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Spatially and chemically resolved source apportionment analysis: Case study of high particulate matter event

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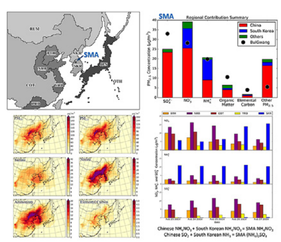
HIGHLIGHTS

- PM source contributions in Seoul, Korea during late February 2014 were analyzed.
- Domestic contributions grew throughout the episode.
- China and South Korea contributed 70% and 21% of $PM_{2.5}$.
- Major secondary $PM_{2.5}$ components were Chinese SO_4^{2-} and NO_3^- and Korean NH_4^+.
- PM controls for the SMA requires refined strategies in space, time, and chemicals.

GRAPHICAL ABSTRACT



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ABSTRACT

This article presents the results of a detailed source apportionment study of the high particulate matter (PM) event in the Seoul Metropolitan Area (SMA), South Korea, during late February 2014. Using the Comprehensive Air Quality Model with Extensions with its Particulate Source Apportionment Technology (CAMx-PSAT), we defined 10 source regions, including five in China, for spatially and chemically resolved analyses. During the event, the spatially averaged PM_{10} concentration at all PM_{10} monitors in the SMA was 129 μg/m^3, while the PM_{10} and $PM_{2.5}$ concentrations at the BulGwang Supersite were 143 μg/m^3 and 123 μg/m^3, respectively. CAMx-PSAT showed reasonably good PM model performance in both China and the SMA. For February 23–27, CAMx-PSAT estimated that Chinese contributions to the SMA PM_{10} and $PM_{2.5}$ were 84.3 μg/m^3 and 80.0 μg/m^3, respectively, or 64% and 70% of the respective totals, while South Korea's respective domestic contributions were 36.5 μg/m^3 and 23.3 μg/m^3. We observed that the spatiotemporal pattern of PM constituent concentrations and contributions did not necessarily follow that of total PM_{10} and $PM_{2.5}$ concentrations. For example, Beijing-Tianjin-Hebei produced high nitrate concentrations, but the two most-contributing regions to PM in the SMA were the Near Beijing area and South Korea. In addition, we noticed that the relative contributions from each region changed over time. We found that most ammonium mass that neutralized Chinese sulfate mass in the SMA came from South Korean sources, indicating that secondary inorganic aerosol in the SMA, especially ammonium sulfates, during this event resulted from different major precursors originating from different regions.

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1. Introduction

Particulate matter (PM) of 10 or 2.5 μm in diameter or less, termed PM_{10} or $PM_{2.5}$, respectively, has raised social policy concerns around the world for decades due to its significant adverse health effects, including premature deaths, identified in numerous studies and references therein (Lipfert, 1994; Vedal, 1997; Kaiser, 1997; Pope and Dockery, 2006; US EPA, 2009; Bell, 2012; Lim et al., 2012; Burnett et al., 2014; Dominici et al., 2014; Héroux et al., 2015). Particulate matter is a complex mixture of directly emitted substances (i.e., primary PM) and products formed chemically (i.e., secondary PM) through atmospheric reactions of their precursors (NARSTO, 2004; Seinfeld and Pandis, 2016). Understanding the causes of PM_{10} and $PM_{2.5}$ air quality problems in an area, therefore, requires assessing the contributions of both local and remote sources while considering the physicochemical processes of PM transport and formation (Stohl et al., 2002; NARSTO, 2004; Chin et al., 2007).

In South Korea, public awareness of serious PM pollution problems has been growing (Kwon et al., 2002; Kim et al., 2007; Chang et al., 2016; Richey and Ohn, 2016). To address these problems, the South Korean government has begun two major efforts. One is the development and operation of the nationwide year-round PM air quality forecast to help public make informed decisions about their outdoor activities since 2012 (Chang et al., 2016). The other is the development and implementation of air quality policies to lower domestic PM levels (Korea Ministry of Environment (2015)). Lowering domestic PM levels is made more difficult by the fact that high PM concentrations observed in South Korea are often linked to emissions from China (National Institute of Environmental Research, 2013; Oh et al., 2015; Shin et al., 2016; Wang et al., 2016). Wherever remote PM sources lie beyond the jurisdictional boundary of the receptor area's authority, as in South Korea, the development of effective control strategies becomes particularly complicated, because addressing air quality problems then requires collaborative efforts (Farrell and Keating, 1998; Dentener et al., 2010; Secretariat of Working Group for LTP project, 2011) This is exactly why the South Korean government has not only implemented local controls (Korea Ministry of Environment, 2015) but has also pursued international collaboration with neighboring countries, such as China and Japan including special campaigns such as ACE-Asia (Han et al., 2004; Seinfeld et al., 2004; Uno et al., 2004; Jung, 2016). Despite these efforts, South Korea still faces challenges in improving the accuracy of its air quality forecasting and in effectively developing a strategy for pollution control. PM is a complex material emitted from both foreign and domestic sources and formed out of precursors from both remote and local sources through atmospheric chemical reactions (NARSTO, 2004; Seinfeld and Pandis, 2016). Thus, improvements to emissions inventories and effective control strategies must account for the geographical origins of both primary PM emissions and precursor emissions for secondary PM.

