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Q1 Identification of long-range transport pathways and potential 2 sources of PM_{2.5} and PM₁₀ in Beijing from 2014 to 2015

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A B S T R A C T

Trajectory clustering, potential source contribution function (PSCF) and concentration-weighted 19
 trajectory (CWT) methods were applied to investigate the transport pathways and identify 20
 potential sources of PM_{2.5} and PM₁₀ in different seasons from June 2014 to May 2015 in Beijing. 21
 The cluster analyses showed that Beijing was affected by trajectories from the south and 22
 southeast in summer and autumn. In winter and spring, Beijing was not only affected by the 23
 trajectories from the south and southeast, but was also affected by trajectories from the north 24
 and northwest. In addition, the analyses of the pressure profile of backward trajectories showed 25
 that backward trajectories, which have important influence on Beijing, were mainly distributed 26
 above 970 hPa in summer and autumn and below 950 hPa in spring and winter. This indicates 27
 that PM_{2.5} and PM₁₀ were strongly affected by the near surface air masses in summer and autumn 28
 and by high altitude air masses in winter and spring. Results of PSCF and CWT analyses showed 29
 that the largest potential source areas were identified in spring, followed by winter and autumn, 30
 then summer. In addition, potential source regions of PM₁₀ were similar to those of PM_{2.5}. There 31
 were a clear seasonal and spatial variation of the potential source areas of Beijing and the airflow 32
 in the horizontal and vertical directions. Therefore, more effective regional emission reduction 33
 measures in Beijing's surrounding provinces should be implemented to reduce emissions of 34
 regional sources in different seasons. 35

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51 Introduction

52 Particulate matter (PM) has a significant effect on human
 53 health, visibility, direct and indirect radiative forcing, climate
 54 change and ecosystem (Andreae et al., 2008; Cao et al., 2004;
 55 Menon et al., 2002; Rosenfeld, 2000; Streets et al., 2006; Watson,
 56 2002; Yu et al., 2014a; Yu et al., 2004; Yu et al., 2014b; Zhao et al.,

2015). Pope et al. (2002) reported that 10 µg/m³ increases in Q3
 long-term average PM_{2.5} ambient concentrations were associ- 58
 ated with an almost 8% increase in the risk of lung cancer 59
 mortality. Some studies have also revealed that the increase in 60
 the daily number of deaths for all ages for a 10 µg/m³ increase in 61
 daily PM₁₀ concentrations was 0.6% (Katsouyanni et al., 2001; 62
 Krewski et al., 2003). In the most serious case, an increase of 63

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10 $\mu\text{g}/\text{m}^3$ of $\text{PM}_{2.5}$ results in an elevation of 4.60% and 4.48% with a lag of 3 days, values far higher than the average level of 0.69% and 1.32% for respiratory mortality and morbidity, respectively, in Beijing (Li et al., 2013).

With a rapidly developing economy, expanding anthropogenic activity and urbanization, rapid industrial growth, and an increasing number of vehicles, Beijing has suffered from heavy haze pollution in the form of PM_{10} (particulate matter, or aerosol particles, with aerodynamic diameters $\leq 10 \mu\text{m}$) and $\text{PM}_{2.5}$ (diameters $\leq 2.5 \mu\text{m}$) in recent years (Guoan et al., 2005; Ji et al., 2012; Sun, 2012; Sun et al., 2004). The average values of PM_{10} and $\text{PM}_{2.5}$ from 2004 to 2012 were 138.5 ± 92.9 and $72.3 \pm 54.4 \mu\text{g}/\text{m}^3$, respectively, in Beijing. In addition, more than 30% of days in each year exceeded the daily average PM_{10} concentration of the Grade II National Ambient Air Quality Standard (NAAQS, daily limit of $150 \mu\text{g}/\text{m}^3$) set by the Ministry of Environmental Protection of China (Liu et al., 2015b).

The Beijing municipal government has implemented 16 rounds of air pollution control counter measures from 1998 to 2010 and a five-year clean air action plan (2013–2017) was carried out (<http://www.bjepb.gov.cn/>) (Wang et al., 2015b). However, the haze pollution still occurred frequently and the pollution levels remained very high (Gao et al., 2014; Liu et al., 2015a; Sun et al., 2013; Wang et al., 2014; Wang et al., 2013; Xin et al., 2016; Zhao et al., 2011a, 2011b; Zheng et al., 2014). In addition, haze pollution is a regional and complex phenomenon (Hu et al., 2015; Li et al., 2012; Ren et al., 2004; Zheng et al., 2014). Regional and even super-regional pollution joint control included very effective measures of improved air quality (Chen et al., 2015b). For example, Beijing and the neighboring provinces such as Hebei, Tianjin, and Shandong implemented stringent emission control measures to ensure the air quality during the 2014 Asia-Pacific Economic Cooperation (APEC) Economic Leaders' Meetings in Beijing, 3–11 November 2014 (<http://www.bjepb.gov.cn/bjepb/324122/412670/index.html>, in Chinese) (Chen et al., 2015a; Meng et al., 2015). The stringent emission control measures implemented in Beijing and the regional joint control over the surroundings (especially in Hebei) were responsible for the good air quality and so-called "APEC Blue," which suggests that these measures were very effective (Li et al., 2015a; Wang et al., 2015a). However, these stringent emission control measures were present only for a temporary period and a permanent solution remains a tremendous challenge. In order to control air pollution over a sustained period, it is necessary to carry out an in-depth study of the seasonal variations in regional transport and potential source areas in Beijing.

