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Local and distant source contributions to secondary organic aerosol in the Beijing urban area in summer

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HIGHLIGHTS

- Few studies have focused on SOA contributions in a large area, especially in China.
- We extended the CAM_X v5.4, replacing the two-product approach by the VBS approach.
- The source contributions to SOA in Beijing were calculated for the first time.
- The VBS approach substantially improved hourly, daily, and monthly SOA simulations.
- Distant sources dominated for both anthropogenic and biogenic SOA in Beijing.

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ABSTRACT

Quantification of local and distant source contributions to particulate matter is a key issue to improving air quality in large urban areas, but few studies have focused on secondary organic aerosol (SOA) source contributions in a large area, especially in China. In this study, we extended the Comprehensive Air Quality Model with Extensions (CAM_X) version 5.4, replacing the two-product approach by the volatility basis-set (VBS) approach, with updated SOA yields based on smog chamber studies. The modules related to the computationally efficient particulate source apportionment technology (PSAT) used in CAM_X v5.4 were extended based on the volatility basis set (VBS) approach. The updated version of the CAM_X model was then used to calculate the local and distant source contributions to SOA in Beijing for the first time. The results indicated that the VBS approach substantially improved hourly, daily, and monthly SOA simulations, compared with the two-product approach and the observations. In August 2007, the local source contributions to anthropogenic and biogenic SOA in Beijing were 23.8% and 16.6%, respectively; distant sources dominated for both anthropogenic and biogenic SOA in Beijing: Northern Hebei, Middle Hebei, Northeast China, Inner Mongolia, Shandong, and Tianjin (including Xianghe) contributed 5.1% −18.2% to anthropogenic SOA in Beijing; whereas, Inner Mongolia, Northern Hebei, and Northeast China contributed 12.2%, 18.6%, and 10.1%, respectively, to biogenic SOA in Beijing. Additionally, other areas outside China respectively contributed 5.3% and 10.8% to anthropogenic and biogenic SOA in Beijing: this could be related to strong summer monsoon.

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1. Introduction

Secondary organic aerosol (SOA) impacts atmospheric visibility, human health, and climate change, and thus has attracted more and more attention in recent years (Kanakidou et al., 2005; Hallquist et al., 2009). SOA is formed from the oxidation of both anthropogenic and biogenic volatile organic compounds (VOCs) (Chung and Seinfeld, 2002; Kanakidou et al., 2005). SOA accounts for a large fraction of organic aerosol (OA) (Cao et al., 2004; Kanakidou et al., 2005; Zhang et al., 2007; De Gouw and Jimenez, 2009; Hallquist et al., 2009; Tsimpidi et al., 2010). A recent study

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showed that, during an extremely severe and persistent pollution event in China, the contribution of SOA and secondary inorganic aerosol (SIA) was of similar importance, with an SOA/SIA ratio ranging from 0.6 to 1.4 (Huang et al., 2014). However, SOA formation processes are very complicated, thus making it very difficult to identify SOA sources and simulate SOA concentrations in the atmosphere (Hallquist et al., 2009).

Representation of SOA formation is based on gas particle partitioning theory (Pankow, 1994) and historically parameterized using the two-product approach of Odum et al. (1996), which has been coupled into regional and global models (Chung and Seinfeld, 2002; Heald et al., 2005; Han et al., 2008; Jiang et al., 2012). The simulated SOA and OA concentrations using the two-product approach are substantially underestimated, when compared to the corresponding observations in most cases, particularly over urban areas (Matsui et al., 2009a; Jiang et al., 2012). Heald et al. (2005) pointed out that a large OA source in the free troposphere was not included in current models. Robinson et al. (2007) proposed a significant SOA source missing in traditional aerosol models. Ervens et al. (2008) and Fu et al. (2009) considered in-cloud SOA formation pathways in atmospheric models. To further improve SOA simulations, a volatility basis set (VBS) modeling approach was developed by Donahue et al. (2006). The VBS approach has been used in regional and global models (Murphy and Pandis, 2009; Tsimpidi et al., 2010; Shrivastava et al., 2011; Ahmadov et al., 2012; Skyllakou et al., 2014; Koo et al., 2014). Compared with the two-product approach, the VBS approach significantly enhances the simulated SOA concentrations and reduces the gap between the simulated and observed OA concentrations (Ahmadov et al., 2012).

