

In situ measurement of PM₁ organic aerosol in Beijing winter using a high-resolution aerosol mass spectrometer

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Organic aerosol (OA) is a crucial component of atmospheric fine particles. To achieve a better understanding of the chemical characteristics and sources of OA in Beijing, the size-resolved chemical composition of submicron aerosols were measured *in-situ* using a High-Resolution Time-of-Flight Aerosol Mass Spectrometer in the winter of 2010, with a high time resolution of 5 min. During this study, the mean OA mass concentration was $20.9 \pm 25.3 \mu\text{g}/\text{m}^3$, varying between 1.9 and $284.6 \mu\text{g}/\text{m}^3$. Elemental analysis showed that the average H/C, O/C and N/C (molar ratio) were 1.70, 0.17, and 0.005, respectively, corresponding to an OM/OC ratio (mass ratio of organic matter to organic carbon) of 1.37. The average mass-based size distributions of OA present a prominent accumulation mode peaking at approximately 450 nm. The prominent presence of ultrafine particles ($D_{\text{va}} < 100 \text{ nm}$) was mainly from the fresh emissions of combustion sources. A Positive Matrix Factorisation (PMF) analysis of the organic mass spectral dataset differentiated the OA into three components, including hydrocarbon-like (HOA), cooking-related (COA), and oxygenated (OOA) organic aerosols, which, on average, accounted for 26.9%, 49.7% and 23.4%, respectively, of the total organic mass. The HOA and COA likely corresponded to primary organic aerosol (POA) associated with combustion-related and cooking emissions, respectively, and the OOA components corresponded to aged secondary organic aerosol (SOA).

high-resolution aerosol mass spectrometer, organic aerosol, size distribution, elemental composition, positive matrix factorisation

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Atmospheric aerosols, especially fine particles, have been found to play an important role in global climate change by altering the radiative balance of the Earth's atmosphere directly and indirectly. These aerosols also pose a significant threat to human health by increasing the risk of morbidity and mortality of sensitive groups [1,2]. These submicron particulates (PM₁) exhibit lengthy atmospheric residence time and are capable of deep pulmonary ingestion [3,4]. The environmental and health impacts of PM₁ are significant. Organic aerosol (OA) is a large and important fraction of PM₁ and has great impact on human health, especially because some of the compounds have proved to be mutagens

or carcinogens, i.e. polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and unsaturated aldehydes [5,6]. Fine particulate matter (PM) in densely populated megacity environments is of particular concern because of their adverse effects on human health and regional air quality [7,8]. Along with fast economic growth, the air quality in Beijing has suffered severe deterioration, with PM being one of the top pollutants [9,10]. Organic matter is the most abundant fraction in fine particles in Beijing, accounting for 30%–50% of the total mass [11], indicating its key role in air pollution control. In addition, because the chemical compositions of OA are known to be effective at providing useful source apportionment information, understanding the characteristics of OA in depth is

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of great importance. Because the chemical compositions and sources of OA are complex (they can be classified as either primary OA (POA), from direct emissions, or secondary OA (SOA), from the oxidation of gas-phase precursors), the related studies on their chemical and physical characteristics have become difficult and a recent focus point.

Most aerosol chemistry studies in Beijing so far have been based on filter sampling that offers information at a coarse time resolution and is vulnerable to interference by absorption and adsorption in the sampling process. High time-resolution online measurements are needed to gain insight into aerosol chemical characteristics, sources and processes. The Aerodyne High-resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS), which is one of the most advanced instruments in the field of aerosol research, can be used for the real-time measurement of the highly time- and size-resolved chemical compositions of non-refractory (NR) components in submicron aerosols. Compared with many developed countries, the application of AMS for ambient studies in China has been relatively new and has begun only in recent years [6]. The rare AMS in field measurements in Beijing are mainly conducted in summer [12–14], although many previous studies reported that the seasonal variation of fine particulate matter in Beijing, especially organic aerosols, peak in wintertime [15,16]. In addition, due to the impacts of residential heating and special meteorological conditions such as low temperature and low humidity in winter, the compositions and sources of OA should be significantly different compared with other seasons. In this study, to reveal the variation regularity of organic aerosols in the winter in Beijing, the online observation of chemical compositions and size distributions of OA was performed by HR-ToF-AMS with a 5-min time resolution. Meanwhile, based on the data of the mass spectra of organic compounds, major sources of OA were resolved by positive matrix factorisation (PMF).

