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Research article

Spatial-temporal variations and mineral dust fractions in particulate matter mass concentrations in an urban area of northwestern China



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ABSTRACT

PM10 and PM25 concentration data were collected from five air-quality monitoring sites in Lanzhou from October 2014 to October 2015, revealing the spatial-temporal behavior of local particulate matter (PM). The Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) and the PM2.5-to-PM10 ratio model were used to investigate the primary transport path, potential source areas and contributions of the East Asian sandstorm to PM in Lanzhou. The analysis in three functional areas of the city indicated that the monthly variation in PM_{2.5} displayed a unimodal U pattern (the highest value was during the heating period), whereas that of PM₁₀ displayed a bimodal pattern (the primary peak appeared in the spring, and the secondary peak appeared in the winter). These two patterns originated from different PM sources. The PM2.5 was primarily affected by human activities, and the PM₁₀ was influenced by both natural and anthropogenic activities, but the relative contributions of these activities were associated with spatial-temporal variations. The daily PM₁₀ and PM_{2.5} concentration variations displayed a bimodal pattern in the three functional areas: the peak values appeared at 11:00-13:00 and 22:00-1:00, respectively, and the lowest values appeared at 4:00-6:00 and 16:00-18:00, respectively. On the monthly, seasonal and daily scales, the PM concentrations exhibited similar patterns in the industrial, urban and rural areas, indicating that they were partly controlled by the regional natural environment. Meanwhile, due to anthropogenic factors, considerable PM amounts were discharged into the external environment, leading to maximum and minimum concentrations of PM appearing in the industrial and rural areas, respectively. The HYSPLIT model showed that dust storms from the northwest desert and Gobi regions affected Lanzhou three times in March 2015 and contributed 68% and 40% of the total mass of PM10 and PM_{2.5}, respectively.

1. Introduction

Recently, substantial growth in China's energy consumption and the rapid economical development have led to a series of heavy air pollution episodes, particularly those caused by atmospheric PM (Han et al., 2014). The primary atmospheric pollutants in more than 70% of the national key detection cities were PM_{10} and $PM_{2.5}$ (http://www.zhb. gov.cn/?COLLCC=1140769127&). PM has drawn much attention because atmospheric PM at high concentrations can easily lead to smog and haze, which will not only decrease atmospheric visibility and air quality (Hyslop, 2009; Wang and Fang, 2016) but also significantly increase the incidence of human respiratory and cardiovascular diseases and mortality (Poeschl, 2005; Chan et al., 2016).

To effectively reduce PM pollution, studies have focused on the basic characteristics (Han et al., 2010; Yun et al., 2013) and possible sources (Zheng et al., 2005; Feng et al., 2013) of PM pollution in certain

urban areas of China. Most studies have provided PM mass concentrations variations in various metro-climatic (Xie et al., 2005; Sun et al., 2013; Zhang and Cao, 2015), temporal (Quan et al., 2014; Shi et al., 2014) and spatial conditions (Wang et al., 2013; Li et al., 2013). Additionally, studies have shown that PM can be derived from a wide variety of sources, both anthropogenic and natural origins (Harrison et al., 2012). Particularly in urban areas, atmospheric dust is easily polluted by anthropogenic inputs, which are typically mixed aerosols produced by various human activities (Cheng et al., 2005).

The estimation of mineral dust contributions to PM concentrations is not straightforward (Miller-Schulze et al., 2015). There is an increasing trend in East Asia to analyze mineral dust and anthropogenic aerosols using modeling methods such as the HYSPLIT model and other numerical models (Uno et al., 2009; Sugimoto et al., 2015; Huang et al., 2017). The HYSPLIT model is one of the most widely used methods for studying dust storms (Uno et al., 2009; Yan et al., 2015). In contrast to

