Pollution Characteristics Revealed by Size Distribution Properties of Aerosol Particles at Urban and Suburban Sites, Northwest China

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ABSTRACT

High temporal resolution (5 min) particle size distribution data (0.5–20 µm) were collected using aerodynamic particle sizer at an urban (Lanzhou) and a suburban (Yuzhong) site at Lanzhou, northwest China from 1st August to 31st October 2010. Variations of particle concentrations and properties of volume size distributions (PVSD) were analyzed and urban pollution characteristics were investigated using PVSDs and chemical analysis. The average particle number, surface area and volume concentrations for size range 0.5–10 µm were 280.54 ± 270.92 cm⁻³, 331.04 ± 316.95 µm² cm⁻³ and 93.01 ± 127.75 µm³ cm⁻³, respectively, at the urban site, which were 2.87, 1.50 and 1.62 times higher than those at the suburban site. Compared with the suburban site, shifts of accumulation mode (0.5–1.0 µm) to a smaller size and the coarse mode (1.0–10 µm) to a larger size of the PVSDs were observed at the urban site, which may be related to elevated fossil fuel burning and municipal construction or fugitive dust, respectively, in urban area. K-means cluster analysis was used to group the PVSD into six clusters representing the effect of different sources and meteorological conditions. PVSDs at the urban site were dominated by clusters affected by local anthropogenic sources and secondary aerosols, which was characterized by bimodal with peaks at accumulation mode and coarse mode, respectively, while those affected by construction works, wind-borne dust, and dust events were dominated by coarse mode. Chemical composition analysis of PM₂.₅ samples collected on days representing different clusters confirmed the assignment of clusters to different sources.

Keywords: Lanzhou; Atmospheric particles; Cluster analysis; Volume concentration; Sources.

INTRODUCTION

Atmospheric aerosols are primary pollutants affecting the air quality of most urban areas in China and have attracted increasingly attention in recent years due to their effects on visibility (Jung and Kim, 2006), human health (Strak et al., 2012) and climate (Paasonen et al., 2013). Except aerosol particle concentrations, shape and compositions, particle size distributions have been found to be an important factor affecting the behaviour of aerosols in the atmosphere (Dusek et al., 2006; See et al., 2006). Recently, the shape of particle size distributions have been used to obtain information on particle formation mechanisms and their type and origin (Tunved et al., 2004; Charron et al., 2008; Salimi et al., 2014; Vu et al., 2015). Particle size distribution characteristics are becoming important for understanding their effects on climate change, human health, pollution origins, as well as reveal air pollution characteristics.

Aerosol particle concentrations and their size distributions are significantly different for varying regions and atmospheric environments (Morawska et al., 1999; Peng et al., 2014) due to varying emission sources and meteorological processes. Since the 1970s, many important results about particle size distribution properties and their influencing factors have been obtained under various environments, e.g., urban (Hussein et al., 2004; Yue et al., 2013), near highway (Buonanno et al., 2009; Wang et al., 2001; Wehner et al., 2002) and background stations (Tunved et al., 2004; Liu et al., 2008; Shen et al., 2011; Croft et al., 2016). In urban areas, particles emitted from motor vehicles are major particulate pollution sources, especially fine particles (Morawska et al., 1998). Harrison et al. (1999) and Buonanno et al. (2009) found that particle number concentration was 7.5 times and more than 3 times higher than the background level near a busy road and at two urban background locations, respectively. Ketzel et al. (2004) suggested that the average
total particle number could differ by a factor of three at rural, near-city, and urban sites. Currently, most studies on size distribution properties of atmospheric particles in China were mainly carried out in economically developed regions such as central and eastern China and coastal cities (Wu et al., 2008; Gao et al., 2009; Yue et al., 2013). Hu et al. (2006) investigated the effects of high temperature, relative humidity and rainfall on particle size distributions in Beijing in summer. Results from their study indicated that particle concentration and their size distributions had obvious daily variation, and hot and humid environment and rainfall had large impact on size distributions.

Previous studies of size distribution properties of atmospheric particles in northwestern China primarily focused on potential dust source regions and their borders (Cheng et al., 2005; Qiu et al., 2009; Xu et al., 2011). As a fast developing semi-arid city in northwest China, Lanzhou experienced extensive municipal construction works and significant increase in vehicle ownership and traffic congestion. Like many cities in northern China, particulate matter is one of the most formidable air quality and health issues in Lanzhou. Some studies about atmospheric particulate matter pollution characteristics have been conducted in Lanzhou, with most of them focusing on particle mass concentrations (Wang et al., 2009; Wang et al., 2010) and very little on particle size distributions (Gao et al., 2011; Zhao et al., 2015). Specifically, most studies were based on observations from a single station. Until now, little work has been done to investigate the differences of particle size distributions between urban and rural/suburban sites in this area, which limits the understanding of the properties of urban particle pollution, their sources and atmospheric transformation processes in the area.

