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# Development of an on-line source-tagged model for sulfate, nitrate and ammonium: A modeling study for highly polluted periods in Shanghai, China

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ABSTRACT

An on-line source-tagged model coupled with an air quality model (Nested Air Quality Prediction Model System, NAQPMS) was applied to estimate source contributions of primary and secondary sulfate, nitrate and ammonium (S–N–A) during a representative winter period in Shanghai. This source-tagged model system could simultaneously track spatial and temporal sources of S–N–A, which were apportioned to their respective primary precursors in a simulation run. The results indicate that in the study period, local emissions in Shanghai accounted for over 20% of S–N–A contributions and that Jiangsu and Shandong were the two major non-local sources. In particular, non-local emissions had higher contributions during recorded pollution periods. This suggests that the transportation of pollutants plays a key role in air pollution in Shanghai. The temporal contributions show that the emissions from the "current day" (emission contribution from the current day during which the model was simulating) contributed 60%–70% of the sulfate and ammonium concentrations but only 10%–20% of the nitrate concentration, while the previous days' contributions increased during the recorded pollution periods. Emissions that were released within three days contributed over 85% averagely for S–N–A in January 2013. To evaluate the source-tagged model system, the results were compared by sensitivity analysis (emission perturbation of −30%) and backward trajectory analysis. The consistency of the comparison results indicated that the source-tagged model system can track sources of S–N–A with reasonable accuracy.

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## 1. Introduction

In December 2013, a series of severe particulate matter (PM) pollution events occurred, where air quality indices (AQI) over 20 provinces (100 + cities) corresponded to heavy or severe pollution. Fine particulate matter ($PM_{2.5}$) pollution levels were monitored continuously over 9 days in Shanghai, and the maximum hourly $PM_{2.5}$ concentrations reached 602 μg/m$^3$ (Wu et al., 2015). In recent years, the occurrence of these types of pollution events has increased in China, with characteristically high pollution levels experienced for sustained periods of time over large areas.

High $PM_{2.5}$ concentrations, a major contributor to air pollution, create problems for human health and deteriorate visibility (Klemm et al., 2000; Ying et al., 2004; Tie et al., 2009). Previous studies on $PM_{2.5}$ have demonstrated that sulfate, nitrate and ammonium (S–N–A) are its main constituents and that most of these S–N–A ions are secondary aerosols generated through chemical reactions (Stockwell et al., 2000; Lee et al., 2008). S–N–A ions account for over half the components in $PM_{2.5}$ concentrations, making them a critical factor in high air pollution (He et al., 2001; Yao et al., 2003). Additionally, S–N–A ions have strong light scattering effects that lead to a deterioration of visibility (Ying et al., 2004; Dzubay et al., 1982; Appel et al., 1985; Chan et al., 1999). Therefore, it is necessary to study the formation and source contributions of S–N–A to effectively regulate PM emissions and its

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precursors.

Chemical transport models are useful for conducting research on source contributions because they explicitly simulate whole atmospheric processes, such as emissions, transport, removal, chemistry and aerosol physics. There are several methods used to study PM source contributions. Sensitivity analysis is a widely used approach in which sources are quantified by perturbing emissions one at a time and simulating the difference in pollutant concentrations compared to unperturbed control scenarios (Streets et al., 2007; An et al., 2007; Chen et al., 2007). Sensitivity experiments require large amounts of computing resources because each scenario of interest is run individually. This method is not always practical for studies on secondary pollutants due to its non-linear responses to emission changes (Dunker et al., 2002; Koo et al., 2009). The decoupled direct method (DDM) directly solves sensitivity equations derived from the governing equations of the atmospheric processes modeled in the host model. The DDM is an efficient and accurate alternative to sensitivity analyses (Dunker, 1981; Hakami et al., 2003). However, DDM may not be suitable for the analysis of large (e.g., above 50%) perturbations and secondary PM emissions (Dunker et al., 2002). The source-oriented external mixture (SOEM) method categorizes the pollutants of interest into source-specific species and tracks them separately through the simulated atmospheric processes in the model (Ying and Kleeman, 2006). This method is computationally demanding because it implements extra solutions to differential equations for gas and aerosol chemistry modules (Kleeman and Cass, 2001). A more efficient approach to tracking both primary and secondary PM emissions is the particulate source apportionment technology (PSAT, Wagstrom et al., 2008). PSAT utilizes reactive tracers to categorize PM components into different source classes. Similar to the source apportionment tool, the integrated source apportionment method (ISAM) is utilized in the CMAQ model (Kwok et al., 2013). Different methods probably produce different results, so there is no “true” value to which all results can be compared (Koo et al., 2009). Previous studies have predominantly focused on the source of emission types and locations, and only a few studies have discussed the temporal sources of PMs when its precursors or primary pollutants are emitted into the atmosphere (Wagstrom and Pandis, 2009). It is important to understand the temporal contributions of PMs, as this can provide governments with specific guidance regarding when to conduct emission reduction measures. Therefore, there is a need to develop a more accurate and computationally efficient technique for studying source contributions in air quality models.

