Nitrate and sulfate in individual Asian dust-storm particles in Beijing, China in spring of 1995 and 1996

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Abstract

Energy dispersive X-ray analysis of elements and reagent thin-film test of particulate nitrate and sulfate were carried out to examine individual dust particles collected in Beijing during five dust-storm events occurring in spring of 1995 and 1996. Dominant particles were electron-opaque and had irregular shapes during the dust-storm periods, and their size was frequently in the range larger than 1 \(\mu\text{m}\) (diameter). Besides, some mineral particles that showed regular cubic shapes were found in the range from 0.1 to 2 \(\mu\text{m}\). Their X-ray spectrums indicated calcium was abundant and little or no other elements with atomic number larger than 11 existed in such particles. They were supposed to be emitted initially from construction sites, and then formed through crystallization in the atmosphere. Their most possible composition was CaO or Ca(OH)\(_2\). It was estimated that 93\% of the collected electron-opaque particles are dust particles and the cubic particles in term of number frequency. On reagent films, few dust particles reacted apparently with barium chloride suggesting there was no water-soluble sulfate on the surface of dust particles although X-ray spectrums of about 14.6\% of dust particles showed peaks of sulfur. The frequency of nitrate-containing particles in dust particles was 10.8\%, which was much smaller than that in mineral particles collected in non-dust-storm periods. These results suggest that almost no sulfate is formed and nitrate is hardly formed on the surface of dust particles during their transport from source areas to Beijing. \copyright 1999 Published by Elsevier Science Ltd. All rights reserved.

Keywords: Dust particles; Nitrate; Water-soluble sulfate; Ca abundant

1. Introduction

Asian dust-storms inject immense quantities of dust particles from arid and semi-arid areas into the atmosphere. These particles can be blown eastward and widely transported to the East China (Liu et al., 1981), Japan (Ishizaka and Ono, 1982; Iwasaka et al., 1983) and even the North Pacific Ocean (Duce et al., 1983; Uematsu et al., 1983). After being injected into the air, dust particles coalesce with pre-existing particles, grow through condensation of water and other gases, and fall out. At the same time, they can work as reaction sites in the atmosphere and act as carriers of reactants and products as well (Iwasaka et al., 1988). Behavior of the soil particles also attracts the interest in radiative effects of atmospheric particles since recently many investigators have suggested that radiative forcing of particulate matter is noticeable in eastern Asia (Zhou et al., 1994; Tegen and Fung, 1995; Parungo, 1997).

Chemical analysis of Asian dust particles collected in Gobi areas in northwestern China reveals that dust particles mainly contain Mg, Al, Si, K, Ca, Fe, Ti, and Mn (KOSA, 1991; Zhang et al., 1993). This feature is consistent with the composition of soil on the surface of the
As aerosol particles were collected on the roof of the Physics Building of Peking University during five dust-storm events in springs of 1995 and 1996. The building is about 15 m high above the ground and located in the northwest part of Beijing (39°58′N, 116°19′E). Near this site were residences, streets and shopping areas. There was no source of atmospheric pollutants and other higher buildings which might disturb the sampling. Therefore, the observation site was favorable to the measurements of pollutants arriving at Beijing through long-range transport from west or northwest. Particles were collected onto transmission electron microscope grids using two single-stage impactors whose jet diameters were 1 (stage-1) and 0.5 mm (stage-2), respectively. The pumping rate of air was 4.5 l min⁻¹ and airflow passed through stage-1 first and then through stage-2. Therefore, 50% cut-off aerodynamic diameters of the impactors were 0.3 and 0.1 μm, respectively, if the density of particles was assumed to be 2.3 g cm⁻³. The grids were covered with carbon-reinforced holly formvar film. The samples on such grids were applied for morphological analysis and EDX analysis. Nitron film was pre-coated on some sampling grids to detect nitrate-containing particles and barium chloride films were pre-coated on the grids with nitron film to detect sulfate-containing particles and to see internal mixing state of particles of sulfate and nitrate. These reagent films were made in a vacuum evaporator under 10⁻¹⁶ Torr and the thickness of nitron and barium chloride was about 8 and 4 nm, respectively. Sulfate and nitrate in individual particles were identified by the appearance of Liesegang rings, the product of sulfate and barium chloride reaction, and needle-like bundles, the product of nitrate and nitron reaction, respectively. According to the laboratory experiments of Isawa and Ono (1979) and Qian et al. (1991), it was suggested that the detection limits of the reagent film for sulfate and nitrate contained in individual particles were 10⁻¹⁷ and 10⁻¹⁴ g, respectively.

