Chemical characterization of submicron particles during typical air pollution episodes in spring over Beijing

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ABSTRACT
Beijing experienced a long-lasting pollution episode in April 2012. Here, the authors characterize the sources and evolution processes of the pollution, with a focus on a haze and a dust episode that occurred during 15–30 April. All submicron aerosol species (PM$_1$) showed substantial increases during the haze episode (HE), with nitrate being the largest, contributing on average 32.2% of the non-refractory PM$_1$ (NR-PM$_1$) mass (maximum: ~42.0%), which was different from other seasons. Secondary inorganic aerosol (SIA) species (55.1 ± 23.8 μg m$^{-3}$; SIA = nitrate, ammonium, sulfate) played enhanced roles in the haze formation, accounting for 66.5% of the total NR-PM$_1$ mass. In contrast to the HE, organics exhibited their highest levels (8.9 ± 5.4 μg m$^{-3}$) among the NR-PM$_1$ aerosol species during the dust episode, accounting for 64.0% on average. SIA exhibited low concentration levels (4.6 ± 4.2 μg m$^{-3}$) during the dust episode, contributing only 34.0% to the total NR-PM$_1$ mass. This study suggests that air quality improvement strategies should be designed according to aerosol particle characteristics and formation mechanisms.

1. Introduction
China is currently facing tremendous challenges with respect to air quality because of the substantial increases in anthropogenic emissions associated with rapid industrialization, motorization, and urbanization (Chan and Yao 2008). Beijing, the capital of China, is the national center for politics, culture, and international business. The city’s population is 21 million and the population density is greater than that of other global megacities, e.g. London and Tokyo. The high population density and rapid economic growth have inevitably resulted in high air pollutant emission levels in Beijing. Decreased visibility is one of the most notable effects of aerosol pollution. In the Beijing region, visibility showed a decreasing trend during 1980–2008, and the number of hazy days has increased since the 1990s (Zhao et al. 2011). Due to the many negative effects on human health, in recent years air quality problems have received much public and governmental attention (Li et al. 2014; Zhang, Song, and Cai 2007).

Air pollution in Beijing is a complex phenomenon, so a clear understanding of its characteristics and formation mechanisms is the first priority in the development of effective environmental protection policies. Research on the various chemical components of airborne particles can provide useful information for understanding the key aspects of the formation mechanisms (Sun et al. 2013b). Multiple studies have been conducted on the chemical characteristics of atmospheric aerosols in Beijing in recent years. Crustal matter, secondary inorganic aerosols (SIAs), and organic materials have been identified as the three main chemical components of the aerosols in Beijing (Zhao et al. 2013). Sulfate (SO$_4^{2-}$), nitrate (NO$_3^-$), and...
ammonium (NH$_4^+$) represent the dominant water soluble inorganic ions of aerosols during haze episodes (HE) (Sun et al. 2013b). Ammonium sulfate ([NH$_4^+$]$_2$SO$_4$), ammonium nitrate (NH$_4$NO$_3$), and calcium nitrate (Ca(NO$_3$)$_2$) are the major species present on hazy spring days (Wang et al. 2006a). Wang et al. (2006a) found that SO$_4^{2-}$ and calcium ions (Ca$^{2+}$) were the two primary chemical components during dust episodes in Beijing.

Most previous studies have been conducted using filter samples, which have two disadvantages as compared with online analysis when studying the rapid evolution processes of different types of serious air pollution in Beijing. These problems are as follows: (1) The temporal resolution is poor, being one day or even longer, and the sub-micron particulate matter (PM$_{1}$) concentration can increase to more than 500 μg m$^{-3}$ in less than half day (Sun et al. 2014). As such, filter samples do not provide the temporal resolution necessary for investigating the formation and transformation of particles. (2) Some volatile components deteriorate between the time of sampling and analysis. For example, NH$_4$NO$_3$ will evaporate significantly to gas-phase nitric acid (HNO$_3$) and ammonia (NH$_3$) under relatively high temperatures (lanniello et al. 2011). The 15-min online chemical analysis used in our study makes up for these disadvantages, and can adequately measure chemical species in PM$_{1}$, which illustrates the most adverse effects on public health (Polichetti et al. 2009).

In this study, we employed various high temporal resolution instruments to investigate the rapid evolution of aerosol properties during a lengthy haze pollution episode and a dust storm in Beijing (15–30 April 2012). We compared the chemical composition of non-refractory PM$_{1}$ (NR-PM$_{1}$) during hazy, clean, and dusty days to determine the differences in their formation mechanisms. In particular, our online analysis captured a rare occurrence of a high NO$_3^-$ concentration in the NR-PM$_{1}$ mass. We also investigated the chemical composition of NR-PM$_{1}$ during the dust episode, which offered insights into submicron particle formation during alkaline conditions.