1 Methods and materials

(i) Sampling site description. From 14 December 2010 to 15 January 2011, an Aerodyne HR-ToF-AMS was placed at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS), to measure on-line the chemical compositions and size distributions of ambient aerosols. The observation site is located between the north 3rd Ring Road and north 4th Ring Road. The site is approximately 1 km from the 3rd Ring Road, 200 m from the Badaling Highway running north-south to its east, and 50 m from the Beitucheng West Road running east-west to its north. Ambient air was sampled at approximately 15 m above ground. The HR-ToF-AMS mass spectrum was scanned from m/z 12 to 596, and the sampling flow rate was 90 mL/min. The time resolution for all acquisition data was set to 5 min; therefore,

9504 sets of data in total were obtained during the observation. At the same time, an automatic meteorological observation instrument called Milos520 (Vaisala, Finland) was located at the 8 m level and was used to observe the main meteorological parameters, including pressure, temperature, humidity, and the speed and direction of wind in the atmosphere at ground level [17]. The datasets of CO and NO_x were acquired from the Beijing-Tianjin-Hebei Atmospheric Environment Monitoring Network [18].

(ii) The principle of HR-ToF-AMS. In the HR-ToF-AMS, an aerodynamic lens is used to sample and focus ambient particles into a narrow beam that is transmitted to a heated surface (~600°C), where particles are flash vaporised upon impact. The resulting vapour molecules are ionised by 70 eV electron impactation (EI) and then the positive ions are analysed by a ToF mass spectrometer. Because the aerodynamic lens has reduced transmission efficiencies for particles at sizes of approximately 1 μm and only non-refractory species (i.e. ammonium, sulphate, nitrate, chloride and organics) evaporate at the vaporiser temperature (~600°C), the HR-ToF-AMS measurements represent non-refractory PM₁ (NR-PM₁). Details of the AMS are presented in previous publications [19]. Compared with the previous AMS versions (i.e. Q-AMS, C-ToF-AMS), HR-ToF-AMS has higher resolutions and offers a high m/z resolution of approximately 4000–5000, allowing a greatly improved identification of adjacent ions that are slightly different in exact mass due to differences in elemental composition.

(iii) HR-ToF-AMS operation and calibration. According to the ion flight trajectories in the ToF mass spectrometer, the HR-ToF-AMS provided data with two different resolutions (V-Mode and W-Mode). The more sensitive V-mode data are used to generate unit mass resolution mass spectra, from which mass concentrations and size distributions of species are determined. The high mass resolution W mode is used to separate ion fragments that have the same nominal m/z but differing elemental compositions. During the observation, the HR-ToF-AMS was operated under the V-mode, and the detected period was set to 5 min. The AMS cycled through the mass spectrum (MS) mode and the particle time-of-flight (PToF) mode every 20 s, spending 10 s in each mode. The instrument was calibrated for ionisation efficiency (IE) and particle sizing at the beginning and in the middle of this study following the standard protocols [20]. The calibration of IE used size-selected pure ammonium nitrate particles selected by a Differential Mobility Analyzer (DMA, TSI Co., USA) before introduction into the AMS. The particle size calibration was conducted using mono-disperse polystyrene latex spheres (PSL, density = 1.05 g/cm³) (Duke Scientific, Palo Alto, California, USA) with nominal diameters of 100–700 nm [21]. The HR-ToF-AMS detection limits of the different species were determined by filtered particle-free ambient air and defined as three times the standard deviations of the corresponding species signals. The detection limits (DLs) of the NR-PM₁

species were determined as 3 times the standard deviations (3σ) of the corresponding signals in particle-free ambient air through a HEPA filter. The 5-min DL of organics during the campaign was $0.056 \mu\text{g}/\text{m}^3$.

(iv) HR-ToF-AMS data analysis. Standard ToF-AMS data analysis software packages (SQUIRREL version 1.50 and PIKA version 1.08) were used to obtain the information about mass concentration, size distribution and elemental composition of organics [22]. According to previous field studies, a collection efficiency (CE) factor of 0.5 was used to account for the incomplete detection of aerosol species due to particle bounce at the vaporiser and/or the partial transmission of particles by the lens [19]. The relative ionisation efficiency (RIE) values used in this study were 4.0 for ammonium, 1.2 for sulphate, 1.1 for nitrate, 1.3 for chloride and 1.4 for organics [20].

2 Results and discussions

2.1 Mass concentrations and temporal variations

Figure 1 shows the time variations of organic aerosol mass concentrations and the meteorological parameters (i.e. temperature, humidity, wind speed and direction and air pressure) at the sampling site from 14 December 2010 to 15 January 2011. The weather conditions during this study were characterised by low temperature, low humidity, and prevailing northerly winds. The average temperature, humidity and wind speed were -2.4°C (hourly average -11.2 – 13°C), 23% (hourly average 10%–57%) and 2.3 m/s (hourly

average 0–9.4 m/s), respectively.