Sampling date, the weather during the observation periods and films used for the analysis are summarized in Table 1. We collected atmospheric particles 4 times on the grids of stages-1 and 2 during every dust episode listed in Table 1. The particle collection time was decided to be 30–90 s corresponding to the visibility. Right after every sampling, the grids of nitron film and nitron–barium chloride multiple film were exposed to octanol vapor under room temperature for 18 h to promote possible reactions of nitrate and sulfate with the films. Then all grids were viewed and photographed using the JEM-200CX transmission electron microscope of the Electron Microscope Laboratory of Peking University. After that, elemental composition of individual particles was analyzed by use of the EDX analyzer attached to the same microscope. The counting time for every particle was around 180 seconds. The EDX analyzer used is not effective for the elements with atomic number less than 11 (Na), i.e., the analyzer cannot detect light elements, such as H, C, N and O.

**2. Observations and analysis**

Aerosol particles were collected on the roof of Physics Building of Peking University during five dust-storm events in springs of 1995 and 1996. The building is about 15 m high above the ground and located in the northwest part of Beijing (39°58′N, 116°19′E). Near this site were residences, streets and shopping areas. There was no source of atmospheric pollutants and other higher buildings which might disturb the sampling. Therefore, the observation site was favorable to the measurements of pollutants arriving at Beijing through long-range transport from west or northwest. Particles were collected onto transmission electron microscope grids using two single-stage impactors whose jet diameters were 1 (stage-1) and 0.5 mm (stage-2), respectively. The pumping rate of air was 4.5 l min⁻¹ and airflow passed through stage-1 first and then through stage-2. Therefore, 50% cut-off aerodynamic diameters of the impactors were 0.3 and 0.1 μm, respectively, if the density of particles was assumed to be 2.3 g cm⁻³. The grids were covered with carbon-reinforced holly formvar film. The samples on such grids were applied for morphological analysis and EDX analysis. Nitron film was pre-coated on some sampling grids to detect nitrate-containing particles and barium chloride films were pre-coated on the grids with nitron film to detect sulfate-containing particles and to see internal mixing state of particles of sulfate and nitrate. These reagent films were made in a vacuum evaporator under 10⁻¹⁶ Torr and the thickness of nitron and barium chloride was about 8 and 4 nm, respectively. Sulfate and nitrate in individual particles were identified by the appearance of Liesegang rings, the product of sulfate and barium chloride reaction, and needle-like bundles, the product of nitrate and nitron reaction, respectively. According to the laboratory experiments of Isawa and Ono (1979) and Qian et al. (1991), it was suggested that the detection limits of the reagent film for sulfate and nitrate contained in individual particles were 10⁻¹⁷ and 10⁻¹⁴ g, respectively.

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Table 1
Summary of sampling date, weather and films used for analysis

<table>
<thead>
<tr>
<th>Dust-storm Date</th>
<th>Sampling time (BST*)</th>
<th>Pressure (hPa)</th>
<th>Temp. (°C)</th>
<th>RH (%)</th>
<th>Wind Direction</th>
<th>Wind Speed (m s⁻¹)</th>
<th>Visibility (km)</th>
<th>Analysis films*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1 1995-Mar.-11</td>
<td>14:40–15:00</td>
<td>1011</td>
<td>9</td>
<td>22</td>
<td>NW</td>
<td>14</td>
<td>2</td>
<td>C,N,M</td>
</tr>
<tr>
<td>Case 2 1995-Apr.-05</td>
<td>19:00–19:30</td>
<td>1007</td>
<td>20</td>
<td>26</td>
<td>NW</td>
<td>16</td>
<td>1.5</td>
<td>C,N</td>
</tr>
<tr>
<td>Case 3 1995-Apr.-06</td>
<td>14:00–14:30</td>
<td>1006</td>
<td>19</td>
<td>24</td>
<td>NW</td>
<td>15</td>
<td>1</td>
<td>C,N</td>
</tr>
<tr>
<td>Case 4 1995-Apr.-24</td>
<td>18:00–18:30</td>
<td>998</td>
<td>21</td>
<td>27</td>
<td>W</td>
<td>10</td>
<td>5</td>
<td>C,N,M</td>
</tr>
<tr>
<td>Case 5 1996-May-07</td>
<td>8:40–9:30</td>
<td>1009</td>
<td>20</td>
<td>42</td>
<td>NW</td>
<td>10</td>
<td>6</td>
<td>C,M</td>
</tr>
</tbody>
</table>

*aBeijing standard time (8 hours prior to GMT).

