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Source contributions and regional transport of primary particulate matter in China

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ABSTRACT

A source-oriented CMAQ was applied to determine source sector/region contributions to primary particulate matter (PPM) in China. Four months were simulated with emissions grouped to eight regions and six sectors. Predicted elemental carbon (EC), primary organic carbon (POC), and PPM concentrations and source contributions agree with measurements and have significant spatiotemporal variations. Residential is a major contributor to spring/winter EC (50–80%), POC (60%–90%), and PPM (30–70%). For summer/fall, industrial contributes 30–50% for EC/POC and 40–60% for PPM. Transportation is more important for EC (20–30%) than POC/PPM (<5%). Open burning is important in summer/fall of Guangzhou and Chongqing. Dust contributes to 1/3–1/2 in spring/fall of Beijing, Xi'an and Chongqing. Based on sector–region combination, local residential/transportation and residential/industrial from Heibei are major contributors to spring PPM in Beijing. In summer/fall, local industrial is the largest. In winter, residential/industrial from local and Hebei account for >90% in Beijing.

1. Introduction

Airborne particulate matter (PM) has adverse effects on visibility, climate and ecosystems in addition to human health (Cao et al., 2012; Menon et al., 2008; Poschl, 2005; Pui et al., 2014). Fine particle with aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$), is especially harmful because it can penetrate deep into lungs and bloodstream. $PM_{2.5}$ has various components, which can be categorized as primary PM (PPM) because they are directly emitted and secondary PM because they are secondarily formed through chemical and physical atmospheric processes. The chemical composition of $PM_{2.5}$ is complex and is typically composed of elemental carbon (EC), primary organic carbon (POC), metals and trace elements, sulfate, nitrate, ammonium, and secondary organic aerosols (SOA). The physical and chemical formation, transformation and fate of particles are different due to their sources, morphologies, chemical compositions, and mechanical/optical properties (Kleeman and Cass, 2001; Zhang et al., 2014b, 2008).

In the past few decades, developing countries such as China and India have been facing extreme PM pollution problems due to the combination of fast increase of population, industrialization, urbanization and associated energy consumption and lagging of sufficient emission control measures (Chan and Yao, 2008; Hu et al., 2014a; Wang et al., 2014b). However, air pollution control has significant economic cost associated with it due to reduction of economic activities, migration and upgrade of industries, and modernization of energy infrastructures, etc. Thus, it is important to prioritize the emission control targets and determine the cost-effective emission reductions.

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Contributions of different source sectors and regions are the critical information needed for policy makers to design effective emission control strategies. Receptor-oriented techniques have been applied at different regions of China to obtain such information (Tao et al., 2014; Zhang et al., 2013). These receptor-oriented statistical source apportionment methods are highly observation dependent and in need of a large amount of observations with detailed components information, which is not available for most regions where only PM mass concentration measurements are available. State-of-art measurement equipment such as aerosol mass spectrometry (AMS) and aerosol chemical speciation monitor (ACSM) greatly increased the abundance of observation samples in China (He et al., 2011; Sun et al., 2014). Such techniques based on more detailed organic aerosol measurements lead to improved understanding of the sources of PM but they can only resolve contributions to OA and are also limited to only provide local source attribution information at the observation site.

Chemical transport model (CTM) based techniques have also been applied to investigate sources to $PM_{2.5}$ (Kleeman et al., 2007; Ying et al., 2008; Zhang et al., 2014b), but only a few studies have been conducted in China. For example, CMAQ with the brute force method (BFM) was used by Wang et al. (2013) to identify the contributions of both source regions and sectors to $PM_{2.5}$ in Southern Hebei during the 2013 severe haze episode. Decoupled direct method (DDM) as an imbedded sensitivity tool in some CTMs has also been used to determine the importance of different sources (Cohan et al., 2005; Dunker et al., 2002). However, both the BFM and the DDM method are more suitable to estimate the change of PM concentrations due to proposed emission control measures than to determine the contributions of certain sources because removal of PM emissions could affect the transport, chemistry, deposition and interactions with meteorology although they are not chemically reactive (Zhang and Ying, 2011). In addition, the BFM method needs to repeat CTM simulations multiple times and greatly increases the computational cost.

