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Modeling nitrate aerosol distributions and its direct radiative forcing in East Asia with RAMS-CMAQ

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ABSTRACT

The geographical and seasonal characteristics in nitrate aerosol and its direct radiative forcing over East Asia are analyzed by using the air quality modeling system RAMS-CMAQ coupled with an aerosol optical properties/radiative transfer module. For evaluating the model performance, nitrate ion concentration in precipitation, and mixing ratios of PM_{10}, and some gas precursors of aerosol during the whole year of 2007 are compared against surface observations at 17 stations located in Japan, Korea, and China, and the satellite retrieved NO_2 columns. The comparison shows that the simulated values are generally in good agreement with the observed ones. Simulated monthly averaged values are mostly within a factor of 2 of the measurements at the observation stations. The distribution patterns of NO_2 from simulation and satellite measurement are also similar with each other. Analysis of the distribution features of monthly and yearly averaged mass concentration and direct radiative forcing (DRF) of nitrate indicates that the nitrate aerosol could reach about 25–30% of the total aerosol mass concentration and DRF in Sichuan Basin, Southeast China, and East China where the high mass burden of all major aerosols concentrated. The highest mass concentration and strongest DRF of nitrate could exceed 40 $\mu g/m^3$ and $-5\ W/m^2$, respectively. It also indicates that other aerosol species, such as carbonaceous and mineral particles, could obviously influence the nitrate DRF for they are often internally mixed with each other.

1. Introduction

Nitrogen oxides are important chemical species in both the stratosphere and troposphere playing a key role in catalytic production of ozone chemistry, gas-to-particle reaction, and other chemical processes. The nitrate aerosol is a secondary aerosol produced from nitrogen oxides and causes many environmental and climatic issues such as acid rain (Brimblecombe & Stedman, 1982), atmospheric haze (Deng et al., 2008; Wang, Zhuang, Sun, & An, 2006), radiation balance change, and cloud droplet formation (Li & Han, 2011; Li, Wang, Zhuang, & Han, 2009; Zhang, Wang, Guo, & Wang, 2009). The soluble aerosol particles, including nitrate aerosol, also act a part in the core-mantle particles by coating on mineral or black carbon particles (Mishchenko, Liu, Travis, & Lacis, 2004). This internal mixing state could obviously influence the radiative properties by enhancing the absorbing ability and then weakening the negative radiative forcing (Bauer et al., 2007; Jacobson, 2001). As a result of the rapid development of industry and commerce, present air pollution problems caused by aerosol and its gas precursor are becoming more and more serious in East Asia (Zhang, Han, & Zhu, 2007; Zhang, Han, Cheng, & Tao, 2009). Meanwhile, nitrogen oxide emissions from Asia are larger than those from Europe and North America, and they are expected to continue to increase (Akimoto, 2003). Especially anthropogenic emissions associated with fossil fuel burning in China have grown significantly as they experienced a period of rapid economic development and industrial expansion over the last three decades (*e.g.*, Richter, Burrows, Nüss, Granier, & Niemeier, 2005; Wu, Jiang, Liu, & Tang, 2002; Zhang, Streets, et al., 2007). Thus, full

Abbreviations: ALDX, higher aldehyde; AOD, aerosol optical depth; CAM, community atmosphere model; CAMRT, radiative transfer scheme of community atmosphere model; CB05 or CB-IV, Carbon Bond mechanism; CMAQ, community multiscale air quality; DRF, direct radiative forcing; EANET, Acid Deposition Monitoring Network in East Asia; EDGAR, Emission Database for Global Atmospheric Research; GEIA, Global Emissions Inventory Activity; INTEX-B, Intercontinental Chemical Transport Experiment-Phase B; ISORROPIA, inorganic aerosol thermodynamic equilibrium model; MOZART-4, Model for Ozone and Related Chemical Tracers, version 4; NTR, organic nitrate; OMI, Ozone Monitoring Instrument; RAMS, regional atmospheric modeling system; CMAQ, regional air quality modeling system; RAQM, regional air quality Eulerian model; REAS, Regional Emission inventory in Asia; RegCM, regional climate model; SSA, single scattering albedo; TOA, top-of-atmosphere; VOC, volatile organic compound.

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consideration of nitrate aerosol should be important in the study about the atmospheric environment and regional climate change in East Asia.

The modeling system which is capable of resolving the transport, transformation, and deposition mechanisms of nitrogen compounds could help to capture the comprehensive information of associated effects of nitrate aerosol. In recent years, modeling studies have paid more attention to this subject in East Asia. An et al. (2002) used the regional air quality Eulerian model (RAQM) to simulate four monthly nitrate concentrations in precipitation. Zhang, Gao, Ge, and Xu (2007) used the air quality modeling system named as regional atmospheric modeling system and community multiscale air quality model (RAMS-CMAQ) to reproduce the seasonal variation of nitrate mass burden. Li et al. (2009) tried to calculate the first indirect radiative effects of nitrate aerosol in China by using the regional climate model (RegCM3). These studies provided the preliminary release of distribution features of partial nitrate characterization in East Asia. However, the investigation about environmental influence and direct radiative forcing of nitrate is still insufficient for only few relative works have focused on it. Additionally, since the sulfate aerosol, absorbing soot, organic carbon, and soil dust also significantly scatter or absorb the atmospheric radiation flux, previous study has suggested that it is necessary to take all major aerosol compositions into account even though the physical and optical properties of one single aerosol species were simulated and studied (Ghan et al., 2001).