In this study, we attempted to advance the understanding of the root causes of high PM events in South Korea by analyzing spatially and chemically resolved domestic and foreign source apportionment. As a case study, we conducted detailed source apportionment analyses for the high PM_{10} and $PM_{2.5}$ episode that occurred during late February 2014. This period was one of a few high PM events in 2014 that might not be associated with typical Asian Dust cases (Kim et al., 2016a) and showed the likely dominance of anthropogenic emissions. Over the past two years, several studies have examined this event, each focusing on limited aspects: meteorological characteristics of the event without refined chemical analysis, chemical compositions of observed PM during the event period without spatially specifying emissions sources, chemical analysis without a quantitative assessment of the impact of upwind country emissions on the air quality of downwind areas, or only total mass of PMs without considering PM speciation during the event (Yan et al., 2015; Kim et al., 2016b; Lin et al., 2016; Shin et al., 2016). Therefore, we performed a more comprehensive study of the event to identify the spatial and chemical characteristics of PM, using a photochemical grid model instrumented with a source apportionment tool based on a method of tagging chemical species. This approach can provide spatially specific and chemically resolved source apportionment information. For example, we can quantify how much sulfur dioxide (SO_2) emitted from one foreign source region and nitrogen oxides (NO_X) emitted from another foreign source region contribute to elevated sulfate (SO_4^{2-}) and nitrate (NO_3^-) concentrations observed in the Seoul Metropolitan Area (SMA), South Korea.

The remainder of this article is structured as follows. First, we describe the configuration of the model system, observational data, and overall performance of the modeling system. Next, we discuss the characteristics of foreign and domestic contributions to PM_{10} and $PM_{2.5}$ in the SMA, South Korea with respect to geographical origin and chemical composition. Finally, we identify the cause of high PM events during late February 2014 and propose future studies. Ultimately, we anticipate that the study framework presented here can be generally applicable in analyses on other high PM episodes around the world.

2. Model configuration and observational datasets

2.1. CAMx-PSAT modeling setup

For modeling, we used the Comprehensive Air Quality Model with Extensions (CAMx) with the Particulate Source Apportionment Technology (PSAT) tool (hereafter, CAMx-PSAT). CAMx-PSAT tracks pollutants in a computationally efficient fashion by tagging them based on their origin, including emissions sector and/or geography (Wagstrom et al., 2008; Ramboll-Environ, 2016). CAMx-PSAT tracks primary pollutants directly in the model. For secondary pollutants, it designates certain emitted species as tagged (e.g., SO_2 for sulfate), simulating their physical and chemical changes through the host model's physicochemical solvers. Therefore, CAMx-PSAT does not suffer from the significant non-linear effects that can occur with the brute-force method when large emission perturbations are applied (Yarwood et al., 2007; Wagstrom et al., 2008; Ramboll-Environ, 2016; Qu et al., 2016).

The modeling domains for this study comprise one (27-km) master grid and two (9- and 3-km) nested grids, as shown in Fig. 1. CAMx simulates air movement in these domains simultaneously (i.e., two-way nesting) for each time step to resolve transport and transformation correctly so that it can simulate re circulation between the finer-grid domains and the master grid. This approach can simulate regional transport more accurately than one-way nesting. The ambient air quality monitors, used to evaluate the performance of the modeling system, are marked in Fig. 1. During the chosen study period from February 22 to February 28, 2014, spatially averaged PM_{10} concentrations in the SMA were high, above the daily average 100 $\mu g/m^3$ "high" PM_{10} threshold used by Oh et al. (2015); the following section details the observed data, including that from monitor networks. For source apportionment analysis, 10 source regions were defined in the 27-km master domain, as shown in Fig. 2.

For meteorological model inputs, we used the Weather Research and Forecast (WRF) model (Skamarock and Klemp, 2008) v3.5.1, from which outputs were generated for routine air quality forecasting as part of the Integrated Multi-scale Air Quality System for Korea (IMAQS-K). IMAQS-K also uses various combinations of