In the present study, high temporal resolution (5 min) particle volume size distributions (PVSD) in the size range 0.5–20 µm were derived from number size distributions synchronously collected at an urban site and a suburban site – 48 km southeast of the urban site in northwest China from 1st August to 31st October, 2010. This is one of the few dataset that particle sizes were concurrently measured at two sites with different source characteristics. The size distribution properties of particles were resolved with the help of cluster analysis and their possible causes were discussed considering the role of meteorological conditions and pollution sources. Chemical compositions of PM$_{2.5}$ (particulate matter with an aerodynamic diameter smaller than 2.5 µm) were analysed to rationalize the results from cluster analysis. This study will provide basis for further studies on the origin and atmospheric transformation processes of aerosol particles and their climate effects in semi-arid regions of China.

**METHODS**

**Site Description**

The urban site (Lanzhou) (36.05°N, 103.86°E) is located at the Chengguan District of urban Lanzhou, which has an average elevation of 1520 m and is surrounded by mountains and hills rising to 500–600 m. The sampling site is on the roof of a 32-m high academic building of the Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences in the east central district of Lanzhou metropolitan area (Fig. 1). The site is located in a primarily residential and commercial area without obvious industrial sources. There are two 6-lane urban roads with heavy traffic (~2000 cars per hour), i.e., Donggang West Road and Tianshui road, at the south and the west of the sampling site, with a distance of about 40 m and 300 m from the sampling site, respectively (Fig. 1). Due to the close proximity of the measurement site to the roads and residential homes, influence from local traffic and household cooking can be expected.

The suburban site (Yuzhong) is located at the Yuzhong County, about 48 km southeast of the urban site. The sampling site is located at one of the reference sites of the international Coordinated Energy and Water Cycle Observations Project (CEOP) (i.e., SACOL (Semi-Arid Climate & Environment Observatory of Lanzhou University)) (35.95°N, 104.14°E). The site is elevated approximately 200 m from the nearby river valley and the surroundings are covered by short grass. There are no significant local or regional pollution sources close to the site (Huang et al., 2008), representing rural/suburban conditions. Fig. 1 shows the locations of urban Lanzhou and Yuzhong County in Gansu province and the soundings of the two sampling sites.

The study area is characterized by continental semi-dry climate with annual precipitation of 327 mm and annual average temperature of 10.3°C. During the study period, the average temperature was 23.3°C (19.3°C) in August, 18.6°C (15.0°C) in September, and 11.4°C (8.5°C) in October, 2010, with three-month average of 17.7°C (14.3°C) at Lanzhou (Yuzhong). The average relative humidity was 55.1% and 62.9% at Lanzhou and Yuzhong, respectively and the corresponding average wind speeds were 1.5 m s$^{-1}$ and 3.4 m s$^{-1}$. Twenty-five days experienced either trace or light precipitation of which three days had thunderstorms. In addition, there were nine foggy and five floating dust days (including two days affected by regional floating dust).

**Instruments**

Two aerodynamic particle sizers (model 3321, TSI, USA) were operating from 1st August to 31st October, 2010 at Lanzhou and Yuzhong. The observation period covers the end of summer and the autumn of the study area, and is a relatively clean period for the study area (Wang et al., 2009). The APS 3321 is a time-of-flight spectrometer which determines the particle size based on the time required by each particle to flight between two laser beams. Aerosol is drawn into the inlet and is immediately split into a sample flow (1 L min$^{-1}$) through the inner nozzle, and a sheath flow (4 L min$^{-1}$) through the outer nozzle. Each detected particle is assigned to one of four events. Event 1 occurs when only a start pulse is triggered due to marginal scattering of a small particle. Event 3 is a coincident event, and Event 4 is an over range event. Only Event 2 is a valid measurement. Particle sizes (0.5–20 µm) were binned into 52 channels with a time resolution of 5 min. During the
experiment, the inner and the outer nozzles of APS were cleaned every two weeks. At the same time, the total flow rate and the sample flow through the inner nozzle and the sheath flow through the outer nozzle were examined with a bubble flow meter and a mass flow meter, respectively. As part of the study, the aerodynamic particle sizer’s time-of-flight response was calibrated using monodisperse aerosols prior to the deployment in the field. At the urban site, a good correlation \( y = 0.28x, R^2 = 0.91 \) between filter sampled PM\(_{2.5}\) (x) and volume concentrations in the size range of 0.5–2.5 \( \mu \)m (y) was obtained. In addition, event data at the two sites were compared to check the performance of the two APSs. At the urban site, event 1, 2, 3, and 4 were 23.4, 75.7, 0.9 and 0% of the total counts, respectively, at the suburban site, the corresponding values were 24.0, 74.8, 1.2 and 0%, indicating the APSs functioned similarly at the two sites. Similar method has been used by Peters (2006) to inspect APS data from different campaigns. Data were further screened based on the percentage of event 2 data and read flag (RF) indicating the state of the laser, total flow, sheath flow, sample concentration, internal temperature, and detector voltage of the APS. Data were removed from further analysis if event 2 accounted for less than 75% of the total data samples.