In this paper, the nitrate mass concentration and direct radiative forcing (DRF) over East Asia in the whole year of 2007 are investigated by using the regional air quality modeling system RAMS-CMAQ coupled with an aerosol optical properties/radiative transfer module. All major aerosol components, including sulfate, ammonium, black carbon, organic carbon, dust, and sea salt, are considered in this modeling system, besides nitrate. The regional features of nitrate mass burden and DRF are focused upon and analyzed. The paper is organized as follows. The modeling system is described in Section 2. In Section 3, the modeled results are evaluated by comparing with the observation data, and the geographical and seasonal distributions of nitrate aerosol concentration and DRF over East Asia are discussed. Section 4 presents the conclusions.

2. Model description

The regional air quality model CMAQ is used in this study to describe the physical and chemical processes of aerosol emission, coagulation, dry and wet deposition, formation of secondary particles from precursors, and other important characteristics. In this study, the updated and expanded version of the Carbon Bond mechanism CB05 (Sarwar, Luecken, Yarwood, Whitten, & Carter, 2008) was applied to describe the mechanisms of gas-phase chemistry and address the vapor phase precursors. Compared with CB-IV, the new version CB05 mechanism adds several nitrogen oxides (NO_x) recycling reactions to improve the representation of the life time of NO_x over multiday scales (Zaveri & Peters, 1999). Additional radical reactions for improving nighttime chemistry of NO_3 removal are also included. The description of gas-phase consumption of HNO_3 is enhanced because of the inclusion of the photolysis of HNO_3 *via* reactions of $NO_3 + HO_2$, ALDX + NO_3, and NTR + OH with the CB05 mechanism. The CMAQ accounts for the gas-phase nitric acid (HNO_3) production *via* homogeneous and heterogeneous reactions which can then be partitioned to form aerosol nitrate. An inorganic aerosol thermodynamic equilibrium model, "ISORROPIA" is used to determine partitioning of inorganic aerosols in the model (Nenes, Pandis, & Pilinis, 1999). Except nitrate aerosol, sulfate, ammonium, black carbon, organic carbon, dust, and sea salt are also simulated in this modeling system.

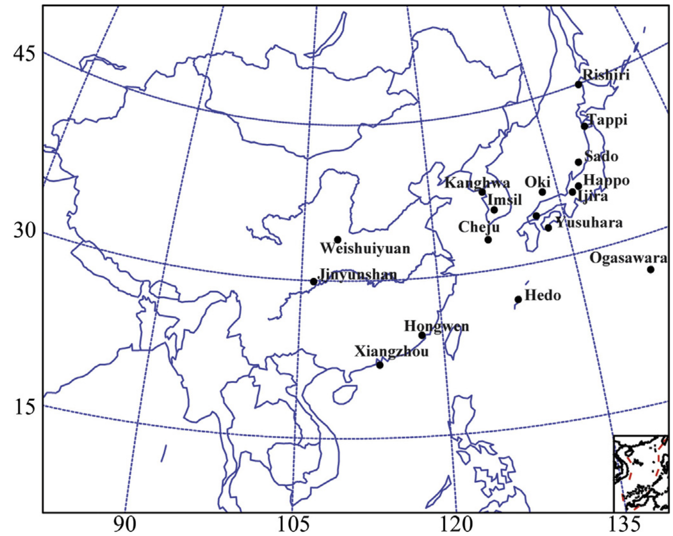

Fig. 1. Geographic locations of the EANET* observation sites in East Asia. *Acid Deposition Monitoring Network in East Asia.

The particle size distribution is represented as the superposition of three lognormal subdistributions: Aitken mode, accumulation mode, and coarse mode. Whereas internal mixing of the aerosol species is assumed within each mode, the modes themselves are externally mixed. The highly versatile numerical code RAMS which could well simulate the boundary layer and the underlying surface is used to provide the meteorological fields for CMAQ.

For the emission inventory, the emissions of nitrogen oxides, sulfur dioxide, carbon monoxide, black carbon, organic carbon from anthropogenic activities (power, industry, residential, and transportation) are obtained from the $0.5^\circ \times 0.5^\circ$ emission inventory for INTEX-B (http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html). The Regional Emission inventory for Asia domain (REAS, http://www.jamstec.go.jp/frsgc/research/d4/emission.htm) was used to provide the emission of NH_3. Nitrogen oxides and ammonia from soil are adopted from the Global Emissions Inventory Activity (GEIA) $1^\circ \times 1^\circ$ monthly global inventory (Benkovitz et al., 1996). Aircraft emissions are based on the Emission Database for Global Atmospheric Research (EDGAR; Olivier, Bouwman, Maas, & Berdowski, 1994). The sea salt and dust emissions were calculated on-line by using an algorithm from Gong (2003) and an empirical mechanism that performs well for describing dust emissions in East Asia (Han et al., 2004), respectively. The boundary conditions for the modeling system were obtained from the MOZART-4 (Pfiser et al., 2008) three-hour output data field. The model domain (Fig. 1) is 6654 km × 5440 km with 64 km grid cells on a rotated polar stereographic map projection centered at 35°N, 116°E. Previous researches have demonstrated that this modeling system performs well on simulating tropospheric ozone, aerosols, and other pollutants in East Asia (Han, Ge, Tao, Zhang, & Zhang, 2012; Zhang, Xu, Zhang, & Han, 2005; Zhang et al., 2006; Zhang, Streets, et al., 2007).

A parameterization of aerosol optical properties and a radiative transfer scheme are coupled into this modeling system. The parameterization could greatly simplify the calculation process of Mie theory by using a Chebyshev polynomial fitting. The water uptake and internal mixing state of aerosol were treated by Kohler theory (Pruppacher & Klett, 1997) and Maxwell–Garnett mixing rule (Chuang et al., 2002), respectively. In previous work, the comparison with satellite and ground-base *in situ* measurements showed that the modeled AOD and SSA are well consistent with observed results (Han, Zhang, Han, Xin, & Liu, 2011). The direct radiative