2. Methods

2.1. Sampling site

Field measurements were carried out at the IAP, Chinese Academy of Sciences (39°58′28″N, 116°22′16″E), which is located between the north 3rd and 4th ring roads in Beijing, during 15–30 April 2012. The sampling site is located on the roof of a second-story building (~8 m high). All aerosol and gaseous species, including NR-PM$_{1}$, species, fine particles (PM$_{2.5}$), nitric oxide (NO), nitrogen dioxide (NO$_2$), total reactive nitrogen (NO$_x$), ozone (O$_3$), sulfur dioxide (SO$_2$), and carbon monoxide (CO), were measured at this site. The meteorological variables, including temperature, RH, wind speed, and wind direction, were obtained from the first floor (8 m) of the meteorological tower (325 m) at the IAP.

2.2. Aerosol and gaseous species measurements

NR-PM$_{1}$ aerosol species, including organics, SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and chloride (Chl), were measured in situ with an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) (Ng et al. 2011). Positive matrix factorization (PMF), using the PMF2.exe algorithm (v4.2) in robust mode (Paatero and Tapper 1994), was performed on the ACSM organic aerosol (OA) mass spectra following the procedures described in Ulbrich et al. (2009). The PMF2 solutions were then evaluated with an Igor Pro-based PMF Evaluation Tool (PET, v2.04) (Ulbrich et al. 2009) following the procedures detailed in Zhang et al. (2011). The two-factor solution, including a hydrocarbon-like OA (HOA) and an oxygenated OA (OOA) with $f_{\text{peak}} = 0$, was chosen in this study.

The PM$_{2.5}$ mass was simultaneously measured using a Tapered Element Oscillating Microbalance (TEOM series 1400a, Thermo Scientific, USA), and the collocated gaseous species, including CO, SO$_2$, NO$_2$, NO/NO$_x$, and O$_3$, were measured using various gas analyzers from Thermo Scientific.

3. Results and discussion

3.1. Identification of two pollution episodes

Figure 1 shows the temporal variations of the mass concentrations of NR-PM$_{1}$ and PM$_{2.5}$ and the meteorological conditions during the measurement period. Two distinct pollution episodes with largely different NR-PM$_{1}$/PM$_{2.5}$ ratios were observed. The HE was formed after a precipitation event on 16 April, and lasted more than one week until 24 April. The average (±1σ) concentrations of NR-PM$_{1}$ and PM$_{2.5}$ were 82.8 ± 34.3 μg m$^{-3}$ and 98.7 ± 50.6 μg m$^{-3}$, respectively. As illustrated in previous work, the averaged NR-PM$_{1}$ concentration was 50 μg m$^{-3}$ in summer (Sun et al. 2012) and 66.8 μg m$^{-3}$ in winter (Sun et al. 2013a). The average PM$_{2.5}$ mass concentration was almost three times the National Ambient Air Quality Standards (NAAQS) of the U.S. Environmental Protection Agency (35 μg m$^{-3}$, 24 h average), and approximately 1.3 times the second grade of China’s NAAQS (75 μg m$^{-3}$, 24 h average).

The average ratio of NR-PM$_{1}$/PM$_{2.5}$ was 0.84 during HE, indicating that submicron particles dominated the total mass of fine particles. The meteorological conditions were stagnant. The winds were dominantly from the south with low speed (~0.6 m s$^{-1}$) and the RH was high (>60%). The stagnant dispersion conditions were also caused by a
low-pressure system, the front of which persisted around Beijing during HE. As a result, the mass concentration of PM$_{2.5}$ remained consistently high during HE until the meteorological conditions had a noticeable change on 24 April. The back trajectories in Figure 1(e) show that the air masses during HE were dominantly from the south and southeast, with relatively short distances. The MODIS true color composite image on 22 April further showed evident gray haze clouds over the North China Plain, including Beijing, indicating that HE formed over a regional scale (Figure 1(f1)).