3. Results

3.1. Weather and backward air trajectories

The Beijing Meteorological Observatory reported dust-storms on the days listed in Table 1. Direction of prevailing wind, as shown in Table 1, was westerly or north-westerly and wind speed was larger than 10 m s⁻¹. Relative humidity was lower than 30% during measurements of 1995 and was 42% during the measurement of 1996. The Beijing Meteorological Observatory reported there was a weak precipitation in the early morning in Beijing on 7 May 1996 and that was the possible reason why the relative humidity of case 5 was 42%, much larger than those of other cases.

In the early morning of 11 March 1995 the wind speed increased rapidly. Weak dust-storm occurred in the morning and it became severe from the noon when the sky was heavily yellow and the visibility was very low (Case 1). During the measurements of 5 and 24 April 1995 (Cases 2 and 4) and 7 May 1996 (Case 5), it was apparent that soil particles were lifted up into the air from the ground with the increase of wind speed. Ambiguous blue sky (cases 2 and 4) or cloud (case 5) could be seen over head suggesting dust particles mainly concentrated in low layer near the ground. Weather of 6 April 1995 (Case 3) had much different features from other cases. Around 10:30 a.m., dust initially appeared at high levels when people saw yellow sky but did not feel obvious dust on the ground and the horizontal visibility was rather good. Two hours later, heavy dust appeared near the ground and the visibility was 1 km. These features suggested that dust particles arrived in Beijing through long-range transport on 6 April 1995.

Fig. 1 shows two-day backward isentropic air trajectories from an altitude of 200 m at Beijing. 1, 2, 3 and 4 refer to cases 1, 2, 3 and 4, respectively. Backward trajectories began at 15:00 BST on 11 March 1995 (case 1), 20:00 BST on 5 April 1995 (case 2), 14:00 BST on 6 April 1995 (case 3), and 18:00 BST on 24 April 1995 (case 4). Open squares are marked on the trajectories for every 24 h.

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Gobi (44°40'N, 109°E) of Mongolia and Darhan areas (42°N, 110°40'E) of China. In cases 2 and 4, the sources of dust were possibly Ulansuhai (41°40'N, 108°15'E) and Darhan areas but the sources were not expected to be too far from Beijing. It is noted that air parcels of cases 1 and 3 moved considerably faster than those of cases 2 and 4.

3.2. Morphologies and elemental composition of dust particles

Fig. 2 shows the typical features of particles collected on carbon film during dust-storm events at Beijing, and
together with the typical X-ray spectrums for electron-opaque particles. Most of the collected particles were electron-opaque and in irregular shape. The X-ray spectrums show the peaks of Si, Al, Mg, K, Ca and Fe. These measurements suggest that dominant aerosol particles were dust particles containing minerals. Fig. 2 also indicates that dust particles were mainly collected on the grids of stage-1 and a few of dust particles were collected on the grids of stage-2. Most of dust particles were larger than or near 1 \( \mu m \). Although a large number of particles appeared in the samples of stage-2 grids, some of them, such as particle C in Fig. 2 which were not electron-opaque, should not be dust particles.

A considerable number of cubic electron-opaque particles, besides dust particles described above, appeared in all samples. The regular cubic particles had X-ray spectrums indicating calcium was abundant and little or no other mineral elements existed (hereafter called as Ca-abundant particles). Examples of X-ray spectrum and electron micrograph of those particles are illustrated in Fig. 3, which shows the scale of the particles was from 0.1 to 2 \( \mu m \) and a great number of such particles appeared in submicron range. Frank et al. (1972) suggested that particles showing crystal cube were sea salt particles. But the Ca-abundant particles found in the present study must be different in view of their elemental composition.

Number frequencies of the dust particles and Ca-abundant particles in the samples on carbon films collected in 1995 were summarized on the basis of EDX analysis. Fig. 4 illustrates the frequencies of dust particles as shown in Fig. 2 and Ca-abundant particles as shown in Fig. 3. The frequencies of dust particles were from 87 to 58%, considerably different in different cases. In case 1 there were 41 dust particles among the 47 analyzed particles while in case 4 there were 30 dust particles among the 52 analyzed particles. However, dust particles and Ca-abundant particles together contributed about 93% of the electron-opaque particles in every case.