At the urban site, 24-h gravimetric samples of PM\(_{2.5}\) were collected on Teflon filters using medium volume sampler (model: TH-150, flow rate: 100 L min\(^{-1}\), Tianhong Instruments Co. Ltd., Wuhan, China) every week during the study period and the mass concentrations of water-soluble inorganic ions (Na\(^+\), NH\(_4\)^+, K\(^+\), Mg\(^{2+}\), Ca\(^{2+}\), Cl\(^-\), NO\(_3\)^– and SO\(_4\)^{2–}\) were determined using two ion chromatography (IC) systems at the State Key Laboratory of Cryospheric Sciences, Chinese Academy of Sciences. Detailed description of ion analysis can be found in Xu et al. (2014b). In addition, one-min meteorological data, including air temperature, relative humidity and wind speed and direction, were obtained with an automatic meteorological station collocated with the APS. At the suburban site, meteorological data at 8 m above the ground were obtained from the boundary layer meteorological measurements system of SACOL. Weather observations from a weather station 1.7 km from the urban site were also used.

**Data Analysis Methods**

The hourly average particle size distributions were considered invalid if more than 30% of the data were missing in an hour. A total of 3977 hourly size distribution samples were obtained at the two sites for further analysis.
The particle number size distribution (PNSD) was measured directly by the APS, while the surface area and the volume size distributions were calculated using Eqs. (1) and (2), respectively, assuming spherical particles. In this study, the particle number, surface area and volume concentrations and their size distributions were exported and analysed using TSI software AIM (Aerosol Instrument Manager).

\[ n_s(D_p) = \frac{4}{3}\pi D_p^3 n_{vol}(D_p) \]  
\[ n_v(D_p) = \frac{4}{3}\pi D_p^3 n_{vol}(D_p) \]  

where \( n_{vol}(D_p) \) and \( n_{vol}(D_p) \) are particle number (cm\(^{-3}\)), surface area (µm\(^2\) cm\(^{-3}\)) and volume (µm\(^3\) cm\(^{-3}\)) concentrations in particle size \( D_p \) (µm), respectively.

Cluster analysis was used to reduce the number of hourly PVSDs obtained at the two sites into several groups with similar characteristics. The K-means clustering routine available in MATLAB\(^\circ\) was used. The K-means clustering routine split the existing multidimensional data into predefined number of subgroups (i.e., clusters), which are as different as possible from each other but as homogeneous as possible within themselves, by iteratively minimizing the sum of squared Euclidean distances from each member to its cluster centroid. K-means clustering method has been used in various studies and has been justified as a preferred technique for particle size distribution data analysis (Beddows et al., 2009). To determine the total number of clusters, some statistics (i.e., RSQ (R\(^2\)), PSF (pseudo F) and PST2 (pseudo t\(^2\))) were calculated using the Statistics Analysis System (SAS\(^\circ\)) and their variations with the number of clusters were evaluated. Result indicates that the best total number of clusters is six. Definitions of the above mentioned statistics and the detailed procedure for determining the number of clusters can be found in Zhao et al. (2017).