As Figure 1 shows, the time variation of organic aerosol displayed strong asymmetric “sawtooth cycles” characteristic of Beijing winters. The cycles begin after a cold front, rising progressively in approximately 4 days and then falling rapidly with the next cold front. The sawtooths are consistent with regional accumulation and removal processes, which were controlled by synoptic cycles, specifically the passage of cold fronts [23]. During the entire campaign, the concentrations of organic aerosol varied dynamically (5-min average of 1.9 – $284.6 \mu\text{g}/\text{m}^3$), with an average of $20.9 \mu\text{g}/\text{m}^3$ and a standard deviation of $25.3 \mu\text{g}/\text{m}^3$, which is lower than that measured in urban Beijing in the summers of 2006 ($28.1 \mu\text{g}/\text{m}^3$) [13] and 2008 ($23.9 \mu\text{g}/\text{m}^3$) [12]. Many previous studies reported that low ambient aerosol loading periods typically occurred with northerly air masses, so the prevailing northerly wind may mainly cause the relatively low OA mass concentrations in most observation periods [24]. In addition, from December 17 to 21, 2010, Beijing city experienced a severe pollution period. The OA mean mass concentration reached $56.1 \mu\text{g}/\text{m}^3$ and a standard deviation of $35.5 \mu\text{g}/\text{m}^3$. As the synchronous meteorological conditions in this period showed, the wind speed was relatively low (less than 1 m/s), the temperature increased and the air pressure continuously remained at a low level. All of these factors indicated that the synoptic system of Beijing in this period was controlled by warm low air pressure. In this case, a strong temperature inversion at low altitudes was often accompanied by a highly stable atmospheric layer. This process made the ground convergence stronger, and

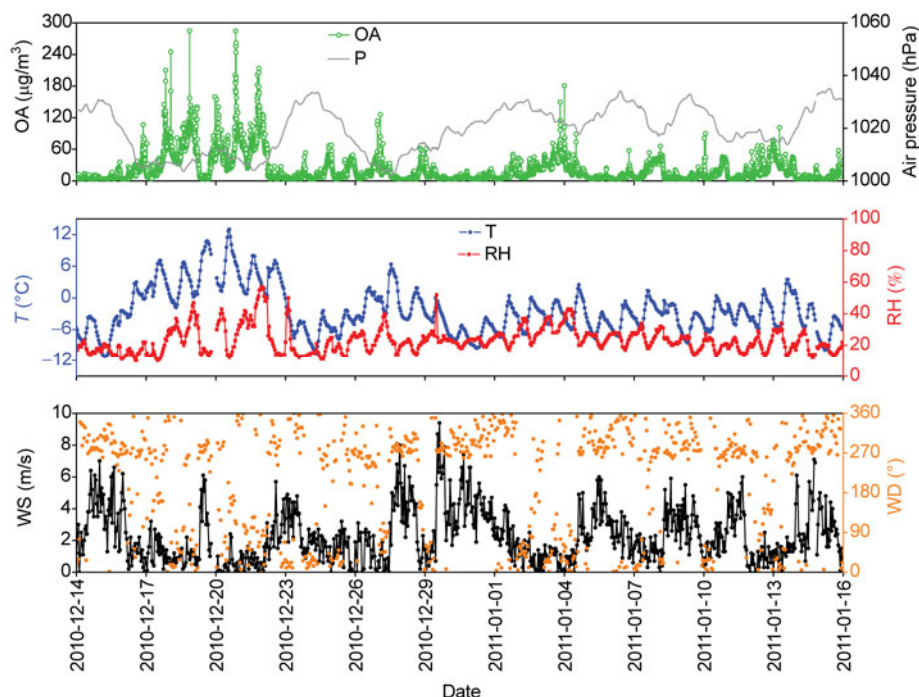


Figure 1 Time series of mass concentration of OA, air pressure (P), temperature (T), relative humidity (RH), wind speed (WS) and wind direction (WD).

not only could the pollutants not be diffused easily but also the atmospheric pollutants of the region circumjacent to Beijing gathered towards the centre, thereby aggravating the degree of air pollution in the city [25]. Meanwhile, the relatively high humidity in this period caused the moisture content in the particles to increase, which subsequently enhanced the absorption capacity of the particles for semi-volatile and volatile organic matter, which ultimately led to the high OA mass concentrations [26,27].

The diurnal variation of OA shows two peaks (Figure 2), a small peak in the noon hour (12:00–13:00) and another large peak in the evening (20:00–21:00). This characteristic is generally similar to the results in Beijing summer [12], but different from some cities abroad, which have a significant peak due to rush hour traffic in the morning (i.e. Pittsburgh and New York City) [28,29]. This unique variation was likely related to the combined influence from local emissions, secondary production, transport and the evolution of boundary layer height. The identification and description of sources and variation characteristics of OA will be discussed in Section 2.4.