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the Moderate-Resolution Imaging Spectroradiometer (MODIS) (Wang et al., 2012; Qu et al., 2015), the Cloud-Aerosol Lidar and the Infrared Pathfinder Satellite Observations (Sugimoto et al., 2013), the PM_{2.5}-to-PM₁₀ ratio model is not just able to distinguish the aerosol type in the PM but also provide the mixing ratio of dust aerosol and anthropogenic aerosols. The method is a simple partitioning based on the assumption of two distinct aerosol types mixing, those having PM_{2.5}-to-PM₁₀ ratios, which provides insights into source characterization of atmospheric particles (Sugimoto et al., 2015; Chen et al., 2017b; Ishak, 2016). Thus, the PM_{2.5}-to-PM₁₀ ratio model is a new method for approximated quantification the contribution of dust storm particles to the total PM in East Asia (Sugimoto et al., 2015).

In China, most research on PM spatial-temporal distributions and PM atmospheric sources has focused primarily on developed areas in eastern China such as the Beijing-Tianjin region (Zhao et al., 2013), the Yangtze River Delta (Hu et al., 2014) and the Pearl River Delta (Ho et al., 2014). Relatively few studies have been performed in inland areas of China. In fact, certain inland areas are frequently attacked by dust storms, particularly in spring, because they are adjacent to sources of the East Asian dust storms. For example, the Lanzhou dust index is more than twice that of certain eastern cities such as Beijing, Tianjin and Shanghai (Feng et al., 2011). As a city with heavy industry dominating its economy, human activity in Lanzhou discharges a high amount of PM, resulting in PM increases in the air. Additionally, its valley topography causes low wind speeds (particularly in winter), which leads to a stagnant PM phenomenon and increases in PM concentrations. Consequently, for a long time, Lanzhou has been one of the cities in China most severely affected by particulate air pollution (Wang et al., 2009). Therefore, it is necessary to develop a better understanding of the spatial-temporal distribution of PM and its possible sources in Lanzhou. We analyzed the distribution and potential sources of PM pollution using PM10 and PM2.5 hourly data recorded at five national monitoring sites in Lanzhou, starting from October 2014 to October 2015. The primary steps were as follows: (1) analyzing the spatial-temporal changes in PM10 and PM2.5 in Lanzhou and its various functional areas, (2) using the HYSPLIT model to determine the primary source of dust storms in Lanzhou, and (3) using numerical simulation to determine the dust aerosol proportion in PM10 and PM25 during the high-incidence period associated with dust storms in Lanzhou. The results of this study will be useful for the government in developing rational measures for effectively alleviating environmental pollution from atmospheric PM.

2. Materials and methods

2.1. Data sources

The hourly $PM_{2.5}$ and PM_{10} concentration data were obtained from the urban air-quality real-time publishing platform of the China National Environmental Monitoring Center (http://113.108.142. 147:20035/emcpublish/), which recorded data at 5 monitoring sites in Lanzhou spanning one year (from October 16, 2014, to October 15, 2015). The five monitoring stations are located in an industrial zone (Site 1), an urban residential district (Sites 2–4) and a rural area (Site 5), and thus provide data for the three typical functional areas (hereafter referred to as the industrial zone, urban area and rural areas) (Fig. S1b).

2.2. Methods

To ensure data validity, we excluded the missing values of $PM_{2.5}$ and PM_{10} concentrations in the raw data. In calculating the daily mean, if the missing monitoring data for any day spans more than 4 h, the data for that day were considered invalid (data from the industrial zone and urban and rural area were missing data for 9, 7 and 7 days, respectively). Based on the real-time concentrations at the five sites, we

calculated the daily average particle concentrations for Lanzhou.