Number frequencies of elements in the individual particles on carbon films collected in spring 1995 are illustrated in Fig. 5. The frequencies suggest that Mg, Al, K, Ca and Fe were the major components of dust
particles besides Si and a few particles contained Ni, Ti and Mn. These features were frequently suggested in previous research (Zhang et al., 1993; Zaizen et al., 1995). The averaged frequency of sulfur in dust particles was 14.6% except case 4. In the samples of case 4, more than one-third of dust particles simultaneously contained Na, Al, Si, S, Cl, K, Ca and Fe, which was much different from the other three cases.
Fig. 5. Number frequencies of elements among the individual dust particles collected in the dust-storms of spring 1995. ND is the total number of dust particles accounted.

Fig. 6. Electron micrograph of dust particles on carbon film and the X-ray spectrum of particle F: Samples of case 5.

In addition, a few of dust particles which were assumed to be quartz particles were found. An example of electron micrograph and X-ray spectrum of such particles are illustrated in Fig. 6. Only the peak corresponding to silicon in the range of atomic numbers larger than 11 appeared. Okada and Kai (1995) found similar particles in samples of dust-storm in the northwestern China as well.

3.3. Sulfate and nitrate in dust particles

A few of dust particles reacted with the multiple films and needle-like bundles appeared on these particles suggesting that they contained nitrate. An example of electron micrograph and X-ray spectrum of those particles are shown in Fig. 7. The reaction spot indicates there was merely a bit of nitrate in particle G. However, most dust particles did not react with the film as particle H in Fig. 8 although some of them might contain sulfur. The spectrum of particle I in Fig. 9 indicates that particle I contained sulfur but no obvious Liesegang rings appeared around it except some ambiguous traces which might be caused by ammonium. No internal mixed particles which contained simultaneously nitrate and sulfate were found. There were only three mineral particles in all
Fig. 7. Electron micrograph of dust particles on multiple film and the X-ray spectrum of nitrate-containing particle G: Samples of case 5.

Fig. 8. Electron micrograph of dust particles on multiple film and X-ray spectrum of particle H: Samples of case 5.

Fig. 9. Electron micrograph of dust particles on multiple film and X-ray spectrum of particle I: Samples of case 1.
of our samples which reacted with barium chloride film and Liesegang rings appeared clearly around them indicating the presence of soluble sulfate in them.

In order to evaluate nitrate in dust particles, the number frequency of nitrate-containing dust particles detected by nitron films as shown in Fig. 10 in the samples of 1995 was estimated. The numbers of accounted particles were 114, 147, 98 and 97 for cases 1, 2, 3 and 4, respectively. It was found that the frequencies of nitrate-containing particles were 12.3, 10.2, 11.2 and 9.3%, respectively, and the average frequency of the four cases was 10.8%.

4. Discussion

4.1. Sulfur and sulfate in dust particles

About 14.6% of the dust particles collected in Beijing contained sulfur but few dust particles contained water-soluble sulfate according to EDX analysis (Fig. 5) and reagent film tests (Fig. 9). EDX analysis in the present study showed the possibility that sulfur in dust particles was frequently accompanied by calcium or sodium. For example, 7 of the 41 dust particles contained sulfur in case 1. All of the 7 particles contained calcium or sodium (6 contained calcium and 4 contained sodium). Moreover, there were 4 sodium sulfate particles, characterized by round shapes and sodium and sulfur peaks in their X-ray spectrum, in the analyzed 47 particles. Besides, the concentration of ammonium in the urban atmosphere in Beijing during dust-storm events is about 2.27 µg m⁻³ (Yang et al., 1991). Therefore, water-soluble sulfate, if existed, in dust particles possibly was present as sodium sulfate, calcium sulfate, ammonium sulfate or their mixture.

Since few dust particles made Liesegang ring through reactions with multiple film, the amount of water-soluble sulfate must be too less to react with barium chloride film. It is unsure dust particles did not contain water-soluble sulfate because the limitation of the detection technique. But dust particles collected in Beijing did not contain considerable water-soluble sulfate from our results.