For analyzing particle concentrations, we divided particles in the size range 0.5–10 µm into 3 size bins (i.e., 0.5–1.0 µm, 1.0–2.5 µm, and 2.5–10 µm). The total particle concentrations refer to particle concentrations within 0.5–10 µm and in different size bins for the urban and the suburban sites. The total particle number, surface area and volume concentrations (0.5–10 µm) were 72.45 ± 52.35 cm\(^{-3}\), 132.58 ± 115.26 µm\(^2\) cm\(^{-3}\) and 35.56 ± 48.42 µm\(^3\) cm\(^{-3}\) at Yuzhong, respectively. Those at Lanzhou were 280.54 ± 270.92 cm\(^{-3}\), 331.04 ± 316.95 µm\(^2\) cm\(^{-3}\) and 93.01 ± 127.75 µm\(^3\) cm\(^{-3}\), respectively, which were 2.87, 1.50 and 1.62 times higher than those at Yuzhong. The number concentration seems much lower than previous studies using the combination of SMPS and APS (e.g., Gao et al., 2011) due to the limitation of APS’s detectable size, but are comparable to Wang et al. (2010), who also used the APS (model 3321, TSI, USA) operated at SACOL and found that the total particle number in 0.5–20 µm were 84.0 cm\(^{-3}\) and 92.0 cm\(^{-3}\) at Yuzhong for summer and autumn of 2008, respectively. The particle number, surface area and volume concentrations in 0.5–1.0 µm were 4.00, 2.53 and 2.18 times higher at the urban site than those at the suburban site, respectively, and accounted for 95.82%, 62.55% and 34.03% of the total particle number, surface area and volume concentrations, while those in 2.5–10 µm were 1.60, 1.96 and 2.11 times higher at the urban site than those at the suburban site, respectively. The above analysis indicate that the most affected size bins by urban anthropogenic emissions are 0.5–1.0 µm for particle number and 2.5–10 µm for volume concentrations.

**General Size Distribution Properties**

Fig. 3 presents the mean particle number, surface area and volume size distributions at Lanzhou and Yuzhong for the study period. Obvious differences exist between the urban and the suburban sites. The PNSD are unimodal with an obvious peak around 0.54–0.58 µm (Lanzhou) and 0.67–0.72 µm (Yuzhong). The average size distributions of surface area are bimodal with a major peak at 0.63–0.67 µm (Lanzhou) and 0.67–0.72 µm (Yuzhong), and a secondary peak at 3.79–4.07 µm (Lanzhou) and 2.46–2.64 µm (Yuzhong). The mean PVSDs are also bimodal with peaks at 0.67–0.72 µm and 4.70–5.05 µm, respectively, at Lanzhou and at 0.72–0.78 µm and 3.52–3.79 µm, respectively, at Yuzhong.

The shift of the accumulation mode (0.5–1.0 µm) to smaller size at the urban site is consistent with more fine particles from elevated anthropogenic emissions, e.g., road traffic, coal combustion, in urban area (Xu et al., 2014a), while the shift of the coarse mode (1.0–10 µm) to larger size at the urban site is consistent with extensive municipal construction works and fugitive dust in urban area. In addition, the volume mode diameter of the coarse mode particles at the urban site (4.70–5.05 µm) is much larger than 3–4 µm reported by Morawska et al. (1999) for urban influenced aerosols, further indicating the extensive coarse particle sources in the arid and semi-arid urban environment.
Fig. 2 Variations of hourly mean particle number, surface area and volume concentrations for different size bins at Lanzhou (urban) and Yuzhong (suburban) from August 1st to October 31st, 2010.
Table 1. Statistics of hourly mean particle concentrations in different size bins during the study $^a$.

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Size range /µm</th>
<th>Mean ± Std</th>
<th>Max.</th>
<th>Min.</th>
<th>Percentage of the total concentration /%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number (cm$^{-3}$)</td>
<td>0.5–1.0</td>
<td>270.55 ± 261.34</td>
<td>2415.02</td>
<td>8.59</td>
<td>95.82</td>
</tr>
<tr>
<td></td>
<td>1.0–2.5</td>
<td>75.0 ± 50.29</td>
<td>284.51</td>
<td>5.20</td>
<td>92.23</td>
</tr>
<tr>
<td></td>
<td>2.5–10</td>
<td>148.1 ± 10.10</td>
<td>97.16</td>
<td>0.10</td>
<td>3.50</td>
</tr>
<tr>
<td></td>
<td>0.5–10</td>
<td>75.0 ± 4.85</td>
<td>59.53</td>
<td>0.12</td>
<td>6.72</td>
</tr>
<tr>
<td></td>
<td>1.0–2.5</td>
<td>148.1 ± 2.71</td>
<td>32.18</td>
<td>0.002</td>
<td>0.68</td>
</tr>
<tr>
<td></td>
<td>2.5–10</td>
<td>0.57 ± 1.12</td>
<td>15.11</td>
<td>0.01</td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>0.5–10</td>
<td>280.54 ± 270.92</td>
<td>2529.07</td>
<td>8.74</td>
<td>100.00</td>
</tr>
<tr>
<td>Surface area (µm$^2$ cm$^{-3}$)</td>
<td>0.5–1.0</td>
<td>197.73 ± 170.54</td>
<td>1114.42</td>
<td>7.35</td>
<td>62.55</td>
</tr>
<tr>
<td></td>
<td>1.0–2.5</td>
<td>59.92 ± 76.97</td>
<td>786.68</td>
<td>0.56</td>
<td>17.25</td>
</tr>
<tr>
<td></td>
<td>2.5–10</td>
<td>73.40 ± 134.07</td>
<td>1672.22</td>
<td>0.05</td>
<td>20.20</td>
</tr>
<tr>
<td></td>
<td>0.5–10</td>
<td>331.04 ± 316.95</td>
<td>2832.59</td>
<td>8.32</td>
<td>100.00</td>
</tr>
<tr>
<td>Volume (µm$^3$ cm$^{-3}$)</td>
<td>0.5–1.0</td>
<td>20.59 ± 18.38</td>
<td>121.70</td>
<td>0.80</td>
<td>34.03</td>
</tr>
<tr>
<td></td>
<td>1.0–2.5</td>
<td>16.41 ± 22.09</td>
<td>230.22</td>
<td>0.14</td>
<td>19.01</td>
</tr>
<tr>
<td></td>
<td>2.5–10</td>
<td>56.00 ± 101.89</td>
<td>1331.28</td>
<td>0.03</td>
<td>46.95</td>
</tr>
<tr>
<td></td>
<td>0.5–10</td>
<td>93.01 ± 127.75</td>
<td>1582.07</td>
<td>1.03</td>
<td>100.00</td>
</tr>
</tbody>
</table>

