Aging impact on sources, volatility, and viscosity of organic aerosols in the Chinese outflows

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Abstract. To investigate the aging impact on sources, volatility, and viscosity of organic aerosol (OA) in the Chinese outflows, a high-resolution time-of-flight aerosol mass spectrometer (HR-AMS) coupled with a thermodenuder (TD) was deployed in the spring of 2018 in Dongying, which is a regional receptor site of metropolitan emissions in North China Plain (NCP). The average mass concentration of PM_1 is 31.5 ± 22.7 μg m^{-3}, which is mainly composed of nitrate (33 %) and OA (25 %). The source apportionment results show that the OA is mainly contributed by oxygenated OA (OOA) from secondary sources, including background-OOA (33 %) representing a background concentration of OA (2.6 μg m^{-3}) in the NCP area, and transported-OOA (33 %) oxidizing from urban emissions. The other two factors include aged hydrocarbon-liked OA (aged-HOA, 28 %) from transported vehicle emissions and biomass burning OA (BBOA, 5 %) from local open burnings. The volatility of total OA (average $C^* = 3.2\times10^{-4}$ μg m^{-3}) in this study is generally lower than those in previous field studies, which is mainly due to the high OA oxidation level resulting from aging processes during transport. The volatilities of OA factors follow the order of background-OOA (average $C^* = 2.7\times10^{-5}$ μg m^{-3}) < transported-OOA (3.7×10^{-4} μg m^{-3}) < aged-HOA (8.1×10^{-4} μg m^{-3}) < BBOA (0.012 μg m^{-3}). Extremely low volatilities in the ambient air indicate that oligomers may exist in the aged plumes. The viscosity estimation suggests that the majority of ambient OA in this study behaves as semi-solid (60 %), liquifies at higher RH (21 %), and solidifies (19 %) during noon time when the RH is low and the oxidation level is high. Finally, the estimated mixing time of molecules in 200nm OA varies dramatically from minutes at night to years in the afternoon, emphasizing the necessity to consider its dynamic kinetic limits when modeling OA. In general, the overall results of this study improve the understanding of the aging impact on OA volatility and viscosity.

1 Introduction

Atmospheric fine particles, which can substantially affect visibility, human health, and climate, have drawn great attention of the public (Song et al., 2018; Quan et al., 2011; Horowitz and Jacob, 1999; Huang et al., 2014; Li et al., 2020b; Huffman et al., 2009a). In the NCP areas, which is one of the most polluted areas in China, high concentrations of fine particles (21–178 μg m^{-3}) in urban cities have been frequently observed (Zhou et al., 2020; Zhao et al., 2019; Duan et al., 2020; Xu et al., 2019). The city clusters in NCP areas act as urban emission sources and have a great impact on the atmospheric chemistry in the downwind regions. During the transport processes, the pollutants undergo physical and chemical reactions continuously, which significantly change the properties of aerosols, especially for organic aerosol (OA), the dominating component of fine particles (Sun et al., 2017; Zhang et al., 2012; Jimenez et al., 2009; Zhang et al., 2007). In the past decade, plenty of field measurements have been conducted to explore the concentration variations and sources of OA in urban and suburban areas in NCP (Duan et al., 2020; Zhang et al., 2018; Sun et al., 2013; Li et al., 2017a; Hu et al., 2016b; Elser et al., 2016; Hu et al., 2017a; Huang et al., 2010; Jiang et al., 2015; Wang et al., 2015; Xu et al., 2018; Xu et al., 2015; Zhang et al., 2015; Zhang et al., 2016; Zhao et al., 2017; Zhou et al., 2018 and references therein), however, few studies were carried out in background areas (Zhou et al., 2020; Zhang et al., 2020; Zheng et al., 2015; Yan et al., 2021; Du et al., 2015). Studies which focus on the physicochemical properties of OA in background areas, i.e., volatility and viscosity, are even scarce. The volatility and viscosity of OA can determine the gas-particle partitioning and particle phases, which are crucial information to understanding aerosol growth and heterogeneous reactions in bulk OA (Li et al., 2020b; Reid et al., 2018; Shiraiwa et al., 2012). Meanwhile, quantifications of volatility and viscosity of OA in aged plumes are the key information to understand the fate of aerosols in the atmosphere.