Observation and modeling studies on regional transport and potential source areas have been introduced in previous studies in Beijing (Han et al., 2015; Li et al., 2015b; Wang et al., 2004; Wang et al., 2013; Wehner et al., 2002; Xia et al., 2007; Xu et al., 2008; Zhao et al., 2007; Zhu et al., 2011). However, little research has been conducted in Beijing with high temporal resolution $\text{PM}_{2.5}$ and PM_{10} data. Previous studies have only focused on PM_{10} or $\text{PM}_{2.5}$ with low temporal resolution data (6 or 24 h resolution) (Wang et al., 2015b; Zhu et al., 2011). High temporal resolution data have been shown to contribute to improved resolutions of the source areas in potential source contribution function (PSCF) calculations (Jeong et al., 2011). In addition, some studies only focused on identifying the

movements of air masses in the horizontal direction (Han et al., 2015; Wang et al., 2015b; Zhu et al., 2011). To obtain comprehensive scientific analyses of the characteristics of the backward trajectory of air masses arriving in Beijing, it is essential to further study the distributions of the backward trajectory in the vertical direction.

Particulate matter is a complex media: it is composed of both primary materials emitted in the atmosphere and secondary aerosols formed in the atmosphere from various chemical processes (Wang et al., 2016). It is difficult to get the potential source regions of each particulate matter species. $\text{PM}_{2.5}$ and PM_{10} as the species of air pollution, however, the potential source regions of $\text{PM}_{2.5}$ and PM_{10} were identified by Trajectory Clustering, PSCF method and CWT method, similar previous studies (Wang et al., 2015b, Wang et al., 2006; Wang et al., 2004; Xin et al., 2016; Zhu et al., 2011). This study aims to improve understanding of the detailed transport pathways and potential sources of $\text{PM}_{2.5}$ and PM_{10} in Beijing. We identified the major air mass transport pathways in the horizontal and vertical directions using cluster analyses and the press profile of backward trajectories. And we identified the main source areas of $\text{PM}_{2.5}$ and PM_{10} in Beijing from June 2014 to May 2015, combining hourly $\text{PM}_{2.5}$ and PM_{10} concentrations using the PSCF method and the CWT method.

1. Experimental methods

1.1. Study location and monitoring data

The area of interest in this study is located in Beijing on the northern part of the North China Plain (Fig. 1a). Western of Beijing is Taihang Mountains and the north and northeast is the Yanshan Mountains. Beijing is the capital of China with a population about 21.705 million in 2015, which covers an area of $16,410.54 \text{ km}^2$.

The hourly $\text{PM}_{2.5}$ and PM_{10} mass concentrations for Beijing during the time period from June 1, 2014 to May 31, 2015 were obtained from the Ministry of Environmental Protection of the People's Republic of China (available at <http://datacenter.mep.gov.cn/>). Hourly $\text{PM}_{2.5}$ and PM_{10} concentrations were calculated by averaging concentrations from thirteen sites in Beijing (Fig. 1b). The thirteen sites included nine urban sites located in the city center (Dongsi, Guanyuan, Tiantan, Wanshouxigong, Aotizhongxin, Nongzhanguan, Gucheng, United States Embassy, Haidianwanliu), two suburban sites located in the Northwest (Dingling and Changping) and two suburban sites located in the Northeast (Huairou and Shunyi New Town).

1.2. Trajectory data

In this study, 72-hour back-trajectories arriving at the center of Beijing ($116^{\circ}25'29''\text{E}$, $39^{\circ}54'20''\text{N}$, 100 m above mean sea level) were calculated every hour (00:00–23:00 local time) using National Centers for Environmental Prediction (NCEP) reanalysis data and the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model version 4.9 developed by the National Oceanic and Atmospheric Administration, Air Resources Laboratory (NOAA ARL). Daily meteorological data were obtained from the global data assimilation system (GDAS) provided by NCEP, which

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