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Source-receptor relationships for $PM_{2.5}$ during typical pollution episodes in the Pearl River Delta city cluster, China

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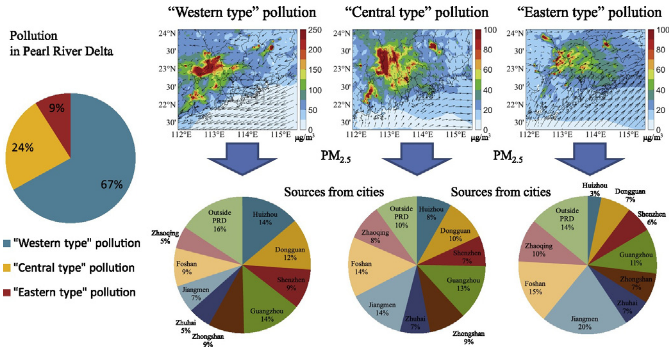
HIGHLIGHTS

- Three types of pollution events were classified in the Pearl River Delta.
- Regional characteristics of $PM_{2.5}$ pollution in the city cluster were analyzed.
- Source-receptor relationships for $PM_{2.5}$ among each city were identified.

GRAPHICAL ABSTRACT



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ABSTRACT

Located in the Southern China monsoon region, pollution days in Pearl River Delta (PRD) were classified into "Western type", "Central type" or "Eastern type", with a relative percentage of 67%, 24% and 9%, respectively. Using this classification system, three typical pollution events were selected for numerical simulations using the WRF-Chem model. The source sensitivity method for anthropogenic emissions of $PM_{2.5}$ and its precursors was applied to identify the source-receptor relationships for $PM_{2.5}$ among 9 cities in PRD. For "Western type" case, the PRD region was under control of a high-pressure system with easterly prevailing winds. The $PM_{2.5}$ concentrations in the western PRD region were higher than those in the eastern region, with emissions from cities in the eastern PRD region having higher contributions. Within the PRD's urban cluster, $PM_{2.5}$ in Huizhou, Dongguan and Shenzhen was mainly derived from local emissions, whereas the $PM_{2.5}$ in the other cities was primarily derived from external transport. For "Eastern type" case, the PRD was influenced by Typhoon Soulik with westerly prevailing winds. Emissions from cities in the western PRD region had the highest impacts on the overall $PM_{2.5}$ concentration. $PM_{2.5}$ in Jiangmen and Foshan was primarily derived from local emissions. Regarding "Central type" case, the PRD region was under control of a uniform pressure field with low wind speed. $PM_{2.5}$ concentrations of each city were primarily caused by local emissions. Overall, wind flows played a significant role in the transport and spatial distribution of $PM_{2.5}$ across the PRD region. Ideally, local governments would be wise to

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establish joint prevention and control measures to reduce regional atmospheric pollution, especially for "Western type" pollution.

1. Introduction

Particulate matter with aerodynamic diameters less than 2.5 μm ($PM_{2.5}$) is not only hazardous to human health but also impacts visibility and contributes to climate change (Gurjar et al., 2010; Feng et al., 2016; Huang et al., 2012; Pipal and Satsangi, 2015; Ying et al., 2004; Yu et al., 2006). In recent years, anthropogenic emissions have increased and the atmosphere has progressively worsened with the rapid development of the Chinese economy and accelerated urbanization (Yang et al., 2011; Saikawa et al., 2011; Wang et al., 2014). Although the Chinese Government has placed great emphasis on regional air pollution and scholars have made major strides in research, China continues to face serious atmospheric environmental problems (Chan and Yao, 2008; Fang et al., 2009; Xu et al., 2011).

Research has shown that China's air pollution problem has evolved from singular pollution events to complex regional ones (Chen et al., 2017; Huang et al., 2015; Zhang et al., 2008). Due to atmospheric transport, one city plays a role not only as the source region of pollutants, but also the receipt region of pollutants. Quantifying the contributions from various emission sources to the total $PM_{2.5}$ concentration of a particular region is a crucial step to develop effective control strategies. The source sensitivity and source apportionment methods in the air quality models are two different source-oriented methods used to determine source-receptor relationships for $PM_{2.5}$ (Burr and Zhang, 2011a). Each method has its own strengths and weaknesses. Whereas source sensitivity can provide useful information about primary and secondary particle species, source apportionment neglects indirect effects by only linking each particle species with its direct primary emission precursor (Burr and Zhang, 2011b). Some widely used numerical models include CAMx (ENVIRON International Corporation, 2016), CMAQ (Byun and Schere, 2006) and WRF-Chem (Grell et al., 2005). Xue et al. (2014) used the Particulate Matter Source Apportionment Technology (PSAT) method of the CAMx framework to quantitatively simulate cross-regional transport patterns of $PM_{2.5}$ and its chemical constituents in China. The results showed that external contributions to the annual $PM_{2.5}$ concentration from regions outside Beijing-Tianjin-Hebei, the Yangtze River Delta, the Pearl River Delta (PRD) and Chengdu-Chongqing urban clusters were 22%, 37%, 28% and 14%, respectively.