With rapid economic development and large energy consumption, China has experienced severe environmental problems, especially air quality issues. Air pollution in China has become a regional problem, particularly in the Beijing–Tianjin–Hebei (BTH) region, and the Yangtze and Pearl River deltas. Quantification of the contributions of local and long range transported pollutants is a key issue for improving air quality in large urban areas. Many methods have been proposed to estimate the role of local and regional sources in different areas. The simplest method is the zero-out or brute-force method (An et al., 2007; Koo et al., 2009; Wang et al., 2012), which is computationally demanding. Dunker (1981) proposed the decoupled direct method (DDM) to calculate the concentration sensitivity to changes in emissions. DDM can provide local derivations of the simulated pollutant levels to model parameters. However, DDM is not suitable for large-scale perturbations (Dunker et al., 2002). Ying and Kleeman (2006) developed the source-oriented external mixture (SOEM) method by dividing each pollutant of interest into different source-specific species and tracking them separately through the model. The SOEM method can accurately track source contributions to pollutant concentrations but is computationally expensive. Wagstrom et al. (2008) developed a computationally efficient particulate source apportionment technology (PSAT) and used it as a first application in investigating the contribution of power plant SO_2 emissions to particulate sulfate concentrations in the Eastern United States. Koo et al. (2009) made a comparison of the PSAT and sensitivity analysis in the Comprehensive Air Quality Model with Extensions (CAM_X, http://www.camx.com) and showed that the DDM and PSAT present similar results only for pollutants that are linearly related with emissions but otherwise differ because of non-linearity and/or indirect effects. Wang et al. (2009) developed a tagged species source apportionment (TSSA) method in the Community Multiscale Air Quality modeling system, whereas Kwok et al. (2013) developed an integrated source apportionment method (ISAM). The TSSA is very similar to the PSAT, but unlike the PSAT, the TSSA adopts an "on-line" approach and explicitly solves tagged species using same algorithms as the host model for physical atmospheric processes like advection and diffusion. The ISAM implementation builds on and improves the structure developed for the TSSA. Recently, Hu et al. (2014) have developed a hybrid fine particulate matter (PM) source apportionment method based on a receptor model species balance and species specific source impacts from a chemical transport model. Skyllakou et al. (2014) calculated contributions of local and regional sources to fine PM in Paris using the PSAT and PMCAMx-2008.

Until now, few studies have focused on SOA source contributions in a large area, especially in China. In this work we first extend the CAM_X version 5.4. The two-product approach used in the CAM_X version 5.4 was replaced by the VBS approach, with updated SOA yields based on smog chamber studies (Murphy and Pandis, 2009). The modules related to the PSAT used in the CAM_X version 5.4 were extended based on the VBS approach. We then used the extended CAM_X version 5.4 to compute the contributions of local and distant sources to SOA in Beijing for the first time.

2. Methodology

2.1. Model setup

Three domains were used in the CAM_X version 5.4 as shown in Fig. 1. The Lambert projection was used with two true latitudes of 60°N and 30°N. Domain 1 covered most East Asia, with a horizontal resolution of 81 km. The horizontal resolutions of Domains 2 and 3 were 27 km and 9 km, respectively. Domain 3 covered the BTH region. There were 17 vertical model layers extending to the top of the troposphere, with the first model layer approximately 30 m above the ground. The SAPRC99 mechanism (Carter, 2000) was chosen as the gas phase chemistry. Aging of anthropogenic organic condensable vapors (VOC oxidation products that condense on particles) (Ahmadov et al., 2012) and chemical reactions of the hydroxyl radical (OH) with sesquiterpenes were added into the SAPRC99 mechanism. The aerosol module used a static two-mode (coarse and fine) scheme. Hourly gridded data of air temperature, relative humidity, air pressure, turbulent diffusivity, wind speed and direction, clouds and precipitation amounts were provided by the Weather Research and Forecast (WRF) model version 3.3 (Skamarock et al., 2008). The parameterization or solution schemes used in this study are listed in Table 1. The terrain and land-use data were obtained from the U.S. Geological Survey (USGS) database. Initial and boundary conditions for meteorological and chemical fields followed Li et al. (2011).

The simulated period was 25 July to 31 August 2007, including seven days of spin-up time. An hourly output frequency was used for both meteorological and chemical fields.

2.2. PSAT description

In our study, we used the PSAT (Wagstrom et al., 2008) to track different pollutant categories originating from different geographic regions and source types. The advantages of the PSAT are high efficiency and flexibility to study different source categories and regions. One fundamental assumption in the PSAT is that PM should be apportioned to the primary precursor for each type of PM. In the PSAT, the general approach to simulating change over a model time step Δt is illustrated for a chemical reaction A → B. The general equation for species production/destruction is:

$$a_i(t+\Delta t) = a_i(t) + \Delta A \frac{w_i a_i}{\sum w_i a_i} \quad (1)$$

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