An unprecedented severe haze occurred over mid-eastern China in January 2013 (Ji et al., 2014). The hourly $PM_{2.5}$ in Shanghai, Nanjing and Hangzhou exceeded 250 μg/m3, which were considered the most serious pollution events since 2000 (with the exception of dust episodes). In this work, we describe the development of an on-line source-tagged model that is applied to studies on the source contributions of S–N–A ions during the heavy-polluted events in January 2013. This on-line source-tagged model coupled with Nested Air Quality Prediction Model System (NAQPMS) simultaneously tracks spatial and temporal source contributions of S–N–A ions, which are categorized into their respective primary precursors. A new dynamical temporal source-tagged method is introduced to mark the period when the primary pollutants are emitted to the atmosphere. To evaluate the accuracy of the on-line source-tagged model, the obtained results were compared via a sensitivity analysis with a −30% emission perturbation and a backward trajectory analysis.

## 2. Materials and methods

### 2.1. Description of the model and experimental setup

A three-dimensional regional Eulerian chemical transport model, the Nested Air Quality Prediction Model System (NAQPMS) was utilized in this study. NAQPMS simulates physical and chemical processes with ambient pollutants that are solved using the mass balance equation in terrain-following coordinates (Wang et al., 2006; Li et al., 2007, 2008). Carbon-Bond Mechanism Z (CBM-Z) and an aerosol thermodynamic model (ISORROPIAI1.7) were employed in this model to calculate the gas and aerosol chemistries, respectively (Zaveri and Peters, 1999; Nenes et al., 1998). A bulk yield scheme of the secondary organic aerosols treated as six SOAs was embedded into NAQPMS (Li et al., 2011). Recent developments on the model can be found in Li et al. (2014).

Fig. 1 shows the model domain used in this study, which had 265 × 265 grid points on a Lambert conformal map projection with 9 km grid resolution. Vertically, NAQPMS used 20 terrain-following layers from the surface to 20 km, with the lowest 10 layers found below 2 km. The meteorological conditions were driven by WRF-ARW3.2.1, which used NCEP/NCAR FNL reanalysis data (1° × 1°) as initial/boundary conditions. Four-dimensional data assimilation (FDDA) (Otte, 2008), consisting of 6-h 3-D analyses of temperature, water vapor mixing ratio and horizontal wind components, was used with a nudging coefficient of $3.0 \times 10^{-4}$ for all the above components of this domain. The simulation was employed from 1 to 30 January 2013, using the first 4 days as the spin-up period.

The emissions applied in this study included anthropogenic and biogenic inventory data. The anthropogenic emission inventory in China was obtained from the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) with 0.25° × 0.25° resolution, while emissions in other countries were derived from the bottom-up Regional Emission inventory in Asia (REAS 2.1) with 0.25° × 0.25° resolution (Kurokawa et al., 2013). To improve the distribution of anthropogenic emissions, we spatially allocated the area emissions over East Asia based on relevant factors. For example, residential emissions were spatially allocated by population data; vehicle emissions were spatially allocated according to road length data; and agricultural emissions were allocated on the basis of cropland intensity data. Besides, biogenic emissions were obtained from a biogenic emission model (MEGANv2) provided by NCAR (http://accent.aero.jussieu.fr/database_table_inventories.php).

### 2.2. The on-line source-tagged model

Currently, chemical transport models (CTMs) treat emissions from all sources as a single entity, resulting in the loss of information on source contributions to pollutant concentrations. The on-line source-tagged model in NAQPMS aimed to assign pollutant concentrations to different sources at each step of the model without disturbing the original calculations. The source-tagged model that calculated the apportionment separately during the emissions, transport, wet/dry deposition, gas phase and aerosol chemistry calculations. There were 3 major processes in the on-line source-tagged model, which are shown in the calculation flow (Fig. 2).

During emissions processing, the on-line source-tagged model categorized each species into different sources. The equation (1) for the impact of PM emissions on the tracer concentrations ($Aer_i$) is:

$$Aer_i(t + \Delta t) = Aer_i(t) + E\_Aer_i \quad (1)$$

where $Aer_i(t + \Delta t)$ and $Aer_i(t)$ are the concentrations of primary PM

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