2.2 Size distribution

Figure 3 presents the OA, m/z 44 and m/z 57 average size distributions determined by AMS during the campaign. The OA showed a major accumulation mode peaking at a vacuum aerodynamic diameter (D_{va}) of approximately 450 nm, and it also had a significant contribution of ultrafine particles ($D_{va} < 100$ nm). In the AMS mass spectrum, fragment m/z 57 (mostly $C_4H_5^+$) indicated a strong association with hydrocarbon organic aerosols (HOA) and is usually a tracer of HOA [30]; however, m/z 44 (mostly CO_2^+) presented a good correlation with oxygenated organic aerosols (OOA) and could be a tracer of OOA [31,32]. As shown in Figure 3, the size distribution of m/z 57 was much broader than that of m/z 44, and its mass concentration was distinctly much

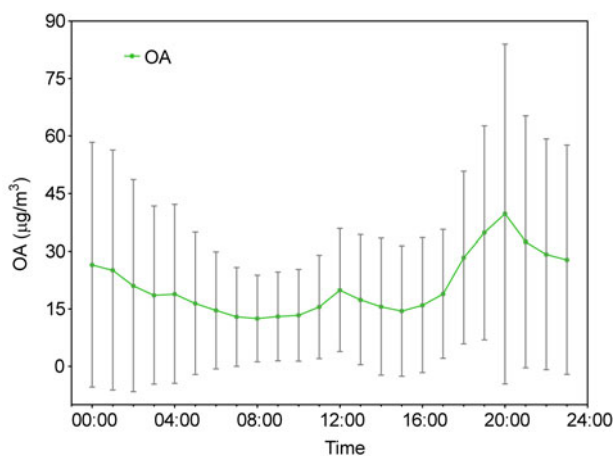


Figure 2 Diurnal cycle of OA mass concentration during the observation period.

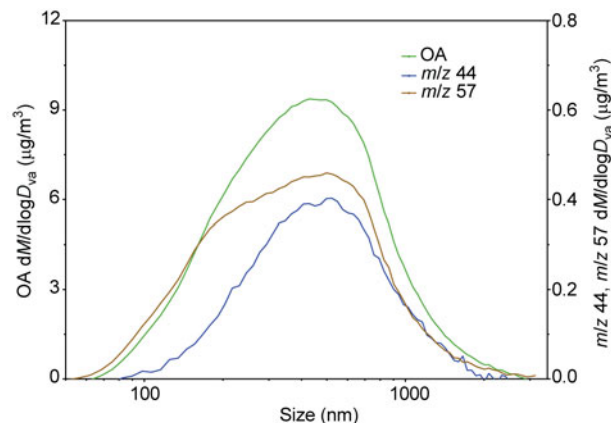


Figure 3 The average size distributions of OA, m/z 44 and m/z 57 for entire study.

higher. In general, the HOA linked to m/z 57 was the major contributor of ultrafine OA, indicated the ultrafine OA in Beijing winter predominately came from “fresh” combustion emissions [33,34].

2.3 Elemental compositions

The OA elemental information was analysed by high-resolution mass spectrum data analysis software (PIKA version 1.08). The average molar percentages of the elemental compositions were 34.8% C, 59.1% H, 0.2% N and 5.9% O, corresponding to a mass contribution rate of 72.7%, 10.4%, 0.4% and 16.5%, respectively. Many studies showed that organic aerosols from different sources had significantly varied O/C (molar ratio) values. In general, primary organic aerosols (POA) had lower O/C ratios than those of secondary organic aerosols (SOA), such as vehicle emissions (O/C range 0.03–0.04), cooking-related emissions (O/C range 0.11–0.14) and aged SOA (O/C range 0.3–1.0) [33–35]. Figure 4 shows the time trend of H/C (molar ratio), O/C (molar ratio), N/C (molar ratio) and OM/OC (mass ratio of organic matter to organic carbon). During this study, the average H/C, O/C and N/C were 1.70, 0.17, and 0.005, respectively, corresponding to an OM/OC ratio of 1.37. Compared to the observation results in 2008 summer in Beijing (H/C 1.48, O/C 0.37, and OM/OC 1.64) [36], the H/C was slightly higher but the O/C and OM/OC were much lower. This result may be due to coal combustion for residential heating and adverse meteorological conditions (i.e. low temperature and low humidity) for secondary organic aerosol formation. The differences of OM/OC between summer and winter indicate that this value should vary with different seasons and observation sites. Therefore, for studies of the chemical compositions of aerosols based on filter sampling, the proper ratios should be chosen when calculating the organic matter mass concentration according to the organic carbon.

As shown in Figure 5, the diurnal variation of O/C is

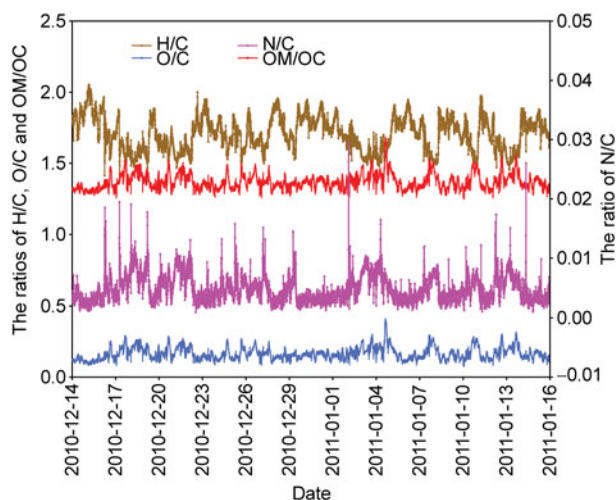


Figure 4 Time series of H/C (molar ratio), N/C (molar ratio), O/C (molar ratio) and OM/OC (mass ratio).