Multiple methods have been used to quantify the volatility of OA (Isaacman-Vanwertz and Aumont, 2021; Cappa and Jimenez, 2010; Saha et al., 2015; Li et al., 2016b; Yli-Juuti et al., 2017; Louvaris et al., 2017a). One of the most common methods is combining a heating module (e.g., thermodenuder, TD) with a detection instrument (e.g., aerosol mass spectrometer, AMS) to measure the mass fraction remaining (MFR) of different OA species at various temperatures. The volatility distribution of OA can be estimated based on either an empirical method (Faulhaber et al., 2009) or an evaporation kinetic model (Riipinen et al., 2010; Saha et al., 2015; Cappa, 2010a; Karnezi et al., 2014; Saha et al., 2017; Park et al., 2012; Epstein et al., 2009; Louvaris et al., 2017a). Based on the TD-AMS system, volatilities of ambient OA from urban/suburban (Xu et al., 2021; Louvaris et al., 2017b; Cao et al., 2018; Huffman et al., 2009a; Cappa and Jimenez, 2010; Saha et al., 2018; Paciga et al., 2016; Kang et al., 2022), rural (Zhu et al., 2021) and forest sites (Hu et al., 2016c; Saha et al., 2017; Kostenidou et al., 2018) have been explored. Huffman et al. (2009a) first directly characterized chemically-resolved volatility of ambient aerosols in urban areas, supporting that OA components are semivolatile and POA (primary organic aerosol) is more volatile than SOA (secondary organic aerosol). Using the similar approach, multiple studies (Huffman et al., 2009a; Louvaris et al., 2017b; Xu et al., 2016; Xu et al., 2021; Zhu et al., 2021) found that the OA volatility varies with different emission sources and oxidation levels, usually shows a trend of vehicle OA > biomass burning OA ≈ cooking OA > secondary OA. In addition, extremely low volatility of SOA due to the existence of oligomers after oxidation has also been reported (Lopez-Hilfiker et al., 2016; Hu et al., 2016c). The varied volatilities of POA and SOA emphasize the necessity to investigate OA volatility during aging processes.

Viscosity, which can be influenced by ambient relative humidity (RH), molecular weight, and oxidation level of OA, determines the mixing state and phase state of aerosols (Koop et al., 2011; Shiraiwa et al., 2017; DeRieux et al., 2018). Multiple studies in the field campaigns and laboratory experiments suggest that, contrary to the traditional understanding, OA may occur as highly viscous semisolids or amorphous glassy solids under certain conditions (Shiraiwa et al., 2017; Virtanen et al., 2010; Reid et al., 2018; Koop et al., 2011; Renbaum-Wolff et al., 2013), which shows important implications on atmospheric chemistry. Diffusion timescales within aerosols are affected by the aerosol size, mass loading, viscosity, and volatility (Li and Shiraiwa, 2019). The diffusivity of species in aerosol is negatively correlated with viscosity. Thereby it takes a longer time for highly viscous species to reach gas-particle equilibrium, which affects a series of atmospheric processes of aerosols (Shiraiwa and Seinfeld, 2012). A new parameterization proposed by Li et al. (2020b) has been used to estimate the diurnal variations of

OA viscosity in ambient air based on the correlation between volatility and viscosity (Li et al., 2020b; Xu et al., 2021). The results show that OA viscosity can differ more than 4 orders of magnitude between day and night, as well as in the daytime among seasons. The great variation signifies the sensitivity of OA viscosity to ambient conditions.

To investigate OA sources, volatility, and viscosity in the outflow of urban plumes, a TD-AMS system was applied in a ground-based campaign at a continental receptor site of downwind areas in NCP. We systematically investigated the dynamic variations of chemical compositions of fine particles. OA sources were analyzed using Positive matrix factorization (PMF) with Multi-Engine (ME-2) analysis. Coupled with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, the impact of regional transport on aerosols was further studied. Finally, a comprehensive understanding of volatilities and viscosities of OA and OA factors during the aging process, as well as their atmospheric implications, are shown and discussed.

2 Experiments

2.1 Field campaign

The field study was conducted at Yellow River Delta National Nature Reserve (YRDNNR) from March 17 to April 12, 2018. The YRDNNR site (37.75° N, 118.97° E, 1 m above sea level) is located in the northeast of Dongying City, Shandong Province, and the estuary of the Yellow River into the Bohai Sea (Yao et al., 2016; Zheng et al., 2019; Zhang et al., 2019), as shown in Fig. 1. Very few industrial or commercial areas were within 15 kilometers of the sampling site, which is considered as a regional background site of NCP. The impacts of natural oil and gas (NO&G) (i.e. Liquified Petroleum Gas, LPG), biomass burning on volatile organic compounds (VOCs), and O_3 during the campaign were found at the site (Lee et al., 2021). The wind rose diagram combined with back trajectory clusters in Fig. 1a suggests that the sampling site during this campaign was affected by southern, southeast, and northwest winds, indicating the observed results were strongly influenced by the NCP urban outflows. The average wind speed was 3.0 ± 1.8 m s^{-1} with a range of 0–9.1 m s^{-1}. The average temperature during this study was 11.5 ± 7.0 ℃ (from −2.8 to 29.2 ℃) and RH was 59.1 ± 19.0 % (from 14 to 97 %).

