Chemical Mass Balance Source Apportionment of Size-Fractionated Particulate Matter in Nanjing, China

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ABSTRACT

In recent years, atmospheric particulate matter has become the primary air pollutant in Nanjing, China. In order to support a scientific basis for particles reduction, it is of great importance to investigate the contribution of different sources to particulate matter. In this paper, the characteristics of chemical composition of observational data for size-fractionated particulate matter at Gulou site and Pukou site in Nanjing from May 2010 to April 2011 were analyzed. Emission samples of particulate matter were collected and size-fractionated particulate chemical compositions were analyzed to obtain the source profiles. A chemical mass balance (CMB) model was applied to apportion the contribution rate of different sources.

Investigations show that the annual average concentrations of PM₃₀ (particles with an aerodynamic diameter of 10 µm or less) at Gulou and Pukou are 135.8 ± 66.4 μg m⁻³ and 132.2 ± 73.0 μg m⁻³, respectively. For PM₂.₅, the concentrations are 55.1 ± 36.3 μg m⁻³ and 64.8 ± 40.2 μg m⁻³, respectively. The dominant chemical constituents are SO₄²⁻, NO₃⁻, Ca, NH₄⁺, Cl⁻, Fe, Al, and K, accounting for 93.02% and 91.56% of all of the measured compositions in PM₃₀ at Gulou and Pukou. These components account for 95.23% and 93.48% of PM₂.₅ at Gulou and Pukou. Seven types of sources, including coal combustion dust, construction dust, smelting dust, soil dust, vehicle exhaust, secondary aerosols and sea salt, are considered. The results of source apportionment show that in coarse particles, construction dust contributes a maximum of 58.9% at Gulou and 47.3% at Pukou. Smelting dust, as the second most important source, contributes 14.1% and 19.7% at these two sites. For fine particulate matter, secondary inorganic aerosols and coal dust contribution are the two largest sources accounting for 58.8% and 39.5% at Pukou. The findings will be useful for the local government to create efficient control strategies to reduce the emission of construction dust and to pay great attention to the prevention of secondary aerosols and coal combustion dust.

Keywords: Size-fractionated particulate matter; PM₂.₅; CMB model; Emission source profiles; Source apportionment.

INTRODUCTION

In the last decade, particulate matter (PM) has become a primary atmospheric pollutant in Nanjing, China. Atmospheric aerosols, especially fine particles (PM₂.₅, i.e., particles with an aerodynamic diameter of 2.5 µm or less), are harmful to human health (HEI, 2002; Lippmann et al., 2003; Neuberger et al., 2004) and have serious effects on visibility (Jung et al., 2009; Cheng et al., 2011) and global climate change (IPCC, 2007). Recent studies have found that PM₂.₅ could not only penetrate into human respiratory and lungs but also influence visibility through the scattering and absorption of solar radiation. Meanwhile, PM plays a major role in the acidification of precipitation and may affect climate change (Khoder and Hassan, 2008). Nanjing, the capital of Jiangsu province, is an important industrial city and economic center of the Yangtze River delta. Aerosol pollution is becoming increasingly severe with urbanization development. The source apportionment of particulate matter is the basis of the control of air particulate matter pollution. Particles mainly consist of many compounds, such as crust elements, nitrate, sulfate, and various organic species. The dominating primary sources of PM are coal combustion, mobile exhaust, biomass burning and construction dust. It is of great significance to identify contributions to the particulate matter less than 10 µm originating from major sources so that policy makers can develop regional emission control strategies and design efficient governance measures.

The chemical mass balance (CMB) model is one of the most widely used receptor models for source apportionment. The CMB model (Friedlander, 1973; Cooper and Watson, 1980; Watson, 1984; Watson et al., 1984; Gordon, 1988;
forms of ions were (NH₄)₂SO₄, NH₄HSO₄, and NH₄NO₃, with mobile exhaust, coal combustion for residential heating, organic aerosol pollution in Beijing was mostly associated with PM₂.5, PM₁₀, and TSP (total suspended particulates) from different types of element, ion and carbon long-term observation data (IMPROVE and STN) with the CMB model for source apportionment in urban and non-urban areas. The largest contribution to secondary inorganic aerosols reached 49%. Deshmukh et al. (2011) studied the mass concentration of PM₂.5 and PM₁₀ and the water-soluble ion mass concentration seasonal variation in Dung City areas in India. Lee (2013) studied the seasonal ion variation and correlation characteristics of size-fractionated particles in the largest industrial city in Korea. Sulfate was the most abundant species in summer, whereas nitrate and ammonium in particles were highest in winter. Choi et al. (2013) used the positive matrix factorization (PMF) model for the source apportionment of PM₂.5 in the coastal area of Korea. The major sources were secondary nitrate, secondary sulfate and motor vehicles. He et al. (2001) studied the characteristics of PM₂.5 from July 1999 to September 2000 in Beijing, and Song et al. (2007) showed the chemical composition and source apportionment of PM₂.5 in Beijing in January and August 2004. Huang et al. (2006) and He et al. (2006) studied the temporal variation and seasonal characteristics of organic compounds in PM₂.5 in Beijing, the capital of China. The organic aerosol pollution in Beijing was mostly associated with mobile exhaust, coal combustion for residential heating, and cooking emissions. Kong et al. (2010) monitored the PM₂.5, PM₁₀, and TSP (total suspended particulates) from June 2007 to February 2008 at a coastal site in Tianjin. The CMB model was applied to identify the source contributions. Coal combustion, marine aerosol, vehicular emission and soil dust explained 5–31%, 1–13%, 13–44% and 3–46% for PM₂.5, PM₁₀ and TSP, respectively. Zhang et al. (2011) measured the PM₁₀ data of 220 meteorological towers in Tianjin and analyzed the concentrations of elements, ions and carbons with a chemical mass balance model, identifying different levels of particulate matter source contributions, with the largest contribution from the secondary sulfate (24.11–30.96%), followed by secondary nitrate (16.19–20.95%).

