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Identification of biased sectors in emission data using a combination of chemical transport model and receptor model

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HIGHLIGHTS

- Source apportionment of $PM_{2.5}$ was conducted on 11 sites in Japan by PMF and CMAQ.
- Contribution of major $PM_{2.5}$ emission sources by each model were well-harmonized.
- CMAQ/BFM underestimated the contribution from biomass burning against PMF.
- A comparison approach allows us to specify which emission sources have large biases.

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ABSTRACT

This study presented a comparison approach with multiple source apportionment methods to identify which sectors of emission data have large biases. The source apportionment methods for the comparison approach included both receptor and chemical transport models, which are widely used to quantify the impacts of emission sources on fine particulate matter of less than 2.5 μm in diameter ($PM_{2.5}$). We used daily chemical component concentration data in the year 2013, including data for water-soluble ions, elements, and carbonaceous species of $PM_{2.5}$ at 11 sites in the Kinki–Tokai district in Japan in order to apply the Positive Matrix Factorization (PMF) model for the source apportionment. Seven PMF factors of $PM_{2.5}$ were identified with the temporal and spatial variation patterns and also retained features of the sites. These factors comprised two types of secondary sulfate, road transportation, heavy oil combustion by ships, biomass burning, secondary nitrate, and soil and industrial dust, accounting for 46%, 17%, 7%, 14%, 13%, and 3% of the $PM_{2.5}$, respectively. The multiple-site data enabled a comprehensive identification of the $PM_{2.5}$ sources. For the same period, source contributions were estimated by air quality simulations using the Community Multiscale Air Quality model (CMAQ) with the brute-force method (BFM) for four source categories. Both models provided consistent results for the following three of the four source categories: secondary sulfates, road transportation, and heavy oil combustion sources. For these three target categories, the models' agreement was supported by the small differences and high correlations between the CMAQ/BFM- and PMF-estimated source contributions to the concentrations of $PM_{2.5}$, SO_4^{2-}, and EC. In contrast, contributions of the biomass burning sources apportioned by CMAQ/BFM were much lower than and little correlated with those captured by the PMF model, indicating large uncertainties in the biomass burning emissions used in the CMAQ simulations. Thus, this comparison approach using the two antithetical models enables us to identify which sectors of emission data have large biases for improvement of future air quality simulations.

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1. Introduction

Fine particulate matter of less than 2.5 μm in diameter ($PM_{2.5}$) suspended in the atmosphere has a serious impact on human mortality as well as respiratory and cardiovascular diseases (Talat et al., 2007; Schwartz et al., 2002; Dockery et al., 1993). $PM_{2.5}$ pollution results from various complex emission sources, which include not only domestic emission sources (e.g., industrial processes, road transportation, and other local sources) but also international sources owing to the long-range transport of air pollution (Shimadera et al., 2016). The specification of $PM_{2.5}$ sources is significant when planning strategies to reduce the $PM_{2.5}$ pollution level. Therefore, receptor models (RMs) and chemical transport models (CTMs) are widely used to identify the impacts of $PM_{2.5}$ sources because source impacts for $PM_{2.5}$ cannot be directly measured at observation sites. RMs are receptor-oriented modeling approaches, which are inferential, whereas CTMs are source-oriented modeling approaches, which are predictive of source contributions. Such source apportionment (SA) approaches have different strengths and limitations (Burr and Zhang, 2011; Hopke, 2016).

The Chemical Mass Balance (CMB) model and the Positive Matrix Factorization (PMF) model are types of RMs that solve the mass balance equations of the observed species concentrations to identify and quantify emission sources of $PM_{2.5}$. The CMB model requires detailed chemical component data of the source particles in advance, which are called the source profiles. Meanwhile, the PMF model does not require these data because it statistically analyzes the environment measurement data without the source profiles. However, a suitable factor number (the number of sources) and a large environmental measurement dataset are necessary for a PMF analysis to obtain a stable and reliable result. When estimating the source contribution of $PM_{2.5}$ at multiple sites, the PMF model is preferable to the CMB model because the source profiles for each receptor site are required prior to a CMB analysis. Moreover, the uncertainties in the profiles need to be considered because most $PM_{2.5}$ sources are unknown and the generation process is complicated. In fact, the study of SA using the PMF model is frequently reported, as discussed by Lin et al. (2010a,b) and Taiwo et al. (2014).