$^a$Lanzhou and Yuzhong were represented in black and gray, respectively.

which is also manifested by the dominance of coarse mode particles in the mean PVSD at the urban site (Fig. 3).

Properties of PVSDs

In order to investigate the properties of particle size distributions and their affecting factors in Lanzhou, the 3977 hourly mean PVSDs obtained at Lanzhou and Yuzhong were grouped using cluster analysis and six characteristic volume size distributions were obtained. Fig. 4 shows the median of PVSDs for the six clusters. The percentage of samples from each site in a cluster is also given in Fig. 4. The occurrence frequency of each cluster and the corresponding mean size-resolved particle volume concentrations at the two sites are summarized in Table 2. Also given in Table 2 are the mean temperature, wind speed, and relative humidity for each cluster.

The six clusters represent two different volume size patterns: bimodal (Clusters 1, 3, and 6) and mono-modal (Clusters 2, 4 and 5), and three different concentrations levels. The PVSDs of Clusters 1 and 6 are bimodal with a major peak at 0.67–0.72 µm and a secondary peak at 5.05–5.43 µm (Cluster 1) or 3.79–4.07 µm (Cluster 6) (Fig. 4). Both clusters are dominated by fine particles. Particles in 0.5–2.5 µm account for 69.27% and 68.84% of the total volume concentrations of Clusters 1 and 6, respectively, at Lanzhou. Cluster 1 accounted for only 3.50% of the total hourly PVSDs at the urban site (Table 2), and all the samples in Cluster 1 were from the urban site (Fig. 4), which included 6.68% of the total hourly PVSDs at the urban site (Table 3). However, as shown in Fig. 6, 94.96% of the PVSDs in Cluster 1 occurred in October, accounting for 17.74% of the total hourly PVSDs in the month (Fig. 5). Cluster 6 occurred much more frequently than Cluster 1 and contains 17.29% of the total hourly size distributions. Cluster 6 contains PVSDs from both urban and suburban sites. About twenty-four percent and 10.13% of the total hourly samples at Lanzhou and Yuzhong belong to Cluster 6, respectively (Table 3). Most of the PVSDs in Cluster 6 (72.05%) are from the urban site, while that from the suburban site accounting for 27.95% (Fig. 4). Cluster 6 mainly appeared in October (Fig. 6), which accounted for 51.48% and 19.93% of the total hourly samples in October at Lanzhou and Yuzhong, respectively (Fig. 5). The main difference between Clusters 1 and 6 is the amplitude of the PVSD. The particle volume concentrations in different size bins for Cluster 1 are higher than those for Cluster 6 and the total particle volume concentration of Cluster 1 is 1.35 times higher than that of Cluster 6 at the urban site (Table 2). A closer inspection indicates that 66.4% of the PVSDs in Cluster 1 were observed during 26—31 October 2010 when the minimum temperature was around 4°C. Additionally, the mean wind speed of Cluster 1 was the lowest and the relative humidity was the second highest among the six clusters (Table 2), indicating stable meteorological conditions. Meanwhile, weather record at a weather station about 1.7 km east of the urban site revealed decreased visibility down to 4 km during this period. A laser spectrometer DustTrak™ DRX Aerosol Monitor (TSI Inc. Model 8533)
The dN/dlog\(D_p\), dS/dlog\(D_p\) and dV/dlog\(D_p\) at urban Lanzhou (pink) and suburban Yuzhong (blue) for the period of August 1st–October 31st, 2010. The solid lines show the median distribution and the shaded area indicate 25th–75th percentile ranges.