Zhang et al. (2011) studied the seasonal variations and sources of water-soluble ions in Xi’an PM₂.5, and the main forms of ions were (NH₄)₂SO₄, NH₄HSO₄, and NH₄NO₃, whose concentration was highest in winter due to coal combustion. These previous studies examined the total PM source apportionment, but also a few studies discussed size-fractionated PM source apportionment. Hu et al. (2013) discussed the size distribution of size-fractionated particles and identified four potential sources for PM₂.5 with enrichment factor and principle component analysis methods.

The ultimate aim of this study is using the CMB model to apportion the size-fractionated PM sources at Gulou and Pukou. This study collected daily aerosol samples from May 2010 to April 2011 at Gulou and Pukou in Nanjing by analyzing the mass concentration and chemical composition, establishing local emission source profiles to use the CMB model to apportion Nanjing size-fractionated particle sources and obtain source contributions. This study has great effects on improving the air quality of Nanjing and providing a scientific basis for governing air pollution in Nanjing.

**Materials and Methods**

**Sampling Site**

Nanjing (118°22′E and 119°14′, 31°14” and 32°37”N) is the capital of Jiangsu province, an important industrial city and economic center in the Yangtze River delta. The size-fractionated PM samples that were used in this study were collected from two sites representing urban and suburban districts in Nanjing. Samples for the downtown area of Nanjing were collected on the roof of an apartment building at the Gulou (118.78°E, 32.05°N) campus of Nanjing University, while the suburban sampling site was located on the roof of a teaching building at the Pukou (118.72°E, 32.21°N) campus of Nanjing University of Information Science & Technology. Fig. 1 shows the two sampling sites in Nanjing.

**Sample Collection**

Daily ambient PM measurements were carried out using the Andersen eight-stage cascade impactor (TE-20-800, Tisch, NY, USA), which can classify nine particle size intervals (<0.43, 0.43–0.65, 0.65–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9.0, and 9.0–10 μm), operated at a constant flow rate of 28.3 L min⁻¹. A total of 70 daily (based on a 96-h period) samples for four seasons from May 2010 to April 2011 were collected almost every ten days on 81-mm Teflon filters (700 filters, including blank filters) using the cascade impactor. The Gulou site accounted for 35 samples, while the Pukou site accounted for the others. Teflon filters were used for mass determination and the analysis of inorganic ions, crust elements and metal elements. The Teflon filters were equilibrated in a desiccator for 24 h before being weighed to determine the mass. The mass concentrations were measured using a gravimetric analysis with an electronic microbalance with a sensitivity of 0.01 mg.

As an important industrial city in the Yangtze River delta, Nanjing has many enterprises. The pillar industries in Nanjing are electronics, petrochemical, automobile and steel smelting (Hu et al., 2012). Nanjing is one of the most rapidly growing cities in China. The economy has been developing rapidly with a large amount of urban infrastructure and an increase in car ownership. According to the actual situation of Nanjing, the emission sources of atmospheric particles are coal combustion dust, construction dust, smelting dust, soil dust, motor vehicle exhaust, secondary aerosols (mainly sulfate, nitrate and ammonium) and sea salt. The emission sources were collected as follows:

Coal combustion dust: the collecting site was a coal-fired electric utility in a chemical industrial park near the highway. The dust was collected from coal ash sorting.
equipment. Construction dust: ashes were collected at an urban construction site and near the construction site. Smelting dust: a Nanjing steel plant was chosen as a representative sample to collect smelting dust. Ashes were collected outside the blast furnace plant. Soil dust: ashes were collected from the bare soil under a tree at the Nanjing University Gulou campus or from large bare land in a suburban area. Motor vehicle exhaust: to avoid the interference of other sources, motor vehicle exhaust was collected directly by an Andersen cascade impactor at the tailpipe. These kinds of sources were collected in Nanjing locally with specialized approaches such as sweeping or grab sampling of soil, unpaved road, fugitive dust using trowels and brooms. These samples were sieved, resuspended, and size-segregated onto filters using ambient cascade impactors. This method might alter the morphological properties and particle size distribution of the chemical components, but this modification is slightly comparing with other effects (Chow et al., 1994; Watson and Chow, 2001).

The collected ashes were separated by the Andersen cascade impactor on 81-mm high-purity quartz filters. Filters that had been sampled for emission sources were analyzed as same as the ambient sampling filters. Numerous of elemental compositions and ions were determined to make chemical abundances in source profiles.

**Sample Treatment and Chemical Analysis**

The collected ambient PM samples and emission sources samples were weighed to determine the aerosol mass. Then, the filters were placed onto clean tinfoil and stored at –20°C until analysis. The sample filters were divided into two parts, half for elemental analysis, and half for water-soluble ion analysis.

The elemental analysis of all of the samples was performed using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Optima 5300, Perkin-Elmer SCIEX, USA). The concentrations of Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Ti and Zn were determined. The method detection limits (MDLs) were determined by adding 3 standard deviations of the blank readings to the average blank values.

Half of the filter samples were extracted into 20-mL ultrapure water using an ultrasonic bath and a shaker before being measured. The ultrasonic bath was operated for 30 min at 40°C, after which mechanical agitations were performed for 1 h. An ion chromatograph (850 Professional IC-AnCat-MCS) was used to measure Cl, NO$_3$-, SO$_4^{2-}$, Na$^+$, NH$_4^+$, K$^+$, Ca$^{2+}$ and Mg$^{2+}$ ions in the aqueous extract. The method detection limit (MDL) was calculated from 1 ppm of the lab-prepared calibration standards. The obtained MDL were 0.01, 0.01, 0.02, 0.01, 0.03, 0.02, 0.01 and 0.01 μg m$^{-3}$ for Cl$^-$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, NH$_4^+$, K$^+$, Ca$^{2+}$ and Mg$^{2+}$, respectively.