The Community Multiscale Air Quality model (CMAQ) (Byun and Schere, 2006) and the Comprehensive Air quality Model with extensions (CAMx) (ENVIRON, 2013) are types of CTMs that are used for air quality simulations to understand the contributions of pollutant sources and to assess the efficacy of target emission control strategies to reduce regional air pollution levels. CTMs, unlike the PMF model, can follow the chemical and physical processes of each pollutant, such as transport, transformation, and the loss of chemical species, and can estimate the secondary formation of $PM_{2.5}$. In CTMs, the brute-force method (BFM) and a reactive tracers method (or a tagged species method) are often used for SA. According to Burr and Zhang, 2011 the BFM is the simplest SA method and has the ability to simulate indirect effects (i.e., the reduction of one PM species or a PM precursor affecting another via aerosol thermodynamic partitioning processes, gas phase oxidation, and aqueous phase neutralization) and oxidant-limiting effects (i.e., the formation of secondary PM species limited by the availability of oxidants) but not the ability to simulate non-linear effects. The reactive tracers method has strengths when handling non-linearity but limitations when handling indirect effects and oxidant-limiting effects. The estimation of source impacts with CTMs is highly uncertain regardless of the SA method because both the meteorological input data and the emission source data contain errors. In addition, there are uncertainties in the chemical and physical processes adopted in CTMs.

There is no way to measure the source impact directly at the receptor sites. This issue makes it difficult to evaluate the accuracy of an SA method. In theory, all results for SA should be consistent regardless of the method if the SA approach is conducted appropriately. Therefore, in the last several years, combined approaches using CTMs and RMs have been applied in order to minimize errors with single SA methods (Balachandran et al., 2012; Maier et al., 2013; Hu et al., 2014; Ivey et al, 2015). At the same time, the weaknesses and strengths of each approach have been discussed by comparing SA results in Italy from RMs and CTMs (Bove et al., 2014; Pirovano et al., 2015). However, there are few reports of evaluations to specify the aspects of the uncertainties in emission sources via direct comparisons between the SA results of CTMs and RMs throughout the year. Particularly in East Asia, as far as we know, there are no published studies focusing on a comparison of these different SA methods.

This study focuses on a comparison approach with antithetical SA methods (i.e., multivariate receptor modeling based on observed data at multiple-sites and numerical modeling based on designed emissions, theoretical physics, and chemistry) to identify uncertain emission sources. First, the PMF model was applied to the assignment of the $PM_{2.5}$ common sources in the 11 site datasets in Japan as a receptor-oriented SA. Second, CMAQ with BFM (CMAQ/BFM) was used to perform a source-oriented SA of $PM_{2.5}$ for the four major source categories at the 11 sites in the same period as the PMF method. Finally, we compared each SA result and introduced the comparison approach to identify which sectors of emission data have large biases via the ambient atmospheric observation data using PMF and CMAQ/BFM.

2. Materials and methods

2.1. Study region and measurement data

Our study region is the Kinki–Tokai district located in the central part of Japan on the coast of the Japan Sea and the Pacific Ocean. It contains a number of significant air pollution sources including heavy road traffic from three megalopolises (Osaka, Kobe, and Nagoya), ship traffic around three major ports (Osaka, Kobe, and Nagoya), industrial regions, other heavy road traffic areas, several villages, and agricultural areas. The locations of the 11 analyzed sites are shown in Table 1 and Fig. 1b.

The CMAQ performance in the outer Kinki–Tokai district was evaluated with the following observed data: hourly $PM_{2.5}$ concentrations by the U.S. Department of State Air Quality at Beijing, Shenyang, Shanghai, and Guangzhou (http://www.stateair.net/web/post/1/1.html, accessed August 20, 2016) and hourly $PM_{2.5}$ mass and SO_4^{2-} concentrations of $PM_{2.5}$ at Oki in Japan using a Continuous Dichotomous Aerosol Chemical Speciation Analyzer (ACSA by KIMOTO ELECTRIC Co. Ltd.) (http://www.nies.go.jp/pmdep/ctype/, accessed December 11, 2016).

In the Kinki-Tokai district, measurement data of chemical components of $PM_{2.5}$ in 2013 (Ministry of Environment in Japan (MOE), http://www.env.go.jp/air/osen/pm/monitoring/data/h25.html (accessed August 11, 2016) is used for the PMF model analysis and evaluation of the CMAQ performance as the atmospheric monitoring data at the 11 sites. The chemical components data were measured by MOE or local governments at air pollution monitoring sites, such as ambient air pollution monitoring stations and roadside air pollution monitoring stations, according to the standard measuring method of $PM_{2.5}$ based on the Air Pollution Control Law (MOE, 2011). $PM_{2.5}$ samples for a 24-h period were collected with polytetrafluoroethylene (PTFE) and quartz filters at every site for chemical analyses. Laboratory analyses quantified the total $PM_{2.5}$ mass, the metallic elements by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) the organic and elemental