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Impact of long-range transport on aerosol properties at a regional background station in Northern China

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ABSTRACT

The impact of long-range transport on aerosol properties at SDZ regional background station in Northern China during 2005–2010, was analyzed using trajectory clustering method with 3-day, 6-hourly backward trajectories determined by using HYSPLIT 4 model. Eleven clusters were determined by using the two-stage cluster method. $PM_{2.5}$ levels, aerosol scattering coefficient (σ_{sp}) and scattering efficiency ($\alpha_{sp\_2.5}$) of $PM_{2.5}$ associated with each cluster were calculated. Based on the levels of $PM_{2.5}$ and σ_{sp}, eleven clusters were classified into a relatively "clean" group (group A) and a "polluted" group (group B). The $PM_{2.5}$ concentration and σ_{sp} of group A were lower than that of group B. Group A was mainly composed of the trajectories from northwest, north and northeast, which originated and passed through the emission areas such as Mongolia and Inner Mongolia. Group B mostly consisted of the air masses from the south and southeast, and the ones from the northwest. It was characterized with short and low trajectories over major anthropogenic emission regions in North China Plain (NCP), northwestern Hebei province and Inner Mongolia. The trajectory pathway of the northwest cluster in group B was lowest and slowest among all clusters from northerly direction, which caused the accumulation of pollutants along this pathway. High PM hours were identified in each cluster for each month, and were found mainly in group B, especially during March to October. Except of the contribution of high $PM_{2.5}$ emissions in NCP, the production of secondary aerosols with the increasing solar radiation and humidity from March to October, and the straw burning that usually occurs in June in NCP are responsible for the high $PM_{2.5}$ as well. The characteristics of $\alpha_{sp\_2.5}$ of each cluster indicated that the northerly clusters were affected by anthropogenic pollutants mixed with dust, but southerly clusters were only influenced by the pollution aerosols. The $\alpha_{sp\_2.5}$ of dust and anthropogenic pollution aerosols had a clear difference and ranged from 0.44 to 1.85, and 3.01 to 5.43, respectively. The higher $\alpha_{sp\_2.5}$ of anthropogenic pollutant occurred mainly in the southerly trajectory pathways and partially in northwest pathways. The primary emissions and secondary formation of $PM_{2.5}$ along these pathways were significant contributors to the aerosol properties of SDZ. However, emissions northwest of SDZ also make significant contributions to the $PM_{2.5}$ mass loading and $\alpha_{sp\_2.5}$, especially in spring and winter.

1. Introduction

In China, the fast economic development has resulted in an increase in energy consumption, air pollution and associated health effects. The large emissions were concentrated in the

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megacity clusters, such as Jing–Jin–Ji (the abbreviation for Beijing, Tianjin, and Hebei provinces in China), the Pearl River delta (PRD) and the Yangtze River delta (YRD) region (Parrish et al., 2009; Chan and Yao, 2008). With the rapid economic development, population expansion and urbanization, the Jing–Jin–Ji region has been experiencing a severe air pollution problem. In January 2013, persistent severe haze occurred in the eastern and central China. This month has been reported as the haziest month for the past 60 years in Beijing. The daily concentrations of $PM_{2.5}$ at some sites in Beijing and Shijiazhuang (the capital city of Hebei province) have been over 500 μg m^{-3} (Wang et al., 2013). Beijing and other seven cities in Hebei province were listed in the top ten polluted cities in this month. The problem of $PM_{2.5}$ in China, especially in Jing–Jin–Ji has raised widespread public concerns in China and worldwide. The high anthropogenic emissions in this region not only cause the fine particulate pollution in cities, but also influence the aerosol properties in a regional scale or even a larger scale, which may change the radiation process with the aerosol direct and indirect effects.