**CMB Model**

The CMB receptor model consists of a solution to linear equations that express each receptor chemical concentration as a linear sum of products of the source profile abundances and source contributions. The EPA CMB 8.2 model (US EPA, 2004) uses the effective variance weighted least-squares fitting with both source profiles and ambient concentrations as inputs to be applied to estimate the source contributions and
their uncertainties for individual samples. A brief description of mathematical principle about CMB model is shown below.

\[
C_a = \sum_{i=1}^{N} F_i S_{nt} + E_{nt} \tag{1}
\]

In the formula above, \(C_a\) represents ambient concentration of the \(i\)-th chemical species measured at time \(t\). It is equal to the sum of contributions from all the sources \(N\) in theory. \(F_{nt}\) is the fractional abundance (source profile) of the \(i\)-th species in the \(n\)-th source type. \(S_{nt}\) is the mass contribution of \(n\)-th source at time \(t\). \(E_{nt}\) represents the difference between the measured and estimated ambient concentration.

The measured uncertainties (UNC) of all of the samples and blanks were estimated as follows:

\[
UNC = \sqrt{\frac{\text{MDL}^2}{3} + (P \times \text{Conc})^2} \tag{2}
\]

where \(P\) is a concentration-dependent precision, 7.1% for all species without Na 9.8%, Mg 8.5%, or Al 8.6% (Baumann et al., 2008; Kim et al., 2005).

RESULTS AND DISCUSSION

PM Concentrations

A total of 70 daily samples of size-fractionated PM were collected from May 2010 to April 2011 at two sites in Nanjing. The concentrations of PM\(_{10}\), PM\(_{2.1}\), and PM\(_{1.1}\) and the PM\(_{2.1}\)/PM\(_{10}\) ratio in Nanjing are shown in Table 1.

The daily PM\(_{10}\) concentration ranged from 37 to 290 µg m\(^{-3}\) at Gulou and 45 to 375 µg m\(^{-3}\) at Pukou. The annual average PM\(_{10}\) mass concentration was 135.8 ± 66.4 µg m\(^{-3}\) at Gulou and 132.2 ± 73.0 µg m\(^{-3}\) at Pukou, with negligible differences between the two sites. The annual average PM\(_{2.1}\) mass concentration and standard deviation (SD) were 55.1 ± 36.3 µg m\(^{-3}\) and 64.8 ± 40.2 µg m\(^{-3}\) at Gulou and Pukou, respectively. The data show significant differences in that the PM\(_{2.1}\) concentration is greater at Pukou than at Gulou.

The annual average PM\(_{1.1}\) mass concentration and standard deviation (SD) were 39.4 ± 23.0 µg m\(^{-3}\) and 45.5 ± 34.6 µg m\(^{-3}\) at Gulou and Pukou, respectively. The PM\(_{1.1}\) difference between urban and suburban sites was the same as that of

<table>
<thead>
<tr>
<th>Season</th>
<th>Site</th>
<th>Type</th>
<th>N(^a)</th>
<th>Max</th>
<th>Min</th>
<th>Mean</th>
<th>SD(^b)</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spring</td>
<td>GL</td>
<td>PM(_{10})</td>
<td>10</td>
<td>270.4</td>
<td>92.2</td>
<td>153.7</td>
<td>57.4</td>
<td>0.38</td>
</tr>
<tr>
<td></td>
<td>PK</td>
<td>PM(_{10})</td>
<td>9</td>
<td>208.9</td>
<td>74.9</td>
<td>121.9</td>
<td>41.6</td>
<td>0.52</td>
</tr>
<tr>
<td>Summer</td>
<td>GL</td>
<td>PM(_{10})</td>
<td>8</td>
<td>101.5</td>
<td>37.1</td>
<td>65.2</td>
<td>22.2</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td>PK</td>
<td>PM(_{10})</td>
<td>6</td>
<td>91.3</td>
<td>67.0</td>
<td>75.9</td>
<td>9.8</td>
<td>0.49</td>
</tr>
<tr>
<td>Autumn</td>
<td>GL</td>
<td>PM(_{10})</td>
<td>9</td>
<td>261.9</td>
<td>60.9</td>
<td>129.7</td>
<td>64.0</td>
<td>0.39</td>
</tr>
<tr>
<td></td>
<td>PK</td>
<td>PM(_{10})</td>
<td>9</td>
<td>95.8</td>
<td>18.8</td>
<td>50.0</td>
<td>22.3</td>
<td>0.39</td>
</tr>
<tr>
<td>Winter</td>
<td>GL</td>
<td>PM(_{10})</td>
<td>9</td>
<td>77.0</td>
<td>14.5</td>
<td>36.7</td>
<td>17.3</td>
<td>0.39</td>
</tr>
<tr>
<td></td>
<td>PK</td>
<td>PM(_{10})</td>
<td>9</td>
<td>376.0</td>
<td>45.8</td>
<td>155.5</td>
<td>111.0</td>
<td>0.39</td>
</tr>
<tr>
<td>Annual</td>
<td>GL</td>
<td>PM(_{10})</td>
<td>36</td>
<td>290.0</td>
<td>134.0</td>
<td>184.7</td>
<td>54.2</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td>PK</td>
<td>PM(_{10})</td>
<td>33</td>
<td>376.0</td>
<td>45.8</td>
<td>132.2</td>
<td>73.0</td>
<td>0.49</td>
</tr>
</tbody>
</table>

\(^a\) Number of samples.  
\(^b\) Standard deviation.
PM$_{2.1}$. Because there is no 2.5-µm cut-point of the Andersen
cascade impactor sampler, 2.1 µm is defined as the boundary
between fine and coarse particles in this study. Referring to
the latest Ambient Air Quality Standards (GB3095-2012)
annual average of PM$_{2.5}$ in China (35 µg m$^{-3}$), the PM$_{2.1}$ mass
concentrations were overweight at both sites and were much
higher than the annual average of 12 µg m$^{-3}$ for USA National
Ambient Air Quality Standard (NAAQS) for PM$_{2.5}$. But the
PM$_{2.1}$ concentration in Nanjing was lower than that in other
mega cities in China but much higher than that in the USA
and other countries (Table 2).

