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Characteristics and source distribution of air pollution in winter in Qingdao, eastern China☆

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ABSTRACT

To characterize air pollution and determine its source distribution in Qingdao, Shandong Province, we analyzed hourly national air quality monitoring network data of normal pollutants at nine sites from 1 November 2015 to 31 January 2016. The average hourly concentrations of particulate matter <2.5 μm ($PM_{2.5}$) and <10 μm (PM_{10}), SO_2, NO_2, 8-h O_3, and CO in Qingdao were 83, 129, 39, 41, and 41 μg m^{-3}, and 1.243 mg m^{-3}, respectively. During the polluted period, 19–26 December 2015, 29 December 2015 to 4 January 2016, and 14–17 January 2016, the mean 24-h $PM_{2.5}$ concentration was 168 μg m^{-3} with maximum of 311 μg m^{-3}. $PM_{2.5}$ was the main pollutant to contribute to the pollution during the above time. Heavier pollution and higher contributions of secondary formation to $PM_{2.5}$ concentration were observed in December and January. Pollution pathways and source distribution were investigated using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model and potential source contribution function (PSCF) and concentration weighted trajectory (CWT) analyses. A cluster from the west, originating in Shanxi, southern Hebei, and west Shandong Provinces, accounted for 44.1% of the total air masses, had a mean $PM_{2.5}$ concentration of 134.9 μg m^{-3} and 73.9% trajectories polluted. This area contributed the most to $PM_{2.5}$ and PM_{10} levels, >160 and 300 μg m^{-3}, respectively. In addition, primary crustal aerosols from desert of Inner Mongolia, and coarse and fine marine aerosols from the Yellow Sea contributed to ambient PM. The ambient pollutant concentrations in Qingdao in winter could be attributed to local primary emissions (e.g., coal combustion, vehicular, domestic and industrial emissions), secondary formation, and long distance transmission of emissions.

1. Introduction

Owing to rapid industrialization and urbanization in recent decades, China experiences severe and persistent haze or smog pollution, which mainly comprises high levels of fine particulate matter ($PM_{2.5}$) and ozone (O_3). Reduced visibility is frequently reported. This has attracted attention from the government, the general public, and researchers. In 2014, the Chinese government enacted the strict Atmospheric Pollution Prevention and Control Action Plan (air "national ten items") (Chinese State Council, 2013). Overall, air quality has improved greatly, but urban and regional air pollution remains severe. During haze events, daily average $PM_{2.5}$ concentrations have reached 345 μg m^{-3} (Huang et al., 2014). In 2015, 78.4% cities did not meet the National Ambient Air Quality Standards of China (NAAQS) (Chinese Ministry of Environmental Protection, 2016). Measurements from 74 Chinese major cities showed maximum monthly average of $PM_{2.5}$, PM_{10}, sulfur dioxide (SO_2), and nitrogen dioxide (NO_2) concentrations of 214, 306, 138, and 100 μg m^{-3}, respectively, in December 2015 (China National Environmental Monitoring Centre, 2016). In addition, the population-weighted mean $PM_{2.5}$ in Chinese cities was 61 μg m^{-3}, three times higher than the global mean (Zhang and Cao, 2015). These events were accompanied by extremely poor visibility and air quality, as well as a high health risk. To obtain a better understanding of air pollution in the country and to control it, in 2013, a national air quality monitoring network comprised of nearly 950 monitoring stations in 190 cities was established to monitor real-

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time concentrations of routine pollutants, including $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and carbon monoxide (CO). The stations were selected according to the Technical Regulation for Selection of Ambient Air Quality Monitoring Stations (on trial), including the principle, method, and number (Chinese Ministry of Environmental Protection, 2013).

Base on the data from this monitoring network and observations from other research, air pollution in eastern China may be becoming more severe (Wang et al., 2014; Yang and Christakos, 2015; Zhang and Cao, 2015; Sun et al., 2016). In recent decades, eastern China has experienced drastic increases in the number of hazy days in winter, mainly owing to an increase in pollutant emissions, especially in the Shandong Peninsula region (rate of increase >2 day yr^{-1}) (Wang and Chen, 2016; Zhao et al., 2016). Based on moderate-resolution imaging spectroradiometer measurements, eastern China has a high aerosol optical depth (AOD) distribution (Xue et al., 2014; Hodzic et al., 2016; Zheng et al., 2015; Sun et al., 2016). Winter daily $PM_{2.5}$ concentrations exceed 100 $\mu g\ m^{-3}$ over vast regions of eastern China (Xu et al., 2015b). Ozone pollution in eastern China has become a top environmental issue (Kanaya et al., 2013; Wang et al., 2013b; Hayashida et al., 2015; Zhu et al., 2015). Attributed to changes in emissions, the maximum daily 8-h average of ozone underwent a significant increase from 2003 to 2015, with an average rate of increase of 1.13 ± 0.01 ppb $year^{-1}$ (Ma et al., 2016).