The dust episode (DE) formed on 27 April in association with a rapid decrease in RH (from >90% to <10%) and a corresponding increase in wind speed (Figure 1). The PM$_{2.5}$ mass concentration continuously increased and reached a maximum of 259.3 μg m$^{-3}$ at 02:25, 29 April (LST). The average mass concentration of PM$_{2.5}$ during DE was 133.2 ± 70.5 μg m$^{-3}$, which was even higher than that observed during HE. However, the NR-PM$_1$ mass remained consistently low during DE, with an average concentration of 13.9 ± 9.4 μg m$^{-3}$. The average ratio of NR-PM$_1$/PM$_{2.5}$ was ~0.1, which was much lower than that during HE, indicating different chemical characteristics of the pollution between HE and DE. Given that the ACSM does not measure the refractory dust component at a vaporizer temperature of ~600 °C, the low NR-PM$_1$/PM$_{2.5}$ ratio was primarily caused by the largely elevated dust particles during DE. This was also supported by the evident yellow dust
3.2. Temporal variation of aerosol composition

Figure 2 shows the time series of the mass concentration and mass fractions of NR-PM$_1$ species measured by the ACSM. The pie charts show the average chemical compositions of NR-PM$_1$ during HE, DE, and a clean period (25 April 2012). (c) Average diurnal cycles of the NO$_3^-$ mass concentration and PBL height normalized NO$_3^-$ during HE.

Table 1. Summary of the mass concentrations of NR-PM$_1$ species and PM$_{2.5}$ (μg m$^{-3}$); gaseous concentrations of SO$_2$, NO, NO$_2$, and CO (ppb); and meteorological parameters including wind speed (WS, m s$^{-1}$), RH (%), and temperature (Temp, °C).

|          | HE       | DE       | Clean  |
|----------|----------|----------|--------|
| NR-PM$_1$| 82.8     | 13.9     | 5.7    |
| Org      | 25.8     | 8.9      | 4.6    |
| HOA      | 5.4      | 3.9      | 2.3    |
| OOA      | 20.6     | 4.3      | 1.6    |
| SO$_4$   | 12.4     | 1.6      | 0.4    |
| NO$_3$   | 26.7     | 1.4      | 0.2    |
| NH$_4$   | 16.0     | 1.6      | 0.5    |
| Chl      | 1.9      | 0.4      | 0.1    |
| PM$_{2.5}$ | 103.8  | 133.3    | 8.8    |
| NO       | 43.9     | 51.9     | 28.4   |
| NO$^+$   | 14.0     | 15.1     | 12.8   |
| NO$_2$   | 34.7     | 27.5     | 12.9   |
| SO$_2$   | 12.4     | 8.7      | 4.6    |
| O$_3$    | 15.1     | 21.2     | 23.6   |
| CO       | 2286.3   | 575.6    | 398.0  |
| Temp     | 16.6     | 16.8     | 15.9   |
| RH       | 63.1     | 19.0     | 16.9   |
| WS       | 0.6      | 0.8      | 1.1    |

Clouds on 27 April over the North China Plain (Figure 1(f2)). The back trajectories confirmed that the air masses during DE originated from the Gobi Desert in Mongolia and Inner Mongolia (Figure 1(e2)). These results together suggested that DE was an episode with strong dust storm influences.

3.2. Temporal variation of aerosol composition

Figure 2 shows the time series of the mass concentration and mass fractions of NR-PM$_1$ species (organics, SO$_4$, NO$_3$, NH$_4$, and Chl) for the entire study. Sias (NO$_3^-$ + NH$_4^+$ + SO$_4^{2-}$) dominated NR-PM$_1$ during HE, on average accounting for 66.5% of the total NR-PM$_1$ mass. The SIA mass was elevated by a factor of ~11–50 compared to that observed during the clean period (Table 1). Previous results illustrated that SO$_2$, NO$_3^-$, and NH$_4^+$ to together accounted for 54%–61% of the total PM$_{2.5}$ mass, based on filter work carried out in spring in Beijing (Shen et al. 2016), which corresponds well with our finding that NO$_3^-$, SO$_2^-$, and NH$_4^+$ were the most important fractions of submicron particles. In comparison, organics contributed 31% of NR-PM$_1$ during HE, with OOA—a surrogate of secondary OA—accounting for 79% of OA. These results suggest that HE was primarily caused by secondary formation. Enhanced roles of secondary species during HE have also been observed previously at various sites in eastern China (Huang et al. 2012; Li et al. 2011; Sun et al. 2006; Tan et al. 2009; Wang et al. 2006a). However, different from previous studies, NO$_3^-$ was the most abundant secondary species, contributing 32.2% of NR-PM$_1$, on average. The NO$_3^-$ contribution was much higher than those observed in summer (16%–25%) (Sun et al. 2010, 2012) and...
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(Figure 3), which facilitated the formation of NO₃ radicals and dinitrogen pentoxide (N₂O₅), and hence the heterogeneous formation of HNO₃ under high conditions of RH. The results are consistent with the findings of Parrish and Zhu (2009); namely, that NO₃⁻ was most probably formed via the heterogeneous hydrolysis process of N₂O₅ on the surface of moist and acidic aerosols in Beijing and Shanghai, especially under humid and hazy weather. Photochemical production might also have been important, as suggested by the higher concentration of NO₃⁻ in the afternoon by excluding the dilution effects of the PBL (Figure 2(c)).