Similar seasonal variations in the PM concentrations
were observed at two sites. The seasonal variations of size-
fractionated PM were winter > spring > autumn > summer
at both sites except for that of PM$_{10}$ at Pukou, which was
winter > autumn > spring > summer. The maximum mass
concentration in winter was approximately twice higher than
the minimum mass concentration in summer at both sites.
The high concentration in winter was most likely due to the
stable meteorological condition that limited the dispersion
and the reduced amount of surface vegetation that made it
easier for dust to mix with the air. And the increased coal
burning emissions due to space heating also made the PM
concentration higher in winter. The low concentration in
summer was likely because of high-frequency precipitation
and precipitation rate that removed particles through
rainout or washout processes.

The annual average PM$_{2.1}$/PM$_{10}$ ratio was 0.41 and 0.49
at Gulou and Pukou, respectively. The PM$_{2.1}$/PM$_{10}$ seasonal
variation ranged from 0.38 to 0.57. The ratio of PM$_{2.1}$/PM$_{10}$
was lower than that of many other countries, for instance,0.54 in Ulsan, Korea (Lee and Hieu, 2013) and 0.59 in
Navarra, Spain (Aldabe et al., 2011). The annual average
value of PM$_{2.1}$/PM$_{10}$ at Pukou was higher than that at Gulou
possibly due to construction dust. Nanjing has carried out a
large number of urban construction in order to hold the Asian
Youth Games and Youth Olympic Games. So an excessive
number of construction sites in the urban area produced
building dust with particulates of coarse diameter (PM$_{10}$–PM$_{2.1}$),
leading to a lower value of PM$_{2.1}$/PM$_{10}$ at Gulou which is
on behalf of urban area. Meanwhile, a large concentration
of iron, steel smelting, petroleum and chemical enterprises
around the Pukou site may have produced a large amount of
carbon combustion dust, smelting dust and secondary aerosols,
which are mostly fine particulate matter. Therefore, the
value of PM$_{2.1}$/PM$_{10}$ at Pukou is higher.

Fig. 2 shows the size-fractionated particulate concentration
variations at Gulou and Pukou as measured by an Andersen
eight-stage cascade impactor during the study period. The
size distribution of the particulate concentration is different.
The PM at Gulou showed a distinct distribution with a strong
peak of 5.8–10 µm and a weak peak of 0.65–1.1 µm. The PM
at Pukou showed significant double-hump distribution
with strong peaks of 5.8–9.0 µm and 1.1–2.1 µm. This study
shows that the average concentration of coarse particles
(PM$_{10}$–PM$_{2.1}$) is higher at Gulou than at Pukou. The increased
concentration of coarse particles would be caused by the
contribution of soil and construction dust with relatively
dry and windy meteorological conditions.

### Table 2. Mass concentrations (µg m$^{-3}$) of fine particles and the major ions at different sites over the world (The cut-sizes of fine particles are 2.5 µm for other works compared to 2.1 µm in this work).

<table>
<thead>
<tr>
<th>Site</th>
<th>Time</th>
<th>Mass</th>
<th>Cl$^{-}$</th>
<th>NO$_3$$^{-}$</th>
<th>SO$_4$$^{2-}$</th>
<th>Na$^{+}$</th>
<th>NH$_4$$^{+}$</th>
<th>Ca$^{2+}$</th>
<th>K$^{+}$</th>
<th>Mg$^{2+}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>GL, Nanjing, China</td>
<td>2010–2011</td>
<td>55.1</td>
<td>1.89</td>
<td>9.07</td>
<td>12.01</td>
<td>0.35</td>
<td>4.51</td>
<td>0.70</td>
<td>0.14</td>
<td>0.14</td>
<td>This work</td>
</tr>
<tr>
<td>PK, Nanjing, China</td>
<td>2010–2011</td>
<td>64.8</td>
<td>2.07</td>
<td>7.32</td>
<td>10.78</td>
<td>0.45</td>
<td>4.07</td>
<td>0.81</td>
<td>0.15</td>
<td>0.15</td>
<td>This work</td>
</tr>
<tr>
<td>Beijing, China</td>
<td>1999–2000</td>
<td>127</td>
<td>NR</td>
<td>NR</td>
<td>0.13</td>
<td>11.52</td>
<td>14.08</td>
<td>1.70</td>
<td>0.55</td>
<td>0.55</td>
<td>He et al. (2001)</td>
</tr>
<tr>
<td>Shanghai, China</td>
<td>2001–2003</td>
<td>154.3</td>
<td>3.07</td>
<td>3.00</td>
<td>11.23</td>
<td>11.52</td>
<td>14.08</td>
<td>1.25</td>
<td>0.57</td>
<td>0.57</td>
<td>Wang et al. (2006)</td>
</tr>
<tr>
<td>Qingdao, China</td>
<td>1997–2000</td>
<td>43.64</td>
<td>1.53</td>
<td>0.83</td>
<td>4.00</td>
<td>4.33</td>
<td>11.40</td>
<td>1.55</td>
<td>0.30</td>
<td>0.30</td>
<td>He et al. (2002)</td>
</tr>
<tr>
<td>Fuzhou, China</td>
<td>2007–2008</td>
<td>44.33</td>
<td>1.55</td>
<td>0.83</td>
<td>4.40</td>
<td>11.94</td>
<td>14.08</td>
<td>1.48</td>
<td>0.64</td>
<td>0.64</td>
<td>Xu et al. (2010)</td>
</tr>
<tr>
<td>Denver, USA</td>
<td>2002–2003</td>
<td>8.50</td>
<td>NR</td>
<td>NR</td>
<td>3.40</td>
<td>11.94</td>
<td>14.08</td>
<td>1.15</td>
<td>0.55</td>
<td>0.55</td>
<td>Dutton et al. (2013)</td>
</tr>
<tr>
<td>Seoul, Korea</td>
<td>2009–2010</td>
<td>13.89</td>
<td>1.16</td>
<td>3.21</td>
<td>1.63</td>
<td>11.94</td>
<td>14.08</td>
<td>1.15</td>
<td>0.28</td>
<td>0.28</td>
<td>Choi et al. (2013)</td>
</tr>
<tr>
<td>Navarra, Spain</td>
<td>2009–2010</td>
<td>15.38</td>
<td>0.08</td>
<td>1.63</td>
<td>0.08</td>
<td>11.94</td>
<td>14.08</td>
<td>1.15</td>
<td>0.28</td>
<td>0.28</td>
<td>Aldabe et al. (2011)</td>
</tr>
</tbody>
</table>