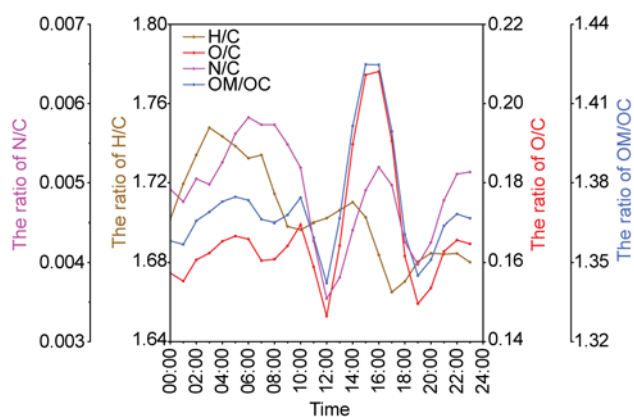


Figure 5 Diurnal cycles of H/C, N/C, O/C and OM/OC during the observation period.

consistent with that of OM/OC and has two main features: (1) the O/C and OM/OC ratios both increase rapidly from noon (~12:00) to late afternoon (~15:00) due to enhanced photochemical SOA formation; and (2) two significant low values of O/C and OM/OC, which appear at 12:00 and 19:00 according to lunch and dinner times, indicate that cooking-related emissions have an important influence on those values. In addition, the H/C ratio was mainly controlled by the variations of boundary layer height and local primary emission, presenting high values at night and low values during the day. In other words, the H/C ratio exhibited a phase relation opposite to the O/C ratio.

2.4 OA components and their sources

A high time resolution is highly favourable to PMF analysis. To further identify the OA chemical compositions and sources, the PMF model was applied to analyse the organic mass spectra data [37,38]. Finally, a three-factor solution was chosen as the optimal solution. According to the char-

acteristics of these factors (such as their mass spectrum features, diurnal variations and correlations with inorganic components), three distinct components, including hydrocarbon-like OA (HOA), cooking-related OA (COA) and oxygenated OA (OOA), were determined.

Figure 6 shows the mass profiles and diurnal variations of the three OA components. The mass spectrum (MS) of HOA was dominated by peaks characteristic of hydrocarbons ($C_nH_{2n+1}^+$), such as m/z 29 (mostly $C_2H_5^+$), m/z 43 ($C_3H_7^+$) and m/z 57 ($C_4H_9^+$). This result is very similar to previously reported reference spectra of POA emitted from gasoline and diesel combustion sources [39]. In addition, the HOA correlated well with the primary gas pollutants NO_x ($R^2=0.66$) and CO ($R^2=0.74$), which were emitted from combustion processes. Compared with the inorganic components, the HOA had a strong correlation with chloride ($R^2=0.86$) (measured by HR-ToF-AMS), which indicated they had a good homology. Because NaCl could not be measured at the vaporiser temperature used in this study, the detected chlorides were mainly in the semi-volatile form of NH_4Cl . This particular formation of chlorides originated from a secondary production of gas-phase HCl emitted by heating combustion sources in the Beijing winter [40]. It is useful to note that biomass burning, a known primary source, significantly contributed to the HOA because there was distinct m/z 60, a tracer ion fragment for biomass burning-emitted aerosols in the HOA MS. The diurnal pattern of HOA concentrations showed the highest values in the late evening and the lowest concentrations in the afternoon. The lower concentrations in the afternoon were a result of the high daytime PBL and reduced heavy-duty diesel truck emissions during the day. Due to Beijing traffic regulations, heavy-duty diesel truck traffic is several times higher at night than during the day. Combined with low PBL at night, a significant peak appeared during the night. A small morning HOA peak was also observed in this campaign and could be mainly attributed to the morning rush hour traffic. The above analysis indicates that the HOA component in Beijing winter was mainly from the primary sources of traffic emission: coal combustion for heating and biomass burning.

The MS of COA is characterised by the most prominent unsaturated ions ($C_nH_{2n-1}^+$), such as m/z 41 (mainly $C_3H_5^+$) and m/z 55 (mainly $C_4H_7^+$), which is consistent with the MS characteristics measured for primary Chinese cooking emissions [41]. The weak correlations between COA and other inorganic components during the observation period (i.e. sulphate, nitrate, ammonium and chloride) indicated that the sources of COA were relatively independent. A clear and unique diurnal pattern of COA provides another piece of strong evidence for it being cooking-related: it presented a small peak at noon and a large peak in the evening, in accordance with the lunch and dinner times of the local residents. Due to the unique Chinese cooking habits and culture, cooking emissions are regarded as one of the major organic