Moreover, the nitrogen oxidation ratio, NOR = \( \frac{n\text{NO}_{3}^-}{n\text{NO}_{3}^- + n\text{NO}_2} \) (where \( n \) refers to molar concentration) during HE (0.21 ± 0.06) was also much higher than that during other periods (Figure 3(b)). All these factors together led to the high concentration of NO₃⁻ in spring. Indeed, NO₃⁻ exceeded winter (14%–16%) (Sun et al. 2013a; Zhang et al. 2014), which indicates that NO₃⁻ played a leading role in the formation of HE. As shown in Figure 2, the NO₃⁻ contribution reached up to 42% during HE. Higher contributions of NO₃⁻ to PM in spring than other seasons were also observed by filter sampling (Zhang et al. 2013). The formation of NO₃⁻ is mainly driven by three processes: daytime photochemical production; gas-particle partitioning; and nighttime heterogeneous reactions. As shown in Table 1, the temperature averaged 16.6 °C during HE, under which NH₄NO₃ will not evaporate significantly to gas-phase HNO₃ and NH₃. This is further supported by the relatively flat diurnal cycle of NO₃⁻ (Figure 2). In addition, the high humidity (63.1%) facilitated the uptake of HNO₃ into the particle phase, and also nighttime heterogeneous reactions. For example, most of the time during HE was associated with high NO₂ and O₃ (Figure 3), which facilitated the formation of NO₃ radicals and dinitrogen pentoxide (N₂O₅), and hence the heterogeneous formation of HNO₃ under high conditions of RH. The results are consistent with the findings of Parrish and Zhu (2009); namely, that NO₃⁻ was most probably formed via the heterogeneous hydrolysis process of N₂O₅ on the surface of moist and acidic aerosols in Beijing and Shanghai, especially under humid and hazy weather. Photochemical production might also have been important, as suggested by the higher concentration of NO₃⁻ in the afternoon by excluding the dilution effects of the PBL (Figure 2(c)).

Moreover, the nitrogen oxidation ratio, NOR = \( \frac{n\text{NO}_{3}^-}{n\text{NO}_{3}^- + n\text{NO}_2} \) (where \( n \) refers to molar concentration) during HE (0.21 ± 0.06) was also much higher than that during other periods (Figure 3(b)). All these factors together led to the high concentration of NO₃⁻ in spring. Indeed, NO₃⁻ exceeded
all other secondary species for most of the time during HE (Figure 2).

SO$_4^{2-}$ also showed a significant increase during HE (12.4 ± 5.7 μg m$^{-3}$) compared to DE (1.6 ± 1.5 μg m$^{-3}$) and the clean period (0.4 ± 0.1 μg m$^{-3}$). The larger contribution of SO$_4^{2-}$ during HE could have been related to the higher oxidation ratio of SO$_2$. The average sulfur oxidation ratio (SOR) was 0.19 ± 0.08 during HE, which was almost five times higher than during DE (0.04 ± 0.01) and the clean period (0.02 ± 0.01). The increased SOR indicated an enhanced secondary formation during HE. The conversion of SO$_2$ to SO$_4^{2-}$ is mainly attributed to the gas-phase oxidation by OH radicals and/or heterogeneous oxidation by hydrogen peroxide, O$_3$, and oxygen/iron/manganese etc. (Calvert et al. 1985; Seinfeld and Pandis 2006). Many studies have suggested that the conversion of SO$_2$ to SO$_4^{2-}$ during HE takes place dominantly via aqueous-phase oxidation (Li et al. 2011; Sun et al. 2006; Tan et al. 2009). In this study, the high RH during HE was a favorable factor for the aqueous-phase oxidation of SO$_2$. Despite this, the contribution of SO$_4^{2-}$ to NR-PM$_1$ remained relatively constant (~15%) during HE. The mass ratio of NO$_x$/SO$_4^{2-}$ in the present study was 2.2 ± 0.8, which was significantly higher than observed in 2006 (0.64) (Wang et al. 2006b) and 1999–2000 (0.43) (Yao et al. 2002; Ye et al. 2003). The higher ratio of NO$_x$/SO$_4^{2-}$ further demonstrates the significant role of NO$_3^-$ in haze formation in spring.