Not reported.
Concentration of Chemical Species

A total of 17 types of chemical elements and 8 types of water-soluble ions concentrations were detected in this study. The crustal materials, such as [Al, Ca, Fe and Ti], were calculated as follows, intending to account for unmeasured mineral oxides:

\[
\text{Crustal material} = 2.2 \times [\text{Al}] + 1.63 \times [\text{Ca}] + 2.42 \times [\text{Fe}] + 1.94 \times [\text{Ti}]
\]

The average concentration of all of the chemical compositions in PM10 is 67.17 μg m⁻³ at the Gulou site and 63.7 μg m⁻³ at the Pukou site with the exceptions of Na⁺, K⁺, Ca²⁺ and Mg²⁺, accounting for average PM₁₀ concentrations of 49.5% and 43.8% at Gulou and Pukou, respectively. The chemical composition concentration in PM₂.₅ is 35.11 μg m⁻³ and 35.61 μg m⁻³, accounting for an average PM₂.₅ concentration of 63.7% and 49.7% at Gulou and Pukou, respectively. The annual average concentrations of chemical species in PM₂.₅ and PM₁₀ at the two sites are shown in Fig. 3.

In PM₁₀, the concentrations of the major abundant chemical compositions follow the order SO₄²⁻ > NO₃⁻ > Ca²⁺ > NH₄⁺ > Cl⁻ > Fe > Al > K > Mg, while the decreasing sequence in PM₂.₅ is SO₄²⁻ > NO₃⁻ > NH₄⁺ > Cl⁻ > K > Fe > Ca²⁺ > Zn > Na > Al. In PM₁₀, the dominant chemical compositions account for 93.02% and 91.56% at Gulou and Pukou, respectively, for all of the measured compositions; meanwhile, in PM₂.₅, these compositions account for 95.23% and 93.48% at Gulou and Pukou, respectively. The remaining chemical components are present in too low of a concentration to be listed. Comparing the chemical composition concentration of PM₂.₅ to those of PM₁₀, the concentration of calcium significantly decreased, suggesting that calcium makes a large contribution to coarse particles. The concentrations of major ions slightly decreased, indicating that the contribution of water-soluble ions is focused in fine particles.

Water-soluble ions play an important role in the atmosphere. The average concentration of all of the chemical compositions in PM10 was 11.6%, and the ratios of Na⁺ and Mg²⁺ were approximately 30%. Na⁺ and Mg²⁺ were the main components of sea salt aerosols, suggesting that sea salt aerosols mainly contribute to coarse particles. The ratios of SO₄²⁻, NO₃⁻, and NH₄⁺ were approximately 70%, 70%, and 95%, respectively, confirming that the contribution of secondary aerosols is mainly concentrated in fine particles. An analysis of the Pukou site also reached the same results.

Through the correlation analysis of water-soluble ions at the Gulou site, we found that Na⁺ and Cl⁻ at levels 0–4 had a high correlation (R was 0.932, 0.900, 0.847, 0.891, and 0.593, respectively), indicating that these two ions may have the same emission source. Considering that NaCl is the main component of sea salt, sea salt aerosol should be considered an emission source in coarse particles.

At levels 0 and 3–5, K⁺ and SO₄²⁻ had a high correlation (R was 0.765, 0.766, 0.851, and 0.921, respectively). The
high correlation in coarse particles maybe indicating that they are belong to the particles from sea salts. K$^+$ and NH$_4^+$ had a high correlation at levels 2–5 (R was 0.798, 0.816, 0.815, 0.784, respectively), indicating that a small part of the NH$_4^+$ in the coarse particles may come from biomass burning (Tsai et al., 2012).

![Graphs of chemical species concentrations in PM10 and PM2.1](a) PM$_{10}$ (b) PM$_{2.1}$

**Fig. 3.** Annual average concentrations of chemical species in (a) PM$_{10}$ and (b) PM$_{2.1}$ at Gulou (GL) and Pukou (PK).