colloqued with the APS revealed a PM\(_{2.5}\) concentration of 184.37 µg m\(^{-3}\) for Cluster 1, which is 5.2 and 2.5 times of the new Grade II ambient air quality standard for the annual (35 µg m\(^{-3}\)) and the daily mean (75 µg m\(^{-3}\)) PM\(_{2.5}\), respectively. Secondary aerosols have been found to be important sources of fine particles during severe pollution in China’s urban areas (Guo et al., 2014). And a recent study by Pan et al. (2016) highlighted the importance of fossil fuel sources of aerosol NH\(_4^+\) during extreme haze episodes in urban Beijing. Although the official winter heating period starts from 1st November in the study area, residential coal burning for heating is expected under such cold weather. Considering the heavy traffic load in the urban area, Cluster 1 was considered to represent urban polluted
Fig. 4. Particle volume size distributions for Clusters 1 to 6 shown separately for urban Lanzhou (black) and suburban Yuzhong (gray). The solid lines show the median distribution and the dashed lines indicate 25th–75th percentile ranges. The percentage of samples from each site in a cluster is also given in the figure.

PVSDs with high loadings of secondary aerosols, which occurs under unfavorable atmospheric conditions, such as weak wind, low temperature and high relative humidity. This is partly manifested by the high concentrations of secondary ions and the increased NH$_4^+$ in the PM$_{2.5}$ sample collected on 28 October 2010 (Table 4). The wind speed and temperature of Cluster 6 are higher than those of Cluster 1. The dominance of urban samples and fine particles in Cluster 6 indicates that those PVSDs are likely affected by local emission sources. At the urban site emissions from traffic, cooking and industry activities cannot be ruled out, while at the suburban site biomass burning is possible. It is noted that the occurrence frequency of Cluster 6 increases when cold season is approached (Fig. 5). This further indicates that PVSDs in Cluster 6 were affected by local emissions which were more likely to be trapped in shallower boundary layer in relatively cold season. Wang et al. (2001) studied the particle size distributions near traffic sources and found that particle mass size distributions were bimodal with peaks around 0.7 µm and in 4–7 µm. These mode diameters are consistent with those of PVSDs in Clusters 1 and 6 in our study. As all the PVSDs in Cluster 1 and most of the PVSDs (72.05%) in Cluster 6 were observed at the urban site, Clusters 1 and 6 may represent urban PVSDs affected by local pollution sources under different meteorological conditions.
Table 2. Occurrence Frequency of Clusters 1 to 6 (C1-6) and their corresponding mean volume concentration and meteorological conditions \(^a\).

<table>
<thead>
<tr>
<th>Item</th>
<th>C1</th>
<th>C2</th>
<th>C3</th>
<th>C4</th>
<th>C5</th>
<th>C6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency of occurrence %</td>
<td>3.50</td>
<td>1.66</td>
<td>61.07</td>
<td>11.45</td>
<td>5.03</td>
<td>17.29</td>
</tr>
<tr>
<td>Mean volume concentration (µm³ cm⁻³)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.5–1.0 µm</td>
<td>71.07</td>
<td>38.01</td>
<td>9.54</td>
<td>10.74</td>
<td>19.26</td>
<td>30.91</td>
</tr>
<tr>
<td>1.0–2.5 µm</td>
<td>14.10</td>
<td>7.43</td>
<td>8.23</td>
<td>8.67</td>
<td>26.71</td>
<td></td>
</tr>
<tr>
<td>2.5–10 µm</td>
<td>47.53</td>
<td>119.74</td>
<td>5.80</td>
<td>12.94</td>
<td>28.83</td>
<td>14.33</td>
</tr>
<tr>
<td>0.5–10 µm</td>
<td>474.28</td>
<td>11.56</td>
<td>61.56</td>
<td>190.38</td>
<td></td>
<td>20.60</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>8.73</td>
<td>21.95</td>
<td>18.72</td>
<td>22.07</td>
<td>20.63</td>
<td>13.77</td>
</tr>
<tr>
<td>Wind Speed (m s⁻¹)</td>
<td>0.96</td>
<td>22.70</td>
<td>14.77</td>
<td>18.31</td>
<td>18.98</td>
<td>11.30</td>
</tr>
<tr>
<td>Relative Humidity (%)</td>
<td>65.24</td>
<td>66.02</td>
<td>52.24</td>
<td>47.35</td>
<td>60.53</td>
<td>58.25</td>
</tr>
<tr>
<td>Characteristics</td>
<td>Fossil fuel + unfavorable atmospheric condition</td>
<td>Regional dust</td>
<td>Back ground</td>
<td>Urban dust</td>
<td>Urban dust</td>
<td>Fossil fuel or biomass</td>
</tr>
</tbody>
</table>

\(^a\)Lanzhou and Yuzhong were represented in black and gray, respectively.