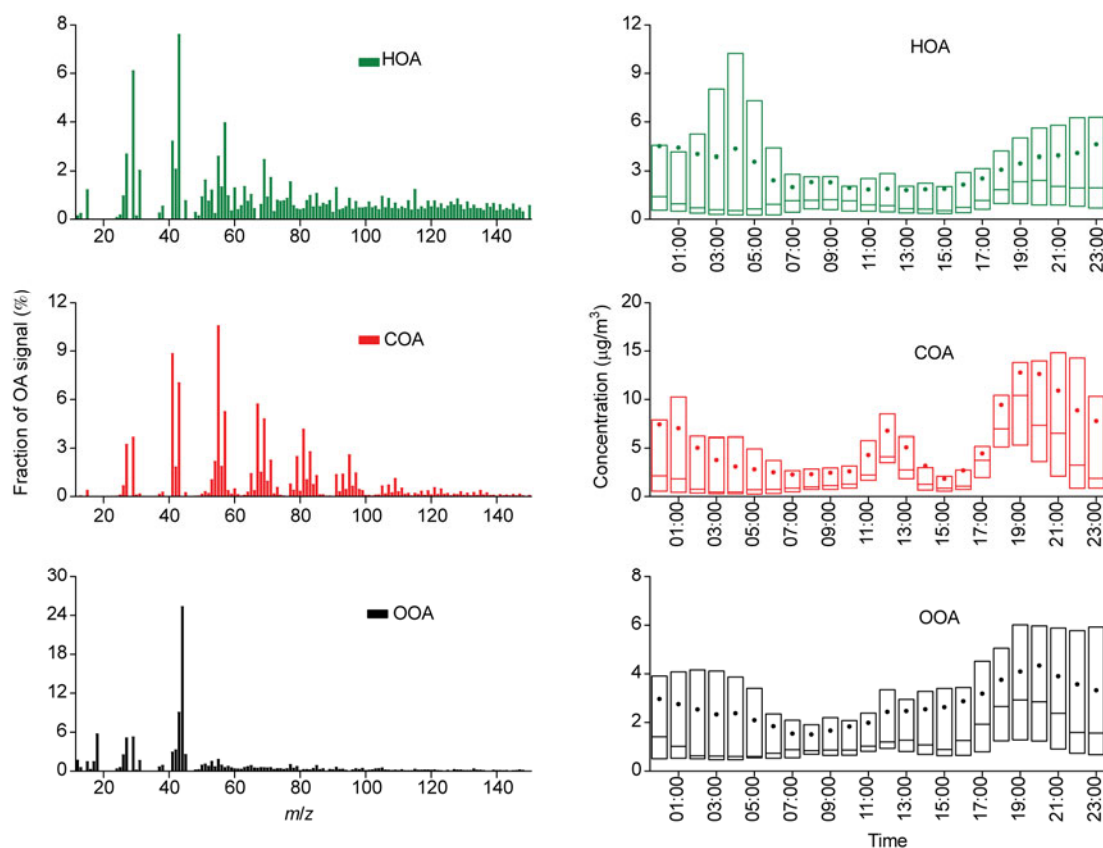


Figure 6 Mass spectrum (left) and time series (right) of OA components during the observation period.

aerosol sources in Chinese urban environments [42].

The OOA component is mainly generated by the oxygenated and photochemical processes of its precursors. The MS of this component is characterised by a dominant peak at m/z 44 (mainly CO_2^+) and does not contain significant contributions from marker peaks of other organic aerosol components, such as at m/z 57 or m/z 60 (Figure 6). The CO_2^+ ions, which are mainly from di- and poly-carboxylic acid functional groups, represent a tracer for oxygenated organic aerosol components [33,34]. In addition, the OOA component correlated well with the sum of sulphate and nitrate ($R^2=0.91$) and showed a similar diurnal pattern with those well-known secondary inorganic species. The diurnal cycle of OOA was characterised by a gradual increase in concentration from approximately 10:00 to 19:00, which was effected by photochemical processes. Two types of OOA with different volatilities have been observed in many previous studies: one is the more oxidised and aged low-volatility OOA and the other is the less oxidised and fresher semi-volatile OOA [12,13,28]. The mass spectrum of OOA in this study is very similar to that of low-volatility OOA, and the semi-volatile OOA did not feature in any of the solutions derived here. That is not to say that semi-volatility OOA is not present but rather that it is not possible to distinguish it from the low-volatility OOA; thus, the fraction is reported as a single factor.

On average, the HOA, COA and OOA components accounted for 26.9%, 49.7% and 23.4% of the total organic mass during this campaign, respectively. The sum of HOA and COA could be regarded as POA, and the OOA corresponded to aged SOA. Subsequently, the POA and SOA accounted 76.6% and 23.4% of the total organic mass, respectively, which indicates that the OA was mainly from the local POA emissions in Beijing winter and that the meteorological conditions of low temperature and low humidity were unfavourable to the formation of SOA. In addition, the large percentage of COA in OA (~50%) indicates its key role in air pollution control.

