Source apportionment of secondary organic aerosol in China using a regional source-oriented chemical transport model and two emission inventories☆

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ABSTRACT

A Community Multiscale Air Quality (CMAQ) model with source-oriented lumped SAPRC-11 (S11L) photochemical mechanism and secondary organic aerosol (SOA) module was applied to determine the contributions of anthropogenic and biogenic sources to SOA concentrations in China. A one-year simulation of 2013 using the Multi-resolution Emission Inventory for China (MEIC) shows that summer SOA are generally higher (10–15 μg m^{-3}) due to large contributions of biogenic (country average 60%) and industrial sources (17%). In winter, SOA formation was mostly due to anthropogenic emissions from industries (40%) and residential sources (38%). Emissions from other countries in southeast China account for approximately 14% of the SOA in both summer and winter, and 46% in spring due to elevated open biomass burning in southeast Asia. The Regional Emission inventory in ASia v2.1 (REAS2) was applied in this study for January and August 2013. Two sets of simulations with the REAS2 inventory were conducted using two different methods to speciate total non-methane carbon into model species. One approach uses total non-methane hydrocarbon (NMHC) emissions and representative speciation profiles from the SPECIATE database. The other approach retains the REAS2 speciated species that can be directly mapped to S11L model species and uses source specific splitting factors to map other REAS2 lumped NMHC species. Biogenic emissions are still the most significant contributor in summer based on these two sets of simulations. However, contributions from the transportation sector to SOA in January are predicted to be much more important based on the two REAS2 emission inventories (~30–40% vs. ~5% by MEIC), and contributions from residential sources according to REAS2 was much lower (~21–24% vs. ~42%). These discrepancies in source contributions to SOA need to be further investigated as the country seeks for optimal emission control strategies to fight severe air pollution.

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1. Introduction

Atmospheric fine particles are correlated with adverse health effects, visibility reduction, and climate change (Englert, 2004; Kaufman et al., 2002; Lu et al., 2015; Pope III and Dockery, 2006; Pui et al., 2014). In January 2013, China experienced a record-breaking severe haze event that covered over 1.4 million square kilometers and affected the health of more than 800 million people in North China Plain (NCP) and central eastern China (Huang et al., 2014; Sun et al., 2014; Wang et al., 2014; Xu et al., 2013). The daily concentrations of PM$_{2.5}$ (airborne particles with aerodynamic diameters less than 2.5 μm) in some cities in NCP such as Beijing and Shijiazhuang were reported as more than 500 μg m^{-3}. Recent field measurements show that organic carbon (OC) was one of the major components during haze events with contributions ranging from 35% to 80% (He et al., 2011; Hu et al., 2016b; Li et al., 2017; Shen

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et al., 2015; Sun et al., 2015; Zhang et al., 2017b; Zheng et al., 2016). It has been suggested that secondary organic aerosol (SOA) could account for a significant fraction of the total observed OC (Crippa et al., 2014; Hallquist et al., 2009; Zhang et al., 2007) but the magnitude and source contributions to SOA in China have not been extensively studied.

Several experimental studies have been reported to determine the magnitude of SOA in China recently. The amount of SOA in total organic aerosol can be estimated by routine measurements of elemental carbon (EC) and OC, using ratios of the mass concentrations of OC to EC collected at a single location over an extended period (Hu et al., 2015; Wu and Yu, 2016). While significant levels of SOA were estimated (Wang et al., 2015), the OC/EC ratio method is considered to have significant uncertainties due to its underlying assumptions. In addition, source-specific SOA organic tracers have been applied to determine the amount of SOA from aromatic and biogenic precursors in organic aerosols (Ding et al., 2012; Feng et al., 2013; Yuan et al., 2013). However, a comparison study showed that SOA concentrations estimated using the tracer method were much lower than those estimated by radiocarbon dating (Liu et al., 2014). Detailed analysis of the aerosol chemical composition using aerosol mass spectrometer (AMS) and the positive matrix factorization technique illustrated that oxygenated organic aerosols, which is considered as surrogates of SOA, could account for more than 50% of organic aerosols (Hu et al., 2016b; Huang et al., 2011; Zhang et al., 2017b). SOA determination based on field measurements is usually expensive and can only provide SOA estimation at the monitoring sites. Modeling analyses are typically needed to illustrate the spatial distribution and regional source apportionment of SOA.