Since primary and secondary $PM_{2.5}$ are formed through different formation pathways and have different chemical composition and regional transport characteristics, it is essential to design source apportionment studies to focus on different PM components separately. Contributions of different source sectors and regions to sulfate and nitrate concentrations in January and August 2009 in China were quantified using a reactive tracer based source apportionment technique (Ying et al., 2014; Zhang et al., 2012). These two studies showed the importance of secondary inorganic components in total $PM_{2.5}$ concentrations. Power sector, transportation, and industrial activities were the dominating source of nitrate and sulfate in both January and August while residential sector contributed to approximately 10–20% of nitrate and sulfate in January. Significant inter-regional transport of nitrate and sulfate was also predicted. However, source and source region contributions to PPM have not been studied in these studies. In addition, the emission inventory used in these studies is for year 2006 and the results cannot be used to interpolate extremely high $PM_{2.5}$ concentrations in recent years. Wang et al. (2014a) quantified the source contributions to both primary and secondary inorganic PM in Xi'an, a major metropolitan in Northwest China and showed that majority of $PM_{2.5}$ was from energy generation (5%), industries (58%) and residential activities (16%) during an extremely polluted month in winter 2013. This study only focused on a small region and a specific month, source contributions to primary and total $PM_{2.5}$ in other areas and the seasonal variations are still not clear.

In this study, a source-oriented air quality model is developed to quantify the contributions of different source sectors and regions to PPM mass as well as its major components EC and POC. The model was applied in a four-month study during 2012–2013 in China to compare the seasonal variations in the region- and source-contributions to PPM. The model results provide valuable information for policy makers to design feasible control measures that balance air pollution reduction and economic costs.

2. Method

2.1. Model description

An updated source-oriented CMAQ model for PPM (CMAQ-PPM) based on CMAQ v5.0.1 (SAPRC-07 photochemical mechanism and AERO6 aerosol module) was developed to determine the source sector and source region contributions to PPM. In the CMAQ-PPM model, tagged non-reactive PM tracers that bear the source sector and region information are used to determine the regional distribution of PPM and its chemical components from multiple emission source sectors and regions in a single model simulation. For example, a tracer ATCR1_2J is used to represent the accumulation mode PPM from source type 1 and source region 2. In this version of the CMAQ-PPM model, emissions from 8 source regions and 9 source types can be tracked simultaneously. These PM tracers are treated as inert PM chemical components and go through the same advection, diffusion, coagulation and deposition processes as other PM components, such as the trace metals in AERO6. As the tracers are considered as non-reactive, no changes to the gas phase chemical mechanism and aerosol chemistry are necessary. In each grid cell, the emission rates of the tracers are set to be a very small fraction (1×10^{-5}) of the PPM emission rates from the corresponding source sectors and regions to make sure that they will not significantly change the particle mass and size distribution to affect other physical and chemical processes. For example, if a grid cell resides in source region 2 and the PPM emission rate of source type 1 in that grid cell is 1 g s^{-1}, the emission rate of the non-reactive PM tracer ATCR1_2J will be set to 1×10^{-5} g s^{-1}. The predicted tracer concentration in a given grid cell, after scaling up by 1×10^5, represents total primary $PM_{2.5}$ mass concentration from a specific source type/region combination in that grid cell. The concentrations of the inert chemical components in primary $PM_{2.5}$ can be determined during the post-processing stage using source specific emission profiles, as shown in equation (1):

$$C_{i,j} = A_{i,j} T_i \quad (1)$$

where $C_{i,j}$ represents the concentration of the jth chemical component from the ith particle emission source category. A is the source profile matrix so that $A_{i,j}$ represents the mass of the jth chemical species per unit mass of PM emitted from the ith emission source. T_i is the model predicted particle mass concentration for the ith source based on the source specific tracer concentrations, as described above. Fig. S1 shows maximum less than 10% difference between the predicted POC and EC in October 2012 by the original CMAQ and CMAQ-PPM models, confirming that the model results were not significantly affected by this technique. Although 72 tracers (9 source regions × 8 source types) are added to the model, the computational time is only slightly increased by less than 10% of the original CMAQ model.

Unlike previous versions of the source-oriented PPM source apportionment models using similar tagging techniques that disabled chemistry and secondary PM formation to reduce run time (Hu et al., 2014b; Wang et al., 2014a), the CMAQ-PPM used in this study includes a full description of the gas and aqueous phase chemistry and gas-to-particle partitioning processes. This is necessary for areas with very high PM concentrations and a significant fraction of secondary PM, as neglecting secondary PM leads to significant biases in the predicted primary particle size and mass concentrations.