**Table 3.** Average annual concentrations (µg m$^{-3}$) of size-fractionated PM major chemical compositions at Gulou.

<table>
<thead>
<tr>
<th>Species</th>
<th>0 level</th>
<th>1 level</th>
<th>2 level</th>
<th>3 level</th>
<th>4 level</th>
<th>5 level</th>
<th>6 level</th>
<th>7 level</th>
<th>8 level</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>10–9.0</td>
<td>9.0–5.8</td>
<td>5.8–4.7</td>
<td>4.7–3.3</td>
<td>3.3–2.1</td>
<td>2.1–1.1</td>
<td>1.1–0.65</td>
<td>0.65–0.43</td>
<td>&lt; 0.43</td>
</tr>
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<td>0.2818</td>
<td>0.1901</td>
<td>0.0690</td>
<td>0.0423</td>
<td>0.0453</td>
</tr>
<tr>
<td>Ca</td>
<td>2.6546</td>
<td>1.7762</td>
<td>1.1146</td>
<td>1.0802</td>
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<td>0.3082</td>
<td>0.1332</td>
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<td>Fe</td>
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<td>0.4406</td>
<td>0.3656</td>
<td>0.2613</td>
<td>0.1461</td>
<td>0.0962</td>
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<tr>
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<td>0.1176</td>
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<tr>
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<td>0.0254</td>
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<td>Zn</td>
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<td>0.0628</td>
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<td>0.0884</td>
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<td>0.0341</td>
</tr>
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<td>0.0678</td>
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<tr>
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<td>0.9990</td>
<td>1.4523</td>
<td>1.1446</td>
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<td>0.0380</td>
<td>0.0531</td>
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</tr>
<tr>
<td>Ca$^{2+}$</td>
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<td>0.7716</td>
<td>0.9490</td>
<td>0.5813</td>
<td>0.2676</td>
<td>0.1922</td>
<td>0.1116</td>
<td>0.1288</td>
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<tr>
<td>Mg$^{2+}$</td>
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<td>0.0593</td>
<td>0.0389</td>
<td>0.0706</td>
<td>0.0482</td>
<td>0.0564</td>
<td>0.0453</td>
<td>0.0203</td>
<td>0.0191</td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>0.3280</td>
<td>0.3197</td>
<td>0.2066</td>
<td>0.3330</td>
<td>0.2919</td>
<td>0.5346</td>
<td>0.5570</td>
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<td>0.3808</td>
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<tr>
<td>NO$_3^-$</td>
<td>0.6445</td>
<td>0.5818</td>
<td>0.4231</td>
<td>0.7726</td>
<td>0.8775</td>
<td>2.3945</td>
<td>3.0111</td>
<td>2.2241</td>
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<td>SO$_4^{2-}$</td>
<td>0.7410</td>
<td>0.6790</td>
<td>0.9785</td>
<td>0.8141</td>
<td>0.9045</td>
<td>2.7785</td>
<td>3.6537</td>
<td>2.6357</td>
<td>2.9412</td>
</tr>
</tbody>
</table>
At levels 5–7, there was a high correlation between SO4\(^{2-}\) and NO\(_3^-\) (R was 0.831, 0.751 and 0.701, respectively). Because these compounds are transformed from SO\(_2\) and NO\(_x\), the high correlation suggests that they come from the same air mass and also possibly the same emission sources or the same source regions. NH\(_4^+\) had a good correlation with both SO\(_4^{2-}\) and NO\(_3^-\), as the correlation coefficient R\(^2\) between NH\(_4^+\) and NO\(_3^-\) from levels 5 to 8 were 0.956, 0.888, 0.952 and 0.807, respectively. R\(^2\) was 0.945, 0.865 and 0.857, respectively, from levels 5–7 between NH\(_4^+\) and SO\(_4^{2-}\). The correlation of NH\(_4^+\) and NO\(_3^-\) was better than that of SO\(_4^{2-}\).

Comparisons between the calculated and observed NH\(_4^+\) concentrations were conducted to evaluate the formation of secondary aerosol water-soluble ions were determined. Secondary inorganic aerosols were represented by pure ammonium nitrate (AMNIT; NH\(_4\)NO\(_3\)) and ammonium bisulfate (AMSBUL; NH\(_4\)HSO\(_4\)). Sea salt aerosol was represented by a profile containing exclusively (100%) NaCl.

The main chemical composition of coal combustion dust was Al. The three most common elements in PM\(_{10}\) were Al (9.107%), Fe (2.745%), and Ca (1.572%), and the two most common elements in PM\(_{2.5}\) were Al (8.208%) and Fe (8.050%). The proportion of chemical components of coal combustion dust is small, probably because the main chemical component in coal combustion dust is the residual carbon due to incomplete combustion. Therefore, the OC and EC requires further analysis.

The main chemical composition of smelting dust was Fe. The three most common elements were Fe (26.458%), Ca (5.479%), and Al (3.757%) in PM\(_{2.5}\) and Fe (5.741%), Mg (1.234%) and Al (3.930%) in PM\(_{10}\). The high Fe content was due to the smelting dust that was collected from the Nanjing steel plant.

The main chemical composition of construction dust was Al. The three most common elements were Al (4.162%), Ca (3.647%), and Fe (3.217%) in PM\(_{10}\), which have similar percentages. The three most common components were Al (19.095%), Mg (5.031%), and SO\(_4^{2-}\) (3.920%) in PM\(_{2.5}\). The chemical composition of cement affected the composition of building dust. Cement was mainly composed of CaO, SiO\(_2\), Al\(_2\)O\(_3\), Fe\(_2\)O\(_3\), and SO\(_2\). In addition, a limited amount of calcium sulfate (CaSO\(_4\)) is often added as an additive to Portland-type cement. Without an analysis of the element Si, then Al, Ca and Fe are the main components.