Investigation of the variation of atmospheric composition with the transport pathways in background areas is a common way to understand the influence of long-range transport of pollutants on the atmospheric environment (Schichtel et al., 2006; Salvador et al., 2008, 2010; Das and Jayaraman, 2012; Tang et al., 2014). Studies have been carried out at three regional background stations Shangdianzi, Lin'an, and Longfengshan in China, respectively (Xu et al., 2009). Lin'an and Longfengshan stations are located in YRD region and Northeast China, respectively. Shangdianzi (SDZ) station is one of the regional Global Atmosphere Watch (GAW) stations in North China. The polluted air masses influencing the gases concentrations at the three stations are different. The polluted air masses are mainly from southeast and northeast at Lin'an, southwest at Longfengshan, and southeast and southwest at SDZ, respectively. Meng et al. (2009) analyzed the seasonal and diurnal variations of the trace gaseous pollutants at SDZ, and found that the air masses from the North China Plain and from the major coal mining regions west of SDZ was responsible for the high concentrations of the gaseous pollutants. The pollution emissions sources in the south regions to SDZ also contributed to the high halogenated gases at this station (An et al., 2012). However, these studies mainly focused on the gaseous pollutants. Works about the transport effect on the particulate pollution, which is the biggest concern of environmental management in China, are comparatively sparse at SDZ.

In our previous study, the annual averages of $PM_{2.5}$ were calculated for three years (2005, 2006, and 2007) with values of 45.9, 66.1, and 52.3 μg m^{-3} at SDZ, respectively (Zhao et al., 2009). The values are much higher than the guideline of 10 μg m^{-3} recommended by the World Health Organization (WHO, 2005), and also higher than the annual average limit of the second-class standard for $PM_{2.5}$ (35 μg m^{-3}) in China (GB3095-2012, 2012). With analyzing the variations of $PM_{2.5}$ and aerosol scattering coefficient (σ_{sp}) with surface wind, we also found that the southerly wind carried pollutants from southern regions and caused the increase of $PM_{2.5}$ and σ_{sp} at SDZ (Zhao et al, 2009, 2011). However, we only got a general idea of the effect of pollution transport. In order to improve our understanding, we investigated the main transport pathways of air masses arriving SDZ, and analyzed the impact of long-range transport on the aerosol properties with 6-year measurements of $PM_{2.5}$ concentration and σ_{sp} in this study. The results of this work would provide certain reference significance for the $PM_{2.5}$ control in North China.

2. Experimental

2.1. Site and instruments

The Shangdianzi station (SDZ, 40°39′N, 117°07′E, 293.9 m a.s.l.) is one of the regional Global Atmosphere Watch (GAW) stations in China. This station is situated in the transitional region of North China Plain (NCP) and Yanshan mountain area (Fig. 1). It is about 100 km from urban area and 55 km from Miyun township. Within 30 km of the site, there are only small villages in mountainous areas with sparse population and insignificant anthropogenic emission sources. Therefore, the atmospheric pollution level at SDZ station can represent the background concentration of atmospheric pollutants in the economic developed regions of North China. Due to the valley topography, the prevailing winds at SDZ are from the east–northeast and the west–southwest. Polluted air masses from urban areas in NCP, can therefore be easily transported to SDZ by southwesterly winds, while relatively clean air masses arrive from other wind directions (Lin et al., 2008; Zhao et al., 2009).

$PM_{2.5}$ concentration and σ_{sp} have been measured continuously at SDZ since 2005. The TEOM (R&P model 1400a, Tapered Element Oscillating Microbalance) is used for $PM_{2.5}$ measurement. It is operated at a standard flow-rate of 1 L min^{-1} with a 2.5 μm cyclone inlet. The sample stream is preheated to 50 °C before entering the mass transducer with an inlet humidity control system, and hence semivolatiles such as ammonium nitrate and water are not measured. The filter loading percentage and flow rates of TEOM are checked once a week, and the filter is replaced when the filter loading percentage is greater than 30%.

An integrating nephelometer (Model M9003, EcoTech) was used to measure the σ_{sp} at the wavelength of 525 nm. The scattering integration angle is from 10° to 170°, and PM_{10} size-selective inlet was used. The relative humidity in the cell of the instrument was controlled below 60% by an automatic heating inlet provided by the manufacturer to prevent liquid particles going into the optical cell. This heating inlet could cause evaporation of volatile inorganic species (such as nitrate) and volatile organic matters. A background (zero) check was done automatically at midnight by pumping in particle-free air every day, and a weekly span check was performed manually by the operator using particle-free HFCR 134a gas recommended by the manufacturer.

The raw $PM_{2.5}$ concentration and σ_{sp} data are recorded as 5-min averages, and then calculated to hourly averages after quality control, which are used for further analysis in this paper.

2.2. Backward trajectory modeling

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT, version 4) model developed jointly by National Oceanic and Atmospheric Administration (NOAA) and Australia's