Table 3. Occurrence Frequency of Clusters 1 to 6 (C1-6) at each station.

<table>
<thead>
<tr>
<th>Site</th>
<th>C1</th>
<th>C2</th>
<th>C3</th>
<th>C4</th>
<th>C5</th>
<th>C6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lanzhou</td>
<td>6.68</td>
<td>2.88</td>
<td>39.48</td>
<td>18.78</td>
<td>8.41</td>
<td>23.78</td>
</tr>
<tr>
<td>Yuzhong</td>
<td>0.00</td>
<td>0.32</td>
<td>84.85</td>
<td>3.38</td>
<td>1.32</td>
<td>10.13</td>
</tr>
</tbody>
</table>

Fig. 5. Proportion of samples from each cluster in a month.
Fig. 6 Proportion of samples from each month in a cluster.

Table 4. Mass concentration (µg m⁻³) of water-soluble ions in PM₂.₅ for different clusters.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>SO₄²⁻</td>
<td>16.54</td>
<td>8.84</td>
<td>6.04</td>
<td>8.51</td>
<td>17.17</td>
</tr>
<tr>
<td>NO₃⁻</td>
<td>13.25</td>
<td>3.65</td>
<td>0.99</td>
<td>2.42</td>
<td>14.63</td>
</tr>
<tr>
<td>Cl⁻</td>
<td>2.92</td>
<td>1.46</td>
<td>0.67</td>
<td>1.01</td>
<td>3.96</td>
</tr>
<tr>
<td>Ca²⁺</td>
<td>1.53</td>
<td>4.44</td>
<td>1.57</td>
<td>2.80</td>
<td>2.08</td>
</tr>
<tr>
<td>NH₄⁺</td>
<td>9.40</td>
<td>0.84</td>
<td>1.74</td>
<td>1.49</td>
<td>6.00</td>
</tr>
<tr>
<td>Na⁺</td>
<td>0.90</td>
<td>1.61</td>
<td>0.32</td>
<td>1.10</td>
<td>1.03</td>
</tr>
<tr>
<td>Mg²⁺</td>
<td>0.38</td>
<td>0.54</td>
<td>0.20</td>
<td>0.25</td>
<td>0.21</td>
</tr>
<tr>
<td>K⁺</td>
<td>1.71</td>
<td>1.23</td>
<td>0.46</td>
<td>1.77</td>
<td>1.37</td>
</tr>
<tr>
<td>SO₄²⁻/NO₃⁻</td>
<td>0.80</td>
<td>0.41</td>
<td>0.16</td>
<td>0.28</td>
<td>0.85</td>
</tr>
</tbody>
</table>

a) Date in the bracket indicate the day on which PM₂.₅ samples were collected.

The mean PVSD of Cluster 2 is unimodal with a peak in 4.70–5.05 µm (Fig. 4). The cluster occurred at very low frequency and only accounts for 1.66% of the total hourly samples (Table 2). The total particle (0.5–10 µm) volume concentration of Cluster 2 is much higher than those of other clusters, and coarse mode particles (2.5–10 µm) account for 77.66% and 74.19% of the total volume concentrations at the urban and the suburban sites, respectively. Ninety-one percent of the hourly samples in Cluster 2 are from the urban site (Fig. 4), which accounts for 2.88% of the total hourly samples at the site (Table 3). Cluster 2 was exclusively observed in August (Fig. 6) and accounted for 8.68% and 0.89% of the total hourly samples in August at Lanzhou and Yuzhong, respectively (Fig. 5). The occurrence of Cluster 2 was related to a floating dust event which affected the observation site during 12:00 12th–18:00 13th August 2010 (Zhao et al., 2015). In addition, the mean temperature and wind speed of Cluster 2 are almost the highest of all the clusters, and the relative humidity at Yuzhong is the lowest among all the clusters, indicating that Cluster 2 was more likely to occur in an environment with high temperature, strong wind and dry air. Alfaro et al. (1998) also pointed out the significant impact of wind erosion on particle size distributions in arid region.