3 Conclusions

The size-resolved organic matter of submicron aerosols was measured *in situ* using a HR-ToF-AMS in Beijing in the winter of 2010. The mass concentrations, chemical compositions and size distribution of OA were obtained with a high time resolution. The mean OA mass concentration was $20.9 \pm 25.3 \mu\text{g}/\text{m}^3$ during this campaign, varying between 1.9 and $284.6 \mu\text{g}/\text{m}^3$. The average mass-based size distributions of OA present a prominent accumulation mode peaking at approximately 450 nm. Meanwhile, through the analysis of marker ions at m/z 44 and 57, it could be derived that the

prominent presence of ultrafine particles ($D_{va} < 100$ nm) was mainly from the fresh emissions of combustion sources. During this study, the average H/C, O/C and N/C (molar ratio) were 1.70, 0.17, and 0.005, respectively, corresponding to an OM/OC ratio (mass ratio of organic matter to organic carbon) of 1.37. The diurnal pattern of O/C was consistent with that of OM/OC and displayed a typical photochemical characteristic.

Positive Matrix Factorisation (PMF) analysis of the organic mass spectral dataset differentiated the OA into three components: hydrocarbon-like (HOA), cooking-related (COA), and oxygenated (OOA) organic aerosols, which, on average, accounted for 26.9%, 49.7% and 23.4%, respectively. In addition, the POA is a dominant component of OA in Beijing winter, with a large percentage of 76.6%. The large contribution of COA in the POA mass indicates its key role in PM_{10} pollution control in Beijing.