Located in the Southern China monsoon region, the Pearl River Delta is one of China's fastest growing urban clusters, consisting of 9 cities including Guangzhou, Shenzhen, Foshan, Zhuhai, Dongguan, Zhongshan, Huizhou, Jiangmen and Zhaoqing. For this reason, the PRD region generates a complex formation mechanism for $PM_{2.5}$ pollution. Indeed, the interconnected city clusters appear to modulate regional pollution characteristics in the PRD (Wu et al., 2005; Wang et al., 2006), and haze events resulting from a high concentration of particles are now occurring frequently (Fung et al., 2005; Kwok et al., 2010; Lai et al., 2016).

Many studies have focused on the regional transport of $PM_{2.5}$ in the PRD region. For example, Wu et al. (2013) applied CAMx with the source apportionment method to analyze how different emission activities influence the $PM_{2.5}$ concentration of the region. The results show that both the local and regional contributions to the $PM_{2.5}$ concentration in 9 different cities in the PRD region were close to each other in April and December, suggesting that controlling emission source by local governments may lead to a significant reduction in $PM_{2.5}$ concentrations. For further reduction, cooperation between multiple regions should also be considered. Wang et al. (2016) used Nested Air Quality Prediction Modeling System (NAQPMS) with the source apportionment method to quantify the intercity transport of $PM_{2.5}$ during a heavy haze episode in the PRD region in January 2013. During the haze episode, the $PM_{2.5}$ levels in Guangzhou and Foshan were mainly derived from local emissions, accounting for 64.9% and 58.9%, respectively. These cities were also identified to be major sources of regional transport. Additionally, $PM_{2.5}$ levels in Zhongshan and Zhuhai were generated primarily from the surrounding areas (accounting for 51.9% and 66.2%, respectively), and these cities were identified to be major receivers of regional transport. The previous studies focused on either the monthly or heavy pollution episodes. None of them focused on different types of pollution events based on the statistical results by long-term observational data. Moreover, none of them used the source sensitivity method.

Combining statistical analysis and numerical sensitivity experiments on typical pollution cases, this paper seeks to identify the major characteristics of regional $PM_{2.5}$ transport between 9 cities in the PRD region. Research on source-receptor relationships in such events can help to develop the most effective and timely control strategies. To this end, Regional Air Quality Index (RAQI) data during 2006–2013 was used to classify the spatial distribution of pollution in the PRD region. The WRF-Chem model was then used to simulate different types of pollution events. The source sensitivity method was applied to reveal the source-receptor relationships for $PM_{2.5}$ among each city. Additionally, sensitivity experiments were performed, which involved reducing anthropogenic emissions of $PM_{2.5}$ and its precursors of each city. Finally, based on a comparison of the simulation results from the base experiment and the sensitivity experiments, this paper quantified the contribution rates of anthropogenic emissions from each of the 9 cities to $PM_{2.5}$ concentrations in the entire PRD region. This paper is organized as follows: Section 2 introduces the methods, Section 3 contains the results and discussion, and Section 4 summarizes the conclusions.

2. Methods

2.1. Classification of air pollution distribution

As shown in Fig. 1, the urban cluster in the PRD region consists of 9 cities including Guangzhou, Shenzhen, Foshan, Zhuhai, Dongguan, Zhongshan, Huizhou, Jiangmen, and Zhaoqing. The Environmental Protection Bureau of Guangdong Province and the Environmental Protection Department of the Hong Kong Special Administrative Region developed the PRD Regional Air Quality Index (RAQI). The RAQI data used in this study were downloaded from the Hong Kong University of Science and Technology's network resource platform. The RAQI uses five grades to identify air quality of the PRD region, which is represented by five different colors in Fig. 1. We note that higher grades correspond to worse air quality. Pollution days were assigned if the RAQI value was greater than grade 3. Taking the Pearl River Estuary as a reference, the location of pollution over the PRD region can be discerned from the spatial distribution of daily RAQI values. Based on these findings, pollution days in the PRD can be classified into three categories: "Western type" pollution, "Eastern type" pollution, and "Central type" pollution. The spatial distributions of the RAQI assigned to these categories are shown in Fig. 1a, b and c, respectively.

The RAQI data during 2006–2013 were analyzed, showing that there were 1875 pollution days in the PRD over this period. The proportions of pollution days and clear days were 64% and 36%, respectively, which is worrisome. Among the pollution days, "Western type" pollution had the highest proportion of 67%, followed by "Central type" pollution of 24%; "Eastern type" pollution comprised only 9% of the total. Overall, air quality of the western PRD region was worse than that in the eastern region. Fig. 2 shows the monthly trend of the cumulative number of atmospheric pollution days in the PRD region during 2006–2013. There