2.2.1. Backward trajectory analysis

We used the HYSPLIT model, which is a joint research development of the U.S.'s National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology (ABOM) and is used in duststorm backward trajectory simulations. This model is the spread of the Eulerian and Lagrangian hybrid modes and captures transfer, diffusion and settling processes. The 3-hourly archive data at 1°latitude-longitude resolution, GDAS1, from the National Centers for Environmental Prediction (NCEP) was used to drive the model (Yan et al., 2015). To determine the effects of dust storms on Lanzhou and their possible transmission paths, and considering the movement of dust aerosols below a height of 1000 m (Liu, 2015), we divided the simulation into three height levels (100 m, 500 m, 1000 m). According to the known average maximum surface wind speed of dust storms described above, it would generally take around one day for air masses to travel more than 800 km from the farthest sources (e.g., the northwest desert and Gobi regions) to Lanzhou. Therefore, Lanzhou (103.82°E, 36.07°N) as the target city, 24 h backward trajectories of the daily air mass before, during and after dust storms were simulated.

2.2.2. PM_{2.5}-to-PM₁₀ ratios

The numerical simulation method involving $PM_{2.5}/PM_{10}$ ratios developed by Sugimoto et al. (2015) was used to partition anthropogenic aerosols (fine mode dominant aerosols) and dust aerosol (coarse mode dominant aerosols) of the East Asian region (Eqs. (1) through (5)).

If f_{10} (c_{10}) denotes the PM₁₀-to-total mass ratios for the fine (coarse) mode dominant aerosols, and $f_{2.5}$ ($c_{2.5}$) denotes the PM_{2.5}-to-total mass ratios for the fine (coarse) mode dominant aerosols, then F and C are the total concentrations of fine and coarse pattern dominant aerosols, respectively (Christakos et al., 2017). Next, PM_{2.5} and PM₁₀ can be defined as

$$PM_{2.5} = f_{2.5} F + c_{2.5}C$$
(1)

$$PM_{10} = f_{10} F + c_{10}C$$
⁽²⁾

$$R = \frac{PM_{2.5}}{PM_{10}} = \frac{f_{2.5}F + c_{2.5}C}{f_{10}F + c_{10}C}$$
(3)

If $f_{2.5}$ (f_{10}) and $c_{2.5}$ (c_{10}) are known, the fractions of coarse mode dominant aerosols in PM₁₀ and PM_{2.5} can be derived as functions of the measured PM_{2.5}-to-PM₁₀ ratio (R), as in Eqs. (4) and (5), respectively:

Coarse type fraction in PM10 =
$$\frac{c_{10}C}{c_{10}C + f_{10}F} = \frac{\left(\frac{f_{2.5}}{f_{10}}\right) - R}{\left(\frac{f_{2.5}}{f_{10}}\right) - \left(\frac{c_{2.5}}{c_{10}}\right)}$$
 (4)

Coarsetype fraction in $PM_{2.5} = \frac{c_{2.5}C}{c_{2.5}C + f_{2.5}F}$

=

$$\frac{\left(\frac{c_2.5}{c_{10}}\right)\left(\left(\frac{f_{2.5}}{f_{10}}\right)-R\right)}{\left(\left(\frac{f_{2.5}}{f_{10}}\right)-\left(\frac{c_{2.5}}{c_{10}}\right)\right)R}$$
(5)

The coarse mode fractions in PM_{10} and $PM_{2.5}$ depend only on $c_{2.5/}$ c_{10} and $f_{2.5}/f_{10}$, and it was unnecessary to know the c_{10} and f_{10} values. In applying the method, $c_{2.5}/c_{10}$ and $f_{2.5}/f_{10}$ were determined empirically for typical dust and anthropogenic aerosol samples (Sugimoto et al., 2015).

R reflects relative changes in response to the anthropogenic aerosol composition (Sun et al., 2003; Shahsavani et al., 2012; Sugimoto et al., 2015). However, the ratio is still significantly below the natural aerosol ratio (Shahsavani et al., 2012). In addition to dust aerosols, sea salt is a common natural coarse mode aerosol (Sun et al., 2003). Due to the inland of lanzhou, it is not reached by sea salt aerosol. Lanzhou is near the source area of Asian dust. Therefore, aerosols in Lanzhou primarily include dust aerosol (coarse mode dominant aerosols) and

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