Many investigators have suggested that particulate sulfate formed in the atmosphere through homogeneous and heterogeneous reactions (McArkle and Hoffmann, 1983; Martin, 1984; Hoffmann, 1986; Seinfeld, 1986). Heterogeneous reactions occur in the aqueous surface layer of pre-existing particles where SO₂ react with O₃ or H₂O₂ to produce sulfate (McArkle and Hoffmann, 1983; Martin, 1984; Hoffmann, 1986). Roberts and Friedlander (1975) reported that heterogeneous conversion of SO₂ to sulfate was two-orders faster than homogeneous conversion in Los Angeles atmosphere. Recent measurements (Okada et al., 1990; Li-Jones and Prospero, 1998) and model simulation (Dentener et al., 1996) also suggested the effectiveness of heterogeneous formation of sulfate on dust particles. However, heterogeneous conversion is sensitive to humidity and its efficiency decreases with the decrease of humidity. Experiments in a smog chamber suggested the conversion when RH = 30% was slower by two times than that when RH = 80% (Yang and Wang, 1994). It is noted from Table 1 that relative humidity was very small during dust-storm episodes in Beijing. The high relative humidity of 7 May 1996 was possibly due to the precipitation in the early morning but the humidity of the air parcels containing dust particles should not be as large as 42%. Therefore, sulfate was hardly formed through heterogeneous reactions on the dust particles due to the lack of H₂O. Atmospheric SO₂ concentration is certainly an important factor controlling sulfate formation. As described later, SO₂ concentration was expected to be too small.

Another possible mechanism of sulfate formation is dust particles absorbed sulfuric acid (gas or droplets) which were formed through homogeneous reactions of SO₂, OH, O₂, H₂O, etc. (Seinfeld, 1986). Such reactions should not be sensitive to humidity. As shown by the backward trajectories, air parcels containing dust-storm particles were originated from arid or semi-arid areas in Mongolia and northern or northwestern China. In these areas, SO₂ emission is weak (Langner and Rodhe, 1991).
During their transport, the air parcels might have passed over some cities, where anthropogenic SO$_2$ was emitted due to human activities. But the emission was pretty small and not to anticipate to impact those air parcels considerably (Akimoto and Narita, 1994). Therefore, homogeneous formation of sulfate should not be efficient for the lack of SO$_2$.

Chemical analysis of bulk-sampled particles suggests that sulfate concentration in Beijing urban atmosphere during dust-storm periods is almost the same as that during non-dust-storm periods (Yang et al., 1991). Analysis of individual particles suggests that about 20% of coarse particles (diameter > 1 μm) and more than 70% of fine particles (1 μm > diameter > 0.2 μm) collected in urban area of Beijing during non-dust-storm periods contain water-soluble sulfate (Zhang et al., 1995). Based on these results, it is concluded that almost no sulfate is formed on the surface of dust particles during their transport from source areas to Beijing. It is reasonable to consider that a few of dust particles contain sulfur because desert soil particles of China have a relatively large fraction of sulfur in the form of CaSO$_4$ (Hseung and Jackson, 1952; Tanaka et al., 1986).

4.2. Nitrate in dust particles

Nitrate, in opposition to sulfate, was found in some dust particles and the average frequency of nitrate-containing dust particles in the four cases of 1995 was 10.8%. Zhang (1996) reported about 45% of mineral particles collected in the urban atmosphere in Beijing during non-dust-storm periods were nitrate bearers. Observations in Beijing indicate that the concentration of total suspended particles during dust-storm periods is larger by 5-10 times than that during non-dust-storm periods and the weight concentration of nitrate during dust-storm periods is approximately equal to that during non-dust-storm periods (Yang et al., 1991; Quan, 1995).

If mineral particles originated from Beijing areas consisted of similar number of nitrate-containing particles as in non-dust-storm periods and no nitrate were formed on dust particles during their transport, the number frequency of nitrate-containing particles among dust particles collected during dust-storm periods should have been considerably smaller than 10.8%. It is proved that dust particles originated from northern and northwestern China do not contain nitrate in their source areas (KOSA, 1991). Therefore, nitrate must be formed on the surface of some dust particles although the formation was not so efficient as to impact apparently the concentration of nitrate in the urban atmosphere in Beijing.

