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Source contributions to $PM_{2.5}$ in Guangdong province, China by numerical modeling: Results and implications

Xiaohong Yin [a], Zhijiong Huang [a], Junyu Zheng [a,*], Zibing Yuan [a], Wenbo Zhu [a], Xiaobo Huang [a], Duohong Chen [b]

[a] School of Environment and Energy, South China University of Technology, Guangzhou, 510006, China
[b] Guangdong Environmental Monitoring Center, Modiesha Street, Haizhu District, Guangzhou 510308, China

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ABSTRACT

As one of the most populous and developed provinces in China, Guangdong province (GD) has been experiencing regional haze problems. Identification of source contributions to ambient $PM_{2.5}$ level is essential for developing effective control strategies. In this study, using the most up-to-date emission inventory and validated numerical model, source contributions to ambient $PM_{2.5}$ from eight emission source sectors (agriculture, biogenic, dust, industry, power plant, residential, mobile and others) in GD in 2012 were quantified. Results showed that mobile sources are the dominant contributors to the ambient $PM_{2.5}$ (24.0%) in the Pearl River Delta (PRD) region, the central and most developed area of GD, while industry sources are the major contributors (21.5% ~ 23.6%) to those in the Northeastern GD (NE-GD) region and the Southwestern GD (SW-GD) region. Although many industries have been encouraged to move from the central GD to peripheral areas such as NE-GD and SW-GD, their emissions still have an important impact on the $PM_{2.5}$ level in the PRD. In addition, agriculture sources are responsible for 17.5% to ambient $PM_{2.5}$ in GD, indicating the importance of regulations on agricultural activities, which has been largely ignored in the current air quality management. Super-regional contributions were also quantified and their contributions to the ambient $PM_{2.5}$ in GD are significant with notable seasonal differences. But they might be overestimated and further studies are needed to better quantify the transport impacts.

1. Introduction

As one of the most populous and developed provinces in China, Guangdong province (GD) has been experiencing regional haze problems (Zheng et al., 2010; Huang et al., 2012; Lai et al., 2016). Thanks to the continuous emissions control efforts implemented in the past few years, the number of haze days and ambient $PM_{2.5}$ concentrations were in declining trends (Fu et al., 2016; MEPC, 2016), but $PM_{2.5}$ concentrations in some cities were still high and frequently violated the new air quality standard (GDEMC, EPDHK, EPBMC, MGBMC, 2015). Previous reduction efforts mainly targeted on power plants, mobile sources and industrial sources by the way of command-control. Continuous reductions on these sources are still effective for further control on ambient $PM_{2.5,}$ but the cost might increase as well. Other sources, such as NH_3 emissions and cooking emissions, also pose an impact on $PM_{2.5}$ formations. However, they did not receive much attention in previous control strategies. To further reduce ambient $PM_{2.5}$ concentrations, control strategies on more comprehensive sources should be considered, thus a full understanding of the relationship between ambient $PM_{2.5}$ and emission sources is needed.

In general, such evidence-based control strategies are based on source apportionment that quantifies source impacts on ambient pollution and prioritizes emission control plans. Two source apportionment methods are commonly used, including receptor modeling methods and source-based numerical models. Receptor models, e.g. Positive Matrix Factorization (PMF) (Paatero and Tapper, 1994) and Chemical Mass Balance (CMB) (Watson et al., 1984), quantify the relationship between receptors and sources based on measurements. However, these methods cannot distinguish regional or local contributions that play important roles in developing $PM_{2.5}$ control strategies (Yuan et al., 2006). In addition, receptor models use a fixed profile for secondary sources, which might not be in line with the actual case (Habre et al., 2011). The spatial coverage of receptor models is also limited to the locations where samples are collected. On the contrary, numerical models, that simulate dispersion, formation, transport and fate of atmospheric pollutants, have a larger spatial coverage and the ability to quantify transport impacts. Thus, they have been gradually recognized as useful tools for air quality planning in term of $PM_{2.5}$ controls. For instance, Wang et al. (2014b) studied $PM_{2.5}$ control policies in Shanghai, China by using the Particulate Source Apportionment Technology (PSAT) embedded in the Comprehensive Air Quality Model with Extensions (CAMx).

* Corresponding author.
E-mail address: zhengjunyu_work@hotmail.com (J. Zheng).

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Previous source apportionment studies of ambient $PM_{2.5}$ based on air quality models mainly focused on the Pearl River Delta (PRD) region (Guo et al., 2011; Wu et al., 2013), one of the most densely urbanized areas in GD, other than the whole GD. Examining observations in the GD, ambient PM concentrations in the PRD and those outside the PRD showed different trends. In the PRD, trends of ambient PM concentrations in recent years gradually declined, while those outside the PRD remain unchanged or even slightly increased (GDEP, 2014). This might be attributed to two reasons. One is the implementation of the policies of "vacate the cage and change birds" (in Chinese *Teng Long Huan Niao*) initiated by the Guangdong provincial government in 2008 (Li&Fung, 2008; Yang, 2012). Many PRD firms specializing into the so-called low-tech industries are entitled as "birds" that have been relocated outside the PRD. Fig. S1 in the supporting information (SI) shows the spatial map of government-designated industrial relocation in Guangdong province. Another is that previous control strategies mainly focused on sources in the PRD region, while those outside the PRD region received less attention. The economic restructuring and the increasing trend of ambient $PM_{2.5}$ concentrations outside the PRD raise several interesting questions: Where do ambient $PM_{2.5}$ concentrations outside the PRD come from? How much do emissions outside the PRD region affect ambient $PM_{2.5}$ concentrations in the PRD region? How much do super-regional sources contribute to $PM_{2.5}$ in GD and PRD? Understanding the interaction of emissions within and outside the PRD region is beneficial to formulate effective control strategies, and underline the need of a comprehensive source apportionment study across the entire GD region.