Compared to HE, NR-PM$_1$ contributed only ~10% to the PM$_{2.5}$ mass during DE. Given that the ACSM does not measure the refractory dust component at a vaporizer temperature of ~600 °C, the low NR-PM$_1$/PM$_{2.5}$ ratio was primarily caused by the largely elevated dust particles during DE. According to the findings of Wang et al. (2005), the mass fraction of mineral dust can reach up to ~94.3% of PM$_{2.5}$ in a super dust storm and ~91% during a regular dust storm in Beijing (Wang et al. 2006a). The NR-PM$_1$ during DE was mainly composed of organics. Organics exhibited their highest levels (8.9 ± 5.4 μg m$^{-3}$) among NR-PM$_1$ aerosol species during DE, accounting for 64.0% on average. This was significantly different from the dominance of SIA during HE. However, the organic contribution was similar to that observed during winter clean periods (63%) (Sun et al. 2013a), when the air masses were from the upwind areas of Beijing and characterized by a high mass fraction of organics (Sun et al. 2012). A similar dominance of organics at low PM loadings was also observed at the rural site of Yufa (Takegawa et al. 2009) and the urban site of Peking University (Huang et al. 2010). The organics at low mass loadings are characterized by a dominant contribution from local traffic and cooking emissions (Sun et al. 2012). SIA$s$ exhibited low concentration levels (4.6 ± 4.2 μg m$^{-3}$) during DE and contributed 34% to the total NR-PM$_1$ mass. Inorganic NO$_3^-$ showed an apparently lower contribution to the total NR-PM$_1$ mass during DE (10%) than HE (32%), which can be explained by low precursors (nitrogen oxides, NO$_x$) and humidity. Secondary SO$_4^{2-}$ presented a similar contribution to the total NR-PM$_1$ mass during DE and HE (12% and 15%, respectively), although the mass concentrations of the two episodes were markedly different (1.6 μg m$^{-3}$ versus 12.4 μg m$^{-3}$, Table 1). The ratio of NO$_3^-/SO_4^{2-}$ also showed a large decrease, from 2.2 during HE to 0.9 during DE. The NO$_3^-/SO_4^{2-}$ ratio during DE was higher than the value of ~0.3 during 2001–04 dust episodes in Beijing (Wang et al. 2006a). According to the results of Zhang et al. (2016), the control of SO$_2$ emissions has been proven to be successful in Beijing from 1998. In comparison, NO$_x$ reduction was less obvious from 2008 through 2013 in Beijing, possibly attributable to the increase in vehicle emissions counteracting the reduction of coal-burning emissions (Zhang et al. 2015). Because the RH during DE was low, the heterogeneous reactions of SO$_2$ on dust particles might have dominated the SO$_4^{2-}$ formation (He et al. 2014; Wang et al. 2006a). In addition, the surface soil in desert areas might have been another important source of SO$_4^{2-}$ during DE (Wu et al. 2012).

4. Summary

We measured the aerosol particle composition and trace gases at an urban site in the megacity of Beijing during 15–30 April 2012. Two distinct pollution episodes were identified, i.e. HE and DE, on the basis of measurements of visibility, aerosol composition, satellite images, and air mass trajectories. The average concentrations of NR-PM$_1$ and PM$_{2.5}$ during HE were 82.8 ± 34.3 μg m$^{-3}$ and 98.7 ± 50.6 μg m$^{-3}$, respectively, with an average NR-PM$_1$/PM$_{2.5}$ ratio of 0.84. DE, however, showed a much lower NR-PM$_1$ concentration (13.9 ± 9.4 μg m$^{-3}$), and higher concentrations of PM$_{2.5}$ (133.2 ± 70.5 μg m$^{-3}$). The average NR-PM$_1$/PM$_{2.5}$ ratio was −0.1, indicating the dominance of PM$_{2.5}$ dust particles during DE.

The SIA$s$ (NO$_3^-$, NH$_4^+$, and SO$_4^{2-}$) exhibited their highest loading values during HE. The average SIA concentration was 55.1 ± 23.8 μg m$^{-3}$, accounting for 66.5% of the total NR-PM$_1$ mass. These results suggest that the secondary formation of aerosols was the major factor driving the formation of HE. Among the SIA$s$, NO$_3^-$ exhibited the highest loading value during HE, accounting for 32.2% of the total NR-PM$_1$ mass. This demonstrates the important role of NO$_3^-$ in spring haze formation, which differs significantly from that of other seasons. The high concentration of NO$_3^-$ in spring was likely due to the high concentrations of precursors, low temperatures, and high humidity, which inhibited the evaporative loss of NH$_4$NO$_3$ particles. During DE, the NR-PM$_1$ concentration remained at a low ambient level, with OA constituting the major fraction (64.0%).
Disclosure statement
No potential conflict of interest was reported by the authors.

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