- 1 Fenger J. Urban air quality. *Atmos Environ*, 1999, 33: 4877–4900
- 2 Pope C A, Dockery D W. Health effects of fine particulate air pollution: lines that connect. *J Air Waste Manage Assoc*, 2006, 56: 709–742
- 3 Vecchi R, Marcazzan G, Valli G, et al. The role of atmospheric dispersion in the seasonal variation of PM_{10} and $PM_{2.5}$ concentration and composition in the urban area of Milan (Italy). *Atmos Environ*, 2004, 38: 4437–4446
- 4 Zhang L, Ninomiya Y, Yamashita T. Formation of submicron particulate matter (PM_{10}) during coal combustion and influence of reaction temperature. *Fuel*, 2006, 85: 1446–1457
- 5 Schwartz J, Dockery D W, Neas L M. Is daily mortality associated specifically with fine particles? *J Air Waste Manage Assoc*, 1998, 46: 927–939
- 6 Zhang Q, Jimenez J L, Canagaratna M R, et al. Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. *Geophys Res Lett*, 2007, 34, doi: 10.1029/2007GL029979
- 7 Molina M J, Molina L T. Megacities and atmospheric pollution. *J Air Waste Manage Assoc*, 2004, 54: 644–680
- 8 Zhang Q H, Zhang J P, Xue H W. The challenge of improving visibility in Beijing. *Atmos Chem Phys*, 2010, 10: 7821–7827
- 9 Chan C K, Yao X. Air pollution in mega cities in China. *Atmos Environ*, 2008, 42: 1–42
- 10 Duan F K, Liu X D, Yu T, et al. Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing. *Atmos Environ*, 2004, 38: 1275–1282
- 11 He L Y, Hu M, Huang X F, et al. Seasonal pollution characteristics of organic compounds in atmospheric fine particles in Beijing. *Sci Total Environ*, 2006, 359: 167–176
- 12 Huang X F, He L Y, Hu M, et al. Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer. *Atmos Chem Phys*, 2010, 10: 8933–8945
- 13 Sun J Y, Zhang Q, Canagaratna M, et al. Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer. *Atmos Environ*, 2010, 44: 131–140
- 14 Takegawa N, Miyakawa T, Kuwata M, et al. Variability of submicron aerosol observed at a rural site in Beijing in the summer of 2006. *J Geophys Res*, 2009, 114, doi: 10.1029/2008JD010857
- 15 Huang X F, He L Y, Hu M, et al. Annual variation of particulate organic compounds in $PM_{2.5}$ in the urban atmosphere of Beijing. *Atmos Environ*, 2006, 40: 2449–2458
- 16 He K B, Yang F M, Ma Y L, et al. The characteristics of $PM_{2.5}$ in Beijing, China. *Atmos Environ*, 2001, 35: 4959–4970
- 17 Hu B, Wang Y S, He X X, et al. Variation properties of earth's surface solar radiation during a strong dust storm in Beijing 2004 (in Chinese). *Clim Environ Res*, 2005, 10: 265–274
- 18 Xin J Y, Wang Y S, Tang G Q, et al. Variability and reduction of atmospheric pollutants in Beijing and its surrounding area during the Beijing 2008 Olympic Games. *Chin Sci Bull*, 2010, 55: 1937–1944
- 19 Canagaratna M R, Jayne J T, Jimenez J L, et al. Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer. *Mass Spectrom Rev*, 2007, 26: 185–222
- 20 Jimenez J L, Jayne J T, Shi Q, et al. Ambient aerosol sampling using the Aerodyne Aerosol Mass Spectrometer. *J Geophys Res*, 2003, 108, doi: 10.1029/2001JD001213
- 21 DeCarlo P F, Kimmel J R, Trimborn A, et al. Field-deployable, High-Resolution, Time-of-Flight Aerosol Mass Spectrometer. *Anal Chem*, 2006, 78: 8281–8289
- 22 Aiken A C, DeCarlo P F, Jimenez J L. elemental analysis of organic species with electron ionization high-resolution mass spectrometry. *Anal Chem*, 2007, 79: 8350–8358
- 23 Jia Y T, Rahn K A, He K B, et al. A novel technique for quantifying the regional component of urban aerosol solely from its sawtooth cycles. *J Geophys Res*, 2008, 113, doi: 10.1029/2008JD010389
- 24 Ma F M, Gao Q X, Zhou S Q, et al. The simulation and analysis of a typical pollution event around Beijing (in Chinese). *Res Environ Sci*, 2008, 21: 30–36
- 25 Sun Y, Wang Y S, Changchun Z. Measurement of the vertical profile of atmospheric SO_2 during the heating period in Beijing on days of high air pollution. *Atmos Environ*, 2009, 43: 468–472
- 26 Jang M, Czoschke N M, Lee S, et al. Heterogeneous atmospheric aerosol production by acid-catalyzed particle-phase reactions. *Science*, 2002, 298: 814–817
- 27 Jacob D J, Winner D A. Effect of climate change on air quality. *Atmos Environ*, 2009, 43: 51–63
- 28 Sun Y L, Zhang Q, Schwab J J, et al. Characterization of the sources and processes of organic and inorganic aerosols in New York City with a high-resolution time-of-flight aerosol mass spectrometer. *Atmos Chem Phys*, 2011, 11: 1581–1602
- 29 Zhang Q, Canagaratna M R, Jayne J T, et al. Time- and size-resolved chemical composition of submicron particles in Pittsburgh: Implications for aerosol sources and processes. *J Geophys Res*, 2005, 110, doi: 10.1029/2004JD004649
- 30 Aiken A C, DeCarlo P F, Kroll J H, et al. O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry. *Environ Sci Technol*, 2008, 42: 4478–4485
- 31 Alfara M R, Coe H, Allan J D, et al. Characterization of urban and rural organic particulate in the Lower Fraser Valley using two Aerodyne Aerosol Mass Spectrometers. *Atmos Environ*, 2004, 38: 5745–5758
- 32 Bahreini R, Keywood M D, Ng N L, et al. Measurements of secondary organic aerosol from oxidation of cycloalkenes, terpenes, and *m*-xylene using an Aerodyne Aerosol Mass Spectrometer. *Environ Sci Technol*, 2005, 39: 5674–5688
- 33 Jimenez J L, Canagaratna M R, Donahue N M, et al. Evolution of organic aerosols in the atmosphere. *Science*, 2009, 326: 1525–1529
- 34 Ng N L, Canagaratna M R, Zhang Q, et al. Organic aerosol components observed in Northern Hemisphere datasets from Aerosol Mass Spectrometry. *Atmos Chem Phys*, 2010, 10: 4625–4641
- 35 Mohr C, Huffman J A, Cubison M J, et al. Characterization of primary organic aerosol emissions from meat cooking, trash burning, and motor vehicles with high-resolution aerosol mass spectrometry and comparison with ambient and chamber observations. *Environ Sci Technol*, 2009, 43: 2443–2449
- 36 Huang X F, Xue L Y, He L, et al. On-line measurement of organic aerosol elemental composition based on high resolution aerosol mass spectrometry (in Chinese). *Chin Sci Bull (Chin ver)*, 2010, 55: 3391–3396
- 37 Ulbrich I M, Canagaratna M R, Zhang Q, et al. Interpretation of organic components from Positive Matrix Factorization of aerosol

- mass spectrometric data. *Atmos Chem Phys*, 2009, 9: 2891–2918
- 38 Paatero P, Tapper U. Positive matrix factorization: A non-negative factor model with optimal utilization of error estimates of data values. *Environmetrics*, 1994, 5: 111–126
- 39 Schneider J, Weimer S, Drewnick F, et al. Mass spectrometric analysis and aerodynamic properties of various types of combustion-related aerosol particles. *Int J Mass spectrom*, 2006, 258: 37–49
- 40 Hagino H, Takada T, Kunimi H, et al. Characterization and source presumption of wintertime submicron organic aerosols at Saitama, Japan, using the Aerodyne aerosol mass spectrometer. *Atmos Environ*, 2007, 41: 8834–8845
- 41 He L Y, Lin Y, Huang X F, et al. Characterization of high-resolution aerosol mass spectra of primary organic aerosol emissions from Chinese cooking and biomass burning. *Atmos Chem Phys Discuss*, 2010, 10: 21237–21257
- 42 Zhao Y L, Hu M, Slanina S, et al. Chemical compositions of fine particulate organic matter emitted from Chinese cooking. *Environ Sci Technol*, 2006, 41: 99–105

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