Generally, heterogeneous formation of nitrate on pre-existing particles is much more efficient than homogeneous formation in the atmosphere (Seinfeld, 1986). However, H$_2$O is needed on particle surface as reactant in the heterogeneous reactions and atmospheric water vapor also condenses onto particle surface during nitrate formation. As described previously, the air parcels containing dust particles were pretty dry. Subsequently, the conversion of NO$_x$ to nitrate on the surface of dust particles should not be efficient. On the other hand, the absorption of nitric acid by dust particles is independent of the humidity of ambient air (Fenter et al., 1995). Nitric acid can be formed through homogeneous reactions between NO$_x$ and OH or N$_2$O$_5$ and H$_2$O occurring in the atmosphere (Seinfeld, 1986). Although we did not measure the concentration of NO$_x$ and OH during the dust-storm periods, the observation clearly showed the existence of particulate nitrate on the surface of dust particles. Therefore, there should be some nitrate-containing dust particles due to the absorption of nitric acid and/or NO$_x$ gases.

4.3. Ca-abundant particles

Ca-rich particles are found in dust samples collected in northwestern China (Okada and Kai, 1995) and in North Pacific regions (Okada et al., 1990; Zhou et al., 1996). The Ca-rich particles are in irregular shapes and the particles collected in North Pacific regions contain S besides Ca, Si and Al. And they are attributed to be mainly composed of CaCO$_3$. Ca-abundant particles in regular cubic shapes were found in the range from 0.1 to 2 μm in the present study (Fig. 3). Their X-ray spectrums indicated calcium was the main component and little or no other mineral elements existed in them. The Ca-abundant particles seemed not to be composed of CaCO$_3$ because of their regular cubic shapes. CaCO$_3$ is the main component of calcite in the nature and natural calcite particles should not be in regular shapes. Moreover, it is worth pointing out that dust particles originated from Asian continent are generally larger than 1 μm (Zhang et al., 1993; Zaizen et al., 1995). A large number of Ca-abundant, as shown in Fig. 3, were in the range smaller than 1 μm. It has been proved that calcium is dominant cation in the air pollutants in Beijing (Huebert et al., 1988). Analysis of individual particles collected during non-dust-storm periods indicates that cubic particles always appear in the samples in the urban atmosphere in Beijing and a few of them contain sulfate (Zhang et al., 1995; Zhang, 1996). The present chemical tests using multiple film revealed most of such particles did not react with barium chloride, as particle J in Fig. 11.

One possible explanation is that the Ca-abundant particles were formed in air through crystallization in view of their regular cubic shapes and scales of 0.1–2 μm. The frequency of Ca-abundant particles among dust particles in the samples of 24 April 1995 (case 4) was much larger than those of 11 March and 5 and 6 April 1995 (cases 1, 2 and 3) (Fig. 4). Table 1 indicates that the pressure of 24 April 1995 was much smaller than those of the other three cases, and the meteorological records revealed that
a strong low-pressure system was passing over Beijing on 24 April 1995 while the collection was carried out (Gunji, 1995). It is reasonable to consider that local sources of particulate matter in Beijing diffused lots of anthropogenic particles under unstable meteorological conditions associating with the low-pressure system (Gunji, 1995).

The most possible composition of the Ca-abundant particles is CaO or Ca(OH)$_2$, emitted from construction sites because there are a large number of construction sites in urban areas in Beijing in recent years, where lime, mainly composed of CaO or Ca(OH)$_2$, is well applied (Wang, 1985). Consequently, it is suggested that Ca-abundant particles found in the present study should not be dust particles originated from the desert areas in northern and northwestern China.

5. Summary

Aerosol particles were collected in Beijing during five dust-storm events (four in spring 1995 and one in spring 1996). The shape and chemical composition of particles were examined using a transmission electron microscope. Elements with atomic number larger than 11 were obtained by EDX analyzer attached to the microscope. Sulfate and nitrate in dust particles were detected by chemical test of barium chloride and nitron films pre-coated on the collecting grids of electron microscope.

The chemical test and EDX analysis indicated that few dust particles contained water-soluble sulfate although some particles contained sulfur, and 10.8% of dust particles contained nitrate. These results suggested that sulfate was hardly formed and merely a little of nitrate was formed on the surface of dust particles during their long-range transport before arriving in Beijing.

Ca-abundant particles which were in regular cubic shapes and consisted of only calcium among mineral elements were found in all samples of the five cases. Morphology, composition and weather analyses suggested that they were not originated from northern or northwestern China as dust particles but were possibly emitted from construction sites of the local areas initially and formed through crystallization in the air. Their main composition was possibly CaO or Ca(OH)$_2$.

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