The three elements that accounted for the largest proportion of vehicle exhaust source in PM\(_{10}\) were Al (6.095%), Ca (2.050%) and Mg (1.234%); the proportions in PM\(_{2.5}\) were Al (9.108%), Ca (3.016%) and Mg (1.530%), respectively. The total mass concentration in the chemical composition was very small. Because of carbon dioxide, which was not measured, and water, these were the main components of automobile tailpipe emissions. Other pollutants such as

### Table 4. Average annual concentrations (µg m\(^{-3}\)) of size-fractionated PM major chemical compositions at Pukou.

<table>
<thead>
<tr>
<th>Species</th>
<th>0 level</th>
<th>1 level</th>
<th>2 level</th>
<th>3 level</th>
<th>4 level</th>
<th>5 level</th>
<th>6 level</th>
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<tbody>
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<td>PM size range (µm)</td>
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<td>10–9.0</td>
<td>9.0–5.8</td>
<td>5.8–4.7</td>
<td>4.7–3.3</td>
<td>3.3–2.1</td>
<td>2.1–1.1</td>
<td>1.1–0.65</td>
<td>0.65–0.43</td>
<td>&lt;0.43</td>
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<tr>
<td>Al</td>
<td>0.3429</td>
<td>0.4512</td>
<td>0.1927</td>
<td>0.2792</td>
<td>0.2986</td>
<td>0.1916</td>
<td>0.0668</td>
<td>0.0304</td>
<td>0.0334</td>
</tr>
<tr>
<td>Ca</td>
<td>0.9875</td>
<td>1.3268</td>
<td>0.5319</td>
<td>0.6809</td>
<td>0.6088</td>
<td>0.3141</td>
<td>0.1382</td>
<td>0.0823</td>
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<tr>
<td>Fe</td>
<td>0.5441</td>
<td>0.7010</td>
<td>0.3481</td>
<td>0.4194</td>
<td>0.4610</td>
<td>0.3839</td>
<td>0.2336</td>
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<tr>
<td>K</td>
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<td>0.1352</td>
<td>0.0724</td>
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<td>0.2246</td>
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<td>0.1258</td>
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<td>0.0614</td>
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<td>0.1170</td>
<td>0.1343</td>
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<tr>
<td>Ti</td>
<td>0.0281</td>
<td>0.0314</td>
<td>0.0190</td>
<td>0.0219</td>
<td>0.0220</td>
<td>0.0194</td>
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<tr>
<td>Zn</td>
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<tr>
<td>Na(^+)</td>
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<td>Mg(^{2+})</td>
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<td>Cl(^-)</td>
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Characteristics of the Size-Fractionated PM Source Profiles in Nanjing

Coal combustion dust, smelting dust, building dust, soil dust, vehicle exhaust, secondary aerosols and sea salt have been classified as the emission sources according to the actual situation in Nanjing. The source profiles consist of the measured mass fractions (abundances) of chemical components in size-fractionated PM emissions from different source types. Through a correlation analysis, the compositions of secondary aerosol water-soluble ions were determined. Secondary inorganic aerosols were represented by pure ammonium nitrate (AMNIT; NH\(_4\)NO\(_3\)) and ammonium bisulfate (AMSBUL; NH\(_4\)HSO\(_4\)). Sea salt aerosol was represented by a profile containing exclusively (100%) NaCl.
soot, sulfur dioxide, carbon monoxide, nitrogen oxides and volatile hydrocarbons, most of which were in gaseous form, were not collected.

The main chemical composition of soil dust was Al. The three most common elements were Al (13.179%), Ca (11.970%), and Fe (3.980%) in PM$_{10}$ and Al (15.923%), Ca (6.615%), and Mg (2.846%) in PM$_{2.1}$. The soil dust represents the crust element content. Without the detection of Si, then Al, Ca and Fe are the main components of soil dust.

Size-Fractionated Source Apportionment in Nanjing

Table 5 shows the size-fractionated source apportionment results at the Gulou and Pukou sites. The source contributions at the two sites were slightly different. Construction dust was the largest emission source at coarse particles both at the urban (Gulou) and suburban (Pukou) sites. The construction dust proportion at Pukou (47.26%) was lower than that at Gulou (58.92%) indicating that a large number of construction sites in urban areas made the dust contribution higher at Gulou. The smelting dust was the second largest emission source at PM$_{10}$–2.1 at both sites. However, smelting dust at Pukou (19.67%) contributed higher than that at Gulou (14.08%). This was because that a large number of smelting enterprises in suburban areas made the smelting dust contribution higher at Pukou.

The smelting dust was the second largest emission source at PM$_{10}$–2.1 at both sites. However, smelting dust at Pukou (19.67%) contributed higher than that at Gulou (14.08%). This was because that a large number of smelting enterprises in suburban areas made the smelting dust contribution higher at Pukou. The third largest emission source was soil dust at both sites but at Gulou site the vehicle exhaust (8.09%) was slightly lower than soil dust (8.12%) indicating that vehicle exhaust should be more influential at urban at coarse particles. Moreover, farmland and large bare areas in the suburban area lead to greater soil dust emission at Pukou (8.56%). The sea salt dust also made important contributions to coarse particles. We can preliminarily conclude that sea salt particles are mainly distributed from 9.0 to 2.1 μm. The contributions were roughly the same at two sites. The vehicle exhaust made very different contributions at Gulou (8.09%) and Pukou (4.06%) at coarse particles indicating a much heavier traffic in the urban area.

The main emission sources at PM$_{2.1}$ were secondary inorganic aerosols and coal combustion dust. The source contributions were consistent at the two sites. The secondary aerosols made the largest contribution at two sites. The sulfate and nitrate contributions can account for 58.76% and 44.23% at Gulou and Pukou, respectively, followed by coal combustion, which accounts for 15.87% and 39.46%, respectively. The proportion of secondary inorganic aerosols at Gulou was higher than that at Pukou, especially nitrate aerosols. The nitrate aerosols contributed much higher at Gulou (28.05%) than Pukou (17.39%). Because nitrate aerosol mainly comes from vehicle emissions of NO$_x$, and the proportion of vehicle exhaust at Gulou is higher than that at Pukou, the vehicle exhaust had a greater contribution in urban areas than in suburban areas.