In this study, our goals are to quantify seasonal source contributions of ambient $PM_{2.5}$ in different areas of GD in 2012 by using the PSAT module embedded in CAMx (ENVIRON, 2011), coupled with a 2012-based highly-resolved emission inventory for the entire GD. Interactions between PRD and the outside PRD area were revealed as well. We also discussed the regional transport and implications for $PM_{2.5}$ control strategies in GD.

2. Methodology and data source

2.1. Modeling system

CAMx (v5.4), a photochemical dispersion model based on the framework of "one atmosphere", in combination with the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) and the Sparse Matrix Operator Kernel Emissions processor for the PRD region (SMOKE-PRD, derived from SMOKEv3.5) (Wang et al., 2011b), was used for this $PM_{2.5}$ source apportionment study in GD.

The Operational Global Analysis data was selected from National Centers for Environmental Prediction (NCEP, http://rda.ucar.edu/datasets/ds083.2/). In term of physical options, Rapid Radioactive Transfer Model (RRTM), Dudhia scheme, WRF Single-Moment 6-classes scheme and Kain-Fritsch scheme were set up as longwave radiation scheme, short wave radiation, microphysics and cumulus parameterization, respectively. The CB05 and RADM were used as the gas-phase chemistry and aqueous-phase chemistry mechanism in CAMx, respectively. The inorganic aerosol thermodynamics/partitioning (ISORROPIA) and secondary organic aerosol formation/partitioning (SOAP) were selected as aerosol modules in CAMx. The WRF had 26 vertical layers and CAMx employed 18 of them. The January, April, August and December in 2012 were selected to represent the typical seasonal patterns of winter, spring, summer, and autumn. In each period, a spin up period of 3 d was used to minimize the influence of initial conditions.

The model system has two nested domains based on a Lambert-Conformal projection, with resolutions of 27 km × 27 km and 9 km × 9 km respectively, as shown in Fig. S2 of the SI. The coarse domain (D1) covers most parts of East Asia, Southeast Asia and parts of the Pacific Ocean. The domain covers major meteorological fields affecting GD and provides a reasonable boundary condition for simulation. The fine domain (D2) is the target domain in this study, which encompasses the entire GD region.

2.2. Emission inventory

2.2.1. Anthropogenic emission inventory

A high spatiotemporal resolution air pollutant emission inventory in GD was adopted. The spatial distributions are shown in Fig. S4 of the SI. Compiled with the latest domestic emission factors and local activity data, this inventory includes nine types of pollutants (SO_2, NO_X, CO, PM_{10}, $PM_{2.5}$, BC, OC, VOCs and NH_3) and covers power plant, residential combustion, non-road mobile, industrial process, storage, biomass combustion, industrial combustion, on-road mobile, dust, solvent use, agriculture and other anthropogenic sources. To filling the data gap in the area outside GD, the Multi-resolution Emission Inventory for China (MEIC) (Liu et al., 2015) and the Regional Emission inventory in Asia (REAS) (Ohara et al., 2007) were used for the area outside GD in China and the area outside China in D1, respectively.

2.2.2. Biogenic emission inventory

In this study, biogenic VOCs emissions and sea salt were classified together as biogenic sources. Biogenic VOCs emissions were estimated using the Model of Emission of Gases and Aerosols from Nature (MEGAN v2.04) driven by meteorological fields, plant functional type, leaf area index (LAI) and emission factors of VOCs species (Guenther et al., 2012). The plant functional type input files for MEGAN are based on the Multi-source Integrated Chinese Land (MICL), with the resolution of 1 km (Ran et al., 2012). For the LAI input, global gridded LAI data of with the resolution of 30 s from National Center for Atmospheric Research (NCAR, http://cdp.ucar.edu/) was applied. Biogenic VOCs emission factors were provided by MEGAN official website (http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm). Hourly gridded sea salt emissions were estimated by a sea salt emission program (seasalt v3.1, http://www.camx.com).

2.3. Emission processing

The SMOKE-PRD emission processing model was used to generate emission input files for CAMx (Wang et al., 2011b). Spatial, temporal and chemical profiles in SMOKE-PRD were updated to cover the entire GD using the most up-to-date information (Zheng et al., 2013; Huang et al., 2015). The MEIC and REAS emission inventories were processed in SMOKE-PRD as well. To rescale and combine these two emission inventories (with spatial resolution of 0.25° and temporal resolution of 1-month) into the model system (with spatial resolution of 9 km and temporal resolution of 1-h), national population data, road network data, landuse distribution data (as detailed in Table S1 of the SI) and local temporal profiles in SMOKE-PRD were adopted (Zheng et al., 2009; Wang et al., 2011b; Huang et al., 2015).

2.4. Observation data

$PM_{2.5}$ and PM_{10} observations at twelve monitoring sites in the PRD Regional Air Quality Monitoring Network (RAQMN) were employed to evaluate the performance of $PM_{2.5}$ and PM_{10} simulations (Zhong et al., 2013), as shown in Fig. S12 of SI. All measurements of pollutant concentrations in RAQMN stations were subject to strict QA/QC procedures. Additionally, 24-h averaged concentrations of NH_4^+, SO_4^{2-}, NO_3^-, OC and EC measured at four sites in the PRD region were employed to evaluate the simulation of $PM_{2.5}$ compositions (Wang et al., 2015). The Mean Fractional Bias (MFB), Mean Fractional Errors (MFE), Normalized Mean Bias (NMB), Normalized Mean Error (NME) and Mean Bias (MB) were used as model performance statistical metrics in this study, as recommended by U.S. EPA (U.S.EPA, 2007).