The mean PVSDs of Cluster 3 is bimodal with two peaks located at accumulation and coarse modes, respectively, and the volume concentrations of the two modes are comparable. The mode diameters of the accumulation and coarse modes...
are in 0.67–0.72 μm and 3.79–4.07 μm, respectively. Compared with other clusters, Cluster 3 occurred the most frequently during the study period, and accounted for 61.07% of the total hourly samples (Table 2). In addition, 66.13% of the PVSDs in Cluster 3 were from the suburban site (Fig. 4), which accounted for 84.85% of the total hourly samples at the site (Table 3). The mean volume concentration of Cluster 3 is the lowest of all the clusters (Table 2). The corresponding wind speed of Cluster 3 is also higher than that of other clusters with the exception of Cluster 2 (Table 2).

In addition, most PVSDs in Cluster 3 were observed on rainy days. The dominance of suburban samples in Cluster 3 indicates its representation of PVSDs observed under relatively clean conditions. In other words, Cluster 3 could be regarded as the summer and autumn background conditions for the study area. Using the correlation between filter sampled PM2.5 mass concentration and the volume concentrations in the size range of 0.5–2.5 μm, the PM2.5 mass concentration was estimated to be 45.89 μg m\(^{-3}\) for Cluster 3. Compared to the new ambient Grade II air quality standard for annual mean PM2.5 of 35 μg m\(^{-3}\) released in 2012 by Chinese government, the results indicate that the background particle loadings in the study area is very high, implying the great challenge for the local government to work out more effective strategies for the control of particulate pollution.

Clusters 4 and 5 have similar shaped PVSDs. Both of them have a major peak at 4.70–5.42 μm and a weak peak at 0.63–0.67 μm (Cluster 4) or 0.67–0.72 μm (Cluster 5). Nearly eighty-six percent of PVSDs in Cluster 4 and 87.50% of PVSDs in Cluster 5 were from the urban site (Fig. 4).

PVSDs in Clusters 4 and 5 accounted for 18.78% (3.38%) and 8.41% (1.32%) of the total hourly samples at the urban site (suburban site), respectively (Table 3). Clusters 4 and 5 mainly occurred during two floating dust events lasting from 12:00 12th to 18:00 13th August and on 15th September at Yuzhong (see Fig. 2), which were affected by urban dust (fugitive or construction works) under different meteorological conditions.

### CONCLUSIONS

High temporal resolution (5 min) aerosol particle size distributions in the size range 0.5–20 μm from an urban (Lanzhou) and a suburban (Yuzhong) site were obtained for the period of 1st August–31st October, 2010. The total particle number, surface area and volume concentrations (0.5–10 μm) were 280.54 ± 270.92 cm\(^{-3}\), 331.04 ± 316.95 μm\(^2\) cm\(^{-3}\) and 93.01 ± 127.75 μm\(^3\) cm\(^{-3}\) at the urban site, which were 2.87, 1.50 and 1.62 times higher than those at the suburban site, respectively. Shifts of the accumulation mode (0.5–1.0 μm) to a smaller size and the coarse mode (1.0–10 μm) to a larger size of the PVSDs were observed at the urban site, which may be related to more fine particles from elevated anthropogenic emissions, e.g., road traffic, coal burning etc. and more coarse particles (1.0–10 μm) from construction activities and resuspended/fugitive
dust, respectively, in urban area.

K-means cluster analysis of the hourly mean PVSDs at the two sites results in six well separated clusters of PVSDs associated with particle modes and total volume concentrations. The six clusters were attributed to different sources and meteorological conditions based on the cluster-mean PVSD characteristics, their occurrence frequency at different sites, seasonal variations, and the corresponding meteorological conditions. In general, PVSDs affected by local anthropogenic emissions (e.g., traffic emissions, coal burning) and secondary aerosol formation were characterized by bimodal with peaks at accumulation mode (0.67–0.72 µm) and coarse mode (5.05–5.43 µm or 3.79–4.07 µm), respectively, while that affected by construction works, wind-borne dust, dust storms have dominant peaks in coarse mode (4.70–5.05 µm). Chemical analysis of PM$_{2.5}$ filter samples collected during the study period highlight the potential use of PVSDs to provide supporting evidence for particle sources apportionment.

This study analyzed the size distribution characteristics of aerosol particles at an urban and a suburban site in Northwestern China. The possibility of using PVSDs properties to characterize urban pollution was investigated. However, due to the drop of collection efficiency of APS in the submicrometer range, and the lack of SMPS measurement and high temporal resolution chemical composition data, the origin and atmospheric transformation processes of aerosol particles cannot be derived explicitly from our study. A full range particle size distribution and a long-term measurement are needed to reveal the sources and atmospheric processes of aerosol particles in the study area.

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REFERENCES