Fig. 4. Comparison of calculated and measured ammonium at levels 5–8.
Table 5. The size-fractionated source apportionment (%) at Gulou (a) and Pukou (b).

(a) Table 5. The size-fractionated source apportionment (%) at Gulou (a) and Pukou (b).

<table>
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<tr>
<th></th>
<th>coal</th>
<th>smelt</th>
<th>const</th>
<th>vehicle</th>
<th>soil</th>
<th>AMNIT</th>
<th>AMSUL</th>
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<td>-</td>
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<td>PM2.1</td>
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<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

*b Sum of 0–4 levels (10–2.1 µm).

(b) Table 5. The size-fractionated source apportionment (%) at Gulou (a) and Pukou (b).

<table>
<thead>
<tr>
<th></th>
<th>coal</th>
<th>smelt</th>
<th>const</th>
<th>vehicle</th>
<th>soil</th>
<th>AMNIT</th>
<th>AMSUL</th>
<th>seasalt</th>
<th>other</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM10-2.1a</td>
<td>2.31</td>
<td>19.67</td>
<td>47.26</td>
<td>4.06</td>
<td>8.56</td>
<td>-</td>
<td>-</td>
<td>5.62</td>
<td>12.52</td>
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<tr>
<td>PM2.1</td>
<td>39.46</td>
<td>3.59</td>
<td>0.25</td>
<td>6.48</td>
<td>0.04</td>
<td>17.39</td>
<td>26.84</td>
<td>-</td>
<td>5.95</td>
</tr>
<tr>
<td>PM1.1</td>
<td>64.08</td>
<td>0.26</td>
<td>0.47</td>
<td>1.11</td>
<td>0.08</td>
<td>11.21</td>
<td>22.79</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

*b Sum of 0–4 levels (10–2.1 µm).

Fig. 5. Pie chart of source contributions at the two sites.

The second largest emission source was coal combustion dust. The proportion of coal combustion dust was higher at Pukou (39.46%) than Gulou (15.87%). It was because of there was a large-scale power plant near Pukou sites and there was no power plant in the urban areas.

The third largest emission source was vehicle exhaust at both sites. The vehicle exhaust contributed higher at Gulou (8.13%) than Pukou (6.48%) indicating a heavier traffic at urban areas. Due to a lack of the component analysis of OC/EC in the results, the vehicle exhaust emission source contribution in this study was on the low side, and a higher nitrate contribution can be seen as the contribution of automobile exhaust after secondary transformation.

The contribution of each source for PM10 is shown in Fig. 5. The total contribution of PM10 emission source at Gulou was 94.96%, and the contributions of the first three sources were building dust (29.62%), sulfate (16.80%) and nitrate (15.35%). The total contribution of the PM10 emission source at Pukou was 90.88%, and the contributions of the first three sources were building dust (22.89%), coal combustion dust (21.57%) and sulfate (15.35%). The total contribution of the PM2.1 emission source at Gulou was 93.13%, and the contributions of the first three sources were sulfate (30.71%), nitrate (28.05%) and coal combustion...
The size-fractionated PM samples that were collected at the Gulou and Pukou sites in Nanjing during the period from May 2010 to April 2011 were analyzed. The mass concentrations, chemical compositions and source apportionment of the size-fractionated PM at the two sites were investigated. The source contributions were identified using the chemical mass balance (CMB) model.

The annual mean PM$_{10}$ concentrations were 135.8 ± 66.4 μg m$^{-3}$ and 132.2 ± 73.0 μg m$^{-3}$ at Gulou and Pukou, respectively. For PM$_{2.5}$, the annual mean concentrations at the two sites were 55.1 ± 36.3 μg m$^{-3}$ and 64.8 ± 40.2 μg m$^{-3}$. The concentration was at the same level as that of other cities in China except for mega cities, such as Beijing and Shanghai, but was much higher than that in the USA, Europe and Korea.

The size-fractionated PM at Gulou was mainly distributed from 10–5.8 μm and 2.1–0.65 μm, occupying 58.4% of the total concentration of PM$_{10}$. At Pukou, the size-fractionated particles were mainly distributed from 2.1–0.65 μm, 9.0–5.8 μm, and below 0.43 μm, which occupied 51.8% of the total concentration of PM$_{10}$. The chemical compositions that constituted a large proportion of PM$_{10}$ were SO$_4^{2-}$, NO$_3^-$, Ca, NH$_4^+$, Cl$^-$, Fe, Al, K, and Mg. The concentrations of SO$_4^{2-}$, NO$_3^-$, and Ca at Gulou were significantly higher than those at Pukou. The chemical compositions that constituted a large proportion of PM$_{2.5}$ were SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, Cl$^-$, K, Fe, Ca, Zn, Na, and Al.

The contributions of sources as conducted by the CMB model showed that at Gulou, the major sources of PM$_{10}$ were construction dust (29.62%), sulfate (30.71%), nitrate (28.05%) and coal combustion dust (15.87%). At Pukou, the three largest sources in PM$_{10}$ were construction dust (22.89%), coal combustion dust (21.57%) and sulfate (30.71%).

The results in this paper will be beneficial for local management to create an efficient control policy to reduce airborne particles. Limiting the emissions of construction dust and coal combustion dust is necessary. The government should attach great importance to the harm of secondary aerosol and strengthen the control of industrial and vehicle exhaust emissions.

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