Modeling studies of SOA formation in China reported in the literature range from relatively simple box models (Wang et al., 2013) to full-blown three-dimensional (3D) chemical transport models (CTMs) (Hu et al., 2017c; Li et al., 2015; Zhao et al., 2016). Hu et al. (2017c) simulated ground level SOA concentrations using a precursor-tracking method in a regional 3D CTM. The model includes extended isoprene gas phase reactions that lead to the production of epoxydiols of isoprene (IEPOX) and methacrylic acid epoxide (MAE), and an SOA mechanism with updated SOA yields and reactive surface uptake of dicarbonyls and IEPOX and MAE. Lin et al. (2016a) applied a CAMx model with SOA yields based on smog chamber studies (Murphy and Pandis, 2009) to investigate local and distant sources of SOA in Beijing during summertime. It was found that approximately 40% of SOA was from the local sources and the rest of the SOA was formed from distant sources. However, source contributions to SOA were not determined in these studies. The amount of SOA predicted in a CTM model also depends on the spatial and temporal distribution of precursor emissions. Analyses of several inventories showed that estimations of emissions of NOx, volatile organic compounds (VOCs) and NH_3 from China were highly uncertain, leading to different predictions of $PM_{2.5}$ and its chemical components and no single inventory showed definitively better performance than other inventories (Hu et al., 2017b). Thus, the uncertainty in predicted SOA concentrations and source contributions due to different emission inventories need to be further evaluated.

The overall objective of this study is to investigate the contributions of different emission sectors to SOA in China. Contributions of major anthropogenic (industries, power generation, residential sources, extraction and solvent utilization) and natural (biogenic and wildfire) VOC sources to SOA in China are determined using two different emission inventories. This is the first mechanistic SOA source apportionment study using a 3D source-oriented CTM for the entire China. While different estimations of emission inventories in China have been reported in previous studies (Hu et al.,

2017b; Saikawa et al., 2017), none of these studies focused on their impact on SOA predictions and source contributions. To the best of the authors' knowledge, this is the first study that investigates the sensitivity of source apportionment of SOA to different anthropogenic emission inventories in China.

2. Method

2.1. Model setup

The CTM used in this study is based on the Community Multiscale Air Quality (CMAQ) model v5.0.1 (Appel et al., 2013; Byun and Schere, 2006; Foley et al., 2010). An updated SAPRC-11 lumped gas phase photochemical mechanism (S11L) (Carter and Heo, 2013) and a revised SOA module with reactive surface uptake of dicarbonyls and isoprene epoxides are used to improve SOA predictions (Ying et al., 2015 and the refereces therein). Further updates of the SOA module regarding the mass yields of SOA to account for wall loss of organic vapor during chamber experiments were described in detail by Hu et al. (2017a,b,c). The S11L gas phase mechanism and the SOA module are further updated in this study to track emissions of precursors from different sources categories separately so that the formation of SOA from different sources can be directly determined. The general approach for SOA source tracking in source-oriented regional models has been previously described by Zhang and Ying (2011b) and Ying et al. (2015) and is summarized in section 2.2.

The model domain covers China and its neighboring countries in east and southeast Asia, with a horizontal resolution of 36 km. Details of the domain setup have been described by Hu et al. (2016a). Meteorological inputs for the CMAQ model were generated using the Weather Research and Forecasting (WRF) model v3.6.1 with the boundary and initial conditions from the National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Model Global Tropospheric Analyses dataset. In general, the WRF model has acceptable performance on meteorological parameters. The detailed validation of meteorology can also be found in Hu et al. (2016a).

The anthropogenic emissions are based on two widely-used regional inventories: the Regional Emission inventory in ASia v2.1 (REAS2) (Kurokawa et al., 2013) and the Multi-resolution Emission Inventory for China (MEIC) (He, 2012). More details of the two inventories and their processing to generate model ready emission input files are described in section 2.3. Biogenic emissions were generated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 (Guenther et al., 2006) with year-specific leaf area index (LAI) data from the 8-day Moderate Resolution Imaging Spectroradiometer (MODIS) LAI product (MOD15A2). MEGAN v2.1 generates emissions for 18 VOC classes natively and these species were further mapped to generate emissions of 23 SAPRC model species, including isoprene, monoterpenes and sesquiterpenes. More details of the MEGAN input data and its application to simulate biogenic emissions in China are available in Qiao et al. (2015a). The Fire Inventory from National Center for Atmospheric Research (NCAR FINN) was used for open biomass burning emissions (Wiedinmyer et al., 2011). VOC emissions with SAPRC speciation are provided natively (http://bai.acom.ucar.edu/Data/fire/). More details of the FINN inventory can be found in Wiedinmyer et al. (2011) and from the other references on the FINN website (https://www2.acom.ucar.edu/modeling/finn-fire-inventory-ncar). Dust (Appel et al., 2013) and sea salt (Kelly et al., 2010) emissions were generated in line during the CMAQ simulations. In the updated CMAQ model used in this study, dust emission module was updated to be compatible with the 20-category MODIS land use data (Hu et al., 2015).