Qingdao is a beautiful and large coastal city on the Shandong Peninsula, eastern China, and is one of the most developed regions worldwide. It experiences both a monsoon and a marine climate. Its economy is dominated by industrial activity, with a large gross domestic product (GDP) of >930 billion RMB (Qingdao Municipal Bureau of Statistics, 2016). Qingdao covers ~570 km^2 and has a population of >9 million. Coal is the main energy source, with an annual consumption of 12.83 million tons. In addition, there are >2 million vehicles in this region. As an important coastal city with a high urbanization rate of 69.99% (Wang and Fang, 2016), Qingdao suffers increasingly from atmospheric pollution. From 2003 to 2013, there were a total of 487 haze days, mainly from October to December (Bi et al., 2015). Owing to urban sprawl and deforestation, the proportion of days with medium or heavy pollution (visibility $\leq$ 5 km; AOD > 0.5) ranged from 10.77% to 32.79% from 2000 to 2010 (Sun et al., 2016). In 2015, there were 72 days when air pollutants exceeded the Grade II standards of the NAAQS in urban Qingdao (Qingdao Municipal Environmental Protection Bureau, 2016). Qingdao experiences more severe air pollution than other coastal cities, such as Yantai, Rizhao, Haikou, Ningbo, and Tianjin, especially in winter (Wang et al., 2013a; Xu et al., 2015a). For example, the average daily $PM_{2.5}$ concentration was 193.77 $\mu g\ m^{-3}$ in winter, with a high $PM_{2.5}/PM_{10}$ ratio of 0.71 (Xu et al., 2015a). From Aura ozone monitoring instrument observations, high SO_2 and NO_2 levels have been also observed in Qingdao, representing the highest concentrations in the world (Krotkov et al., 2016). Therefore, effective air pollution control is urgently required in Qingdao. To control air pollution, a comprehensive understanding of the air pollution characteristics must be developed; however, there are few detailed reports on air pollution in Qingdao.

In this study, we first comprehensively displayed the characterization and source distribution of air pollution in winter in Qingdao. Hourly monitoring data for $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO at nine sites in Qingdao from the national air quality monitoring network were analyzed together with meteorological data to obtain a better understanding of the characteristics of the spatio-temporal distribution and sources of air pollutants in Qingdao. In addition, we investigated the pathways and source distribution of the pollutants based on the monitoring data using a backward trajectory model and potential source contribution function (PSCF) and concentration weighted trajectory (CWT) analyses. Finally, we compared pollutant concentrations in Qingdao with those of other major cities in China. Our study will be important and helpful in understanding the characterization of air pollution and identifying the potential sources of ambient pollutants in winter in Qingdao, which will help decision makers to effectively control the air pollution.

2. Data and methods

2.1. Data collection

We selected all the nine sites in Qingdao, Shandong Province, from the national air quality monitoring network to be used in our study. Notably, there are other stations set by the provincial and local government in the city but they were not used here due to their unavailable measured data. The monitoring datasets at nine sites were recorded from the Chinese Ministry of Environmental Protection (http://www.zhb.gov.cn/). Specifically, we examined hourly concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO at nine sites in Qingdao from 1 November 2015 to 31 January 2016 for the analysis. The study period occurred during winter, when central heating is used in Qingdao. Fig. 1 illustrates the location of Qingdao and the monitoring sites. The nine air quality monitoring sites are: Yangkou (YK; 36.24°N, 120.66°E), Licang (LC; 36.18°N, 120.38°E), Shibei (SB; 36.06°N, 120.34°E), Eastern Shinan (SN_E; 36.06°N, 120.41°E), Sifang (SF; 36.10°N, 120.36°E), Western Shinan (SN_W; 36.05°N, 120.29°E), Laoshan (LS; 36.08°N, 120.45°E), Huangdao (HD; 35.96°N, 120.20°E), and Chengyang (CY; 36.31°N, 120.41°E). YK is located on Laoshan Mountain with no nearby emission sources, representing background levels for the region. The other stations are urban sites located near traffic and industrial or other emission sources. Because the nine sites are relatively concentrated in the urban area of Qingdao, our study only represents the Shinan, Shibei, Licang, Laoshan, Chengyang, and Huangdao Districts of Qingdao. The study area is bordered by the Yellow Sea to the south and Laoshan Mountain to the east, which has an altitude of 1132.7 m. Jiaozhou Bay is located in the middle of this region.

In this study, hourly meteorological data from Qingdao (measured at Liuting; 36.27°N, 120.37°E), including wind speed, wind direction, and temperature, were derived from the National Climatic Data Center. Because of the lack of data, we applied the meteorological data from one station to represent the general weather. Despite of no detailed analysis at each site, it also could provide us a general understanding to the relationship between meteorology and air pollution in Qingdao. During the study period, the prevailing winds were from the north, with an average speed of 3.3 m s^{-1}. There were nearly half the time when the wind speed was <2 m/s, possibly corresponding to pollutant peaks and lower visibilities. The average temperature was 3.2 °C and the relative humidity was almost >60%, favorable for the secondary formation of $PM_{2.5}$ (Chen et al., 2014; Qu et al., 2015). During the study period, Qingdao had an average visibility of ~7 km.

In this study, the 8-h, 24-h or daily, and monthly concentrations of all the air pollutants were averaged by the hourly data from the monitoring sites. The 24-h or daily concentrations of each pollutant were calculated only when there were valid data for more than 20 h during that day. The 8-h O_3 concentrations were calculated when there were valid data for at least 6 h for every 8 h. The monthly averages were computed by averaging the hourly data in that month. The mean concentration of each pollutant in Qingdao was averaged by the data at all monitoring sites unless special instructions.