations of atmospheric aerosols over East Asia and to examine the model's ability in treating complex reactions during the formation processes of ANO_3, ASO_4, ammonium (ANH_4) and organic carbon (OC) aerosols in East Asia (Zhang, 2004; Zhang, Uno, et al., 2004; Zhang, Xu, Zhang, & Han, 2005). In this paper we limit ourselves to show to what extent the modeling system can reproduce actual concentration distributions of atmospheric aerosols by comparing modeled mixing ratios of ANO_3, ASO_4, ANH_4, BC and OC with observations obtained in East Asia on board of aircrafts during the transport and chemical evolution over the Pacific (TRACE-P; Jacob et al., 2003) and the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia; Huebert et al., 2003) field campaigns, which were conducted consecutively in the springtime of 2001.

2. Model description

The modeling system RAMS-CMAQ has two major components. CMAQ performs simulations following first principles and employs a "one atmosphere" philosophy that tackles the complex interactions not only among multiple atmospheric pollutants, but also between regional and urban scales. In the modeling system, CMAQ uses meteorological fields from RAMS instead of its default meteorological driver, the Mesoscale Modeling system (MM5) to get better resolved atmospheric boundary structure (Zhang, Uno, et al., 2004).

In this study CMAQ is configured with the chemical mechanism of the Regional Acid Deposition Model version 2 (RADM2), including gas-phase and aqueous chemistry and aerosol processes of coagulation, particle growth by the addition of new mass, particle formation, etc., and having been extended to include direct estimates of the primary emissions of BC, OC, sea salt and wind-blown dust (Zhang, 2004; Zhang et al., 2005). These particles are divided into two groups, fine particles and coarse particles, and the fine group contains two interacting modes, i.e., i-mode (nuclei or Aitken) representing fresh particles either from nucleation or from direct emission, and j-mode (accumulation) standing for aged particles. The size distribution of aerosol particles is represented as the superposition of three lognormal sub-distributions (Byun & Ching, 1999).

For the CMAQ inputs, emissions of nitrogen oxides, sulfur dioxide, carbon monoxide, non-methane hydrocarbon species and ammonia from anthropogenic activities and open biomass burning were obtained from the 0.5×0.5 monthly inventory (Streets et al., 2003). Nitrogen oxides and ammonia from soil were derived from the Global Emissions Inventory Activity (GEIA) 1×1 monthly global inventory (Benkovitz et al., 1996). Aircraft emissions were based on the Emission Database for Global Atmospheric Research (EDGAR; Olivier et al., 1996). Sulfur dioxide emissions arising from volcanoes were based on the estimates by Streets et al. (2003). These different emission inventories are then preprocessed to form a unified inventory with the same spatial resolution of CMAQ.

RAMS was exercised in a four-dimensional data assimilation mode using analysis nudging with re-initialization every 4 days, leaving the first 24 h as the initialization period. The three-dimensional meteorological fields for the RAMS input were obtained from the European Center for Medium-Range Weather Forecasts (ECMWF) analyzed datasets, and were available every 6 h with 1×1 resolution. The sea surface temperatures (SST) based on weekly mean values and the observed monthly snow-cover information were adopted to set the boundary conditions for the RAMS calculation.

The model domain (shown in Fig. 1) is 6240 km × 5440 km on a rotated polar-stereographic map projection centered at (25°N, 115°E) with 80 km mesh. RAMS and CMAQ have the same model height. There are 23 vertical layers for RAMS in the σ_z coordinates system unequally spaced from the ground to ~23 km, with about 9 layers concentrated in the lowest 2 km of the atmosphere in order to resolve the planetary boundary layer, while there are 14 levels for CMAQ with the lowest 7 layers being the same as those in RAMS.

Initial and boundary conditions of chemical species in CMAQ were chosen to reflect the East Asian situation. Recent measurements were used whenever possible. To evaluate the impact of the anthropogenic emissions on the distributions of trace gases and aerosols, the initial and boundary conditions were generally chosen at the lower end of their observed range (e.g., the northern and western boundary conditions for ASO_4, ANO_3, ANH_4, BC and OC were 1.0, 0.5, 0.2, 0.1 and 0.1 $\mu g/m^3$, respectively) so as to allow the emissions and chemical reactions to bring them closer to their actual values during the initialization period.