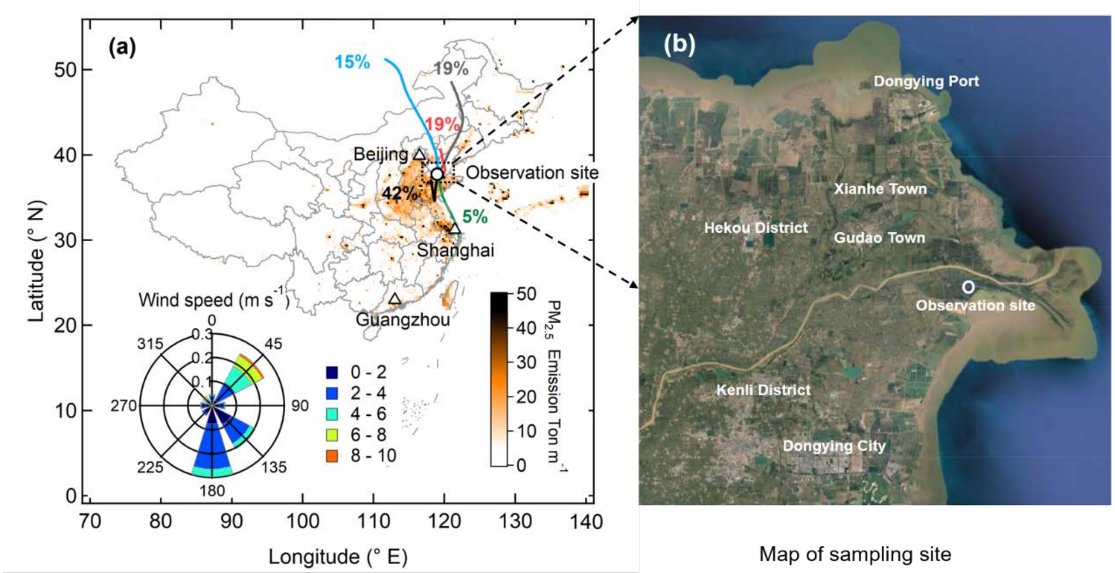

Figure 1. (a) Location of observation site (opaque circle) on the Chinese map, along with wind speeds, wind directions, and back trajectories of air masses during this campaign. The map is color-coded by $PM_{2.5}$ emission inventory (Li et al., 2017b) (b) Map of the Yellow River Delta National Nature Reserve (YRDNNR) region, labeled with the sampling site (opaque circle) and adjacent cities and towns (source: Google Earth).

2.2 Operations and data processing of instruments

2.2.1 HR-ToF-AMS

The chemical composition of non-refractory PM_1 was measured by a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, manufactured by Aerodyne Research Inc. ARI, USA; "AMS" will be used hereinafter). The detailed introduction and operating principle of AMS can be found in previous studies (Canagaratna et al., 2007; Drewnick et al., 2005; Chen et al., 2021b). The setup and calibrations of AMS will be briefly introduced in the following.

During this campaign, the ambient air was sampled through a $PM_{2.5}$ cyclone at a flow rate of 5 L min^{-1} and then introduced into AMS through a 1/4-inch stainless steel tube. The sampling flow rate of AMS was 0.1 L min^{-1}. To avoid the effects of varied phase states on the collection efficiency (CE) of particles, a silicon dryer was set ahead of AMS to keep the RH of sampled air below 40 %. The schematic diagram is shown in Fig. S1.

The calibrations including ionization efficiency (IE), inlet flow rate, and sizing distribution, were conducted before and after the campaign (Jimenez et al., 2003; Jayne et al., 2000; Canagaratna et al., 2007). The IE calibration was performed based on a brute-force single particle (BFSP) mode (Decarlo et al., 2006), in which 400 nm monodisperse pure ammonium nitrate (NH_4NO_3) particles were introduced. For mass concentration calculation, the relative ionization efficiency (RIE) of ammonium was determined to be 3.7 by assuming a neutral balance of cation versus anion from standard NH_4NO_3 signals. The default RIE values of nitrate, sulfate, chloride, and OA were applied in this study (Huffman et al., 2009a). Taking into account particle loss in AMS, the chemical composition-dependent CE (CDCE) followed by Middlebrook et al. (2012) was applied to correct mass concentration. The average value of CDCE is 0.52 ± 0.04.

Pieber et al. (2016) found that the artificial CO_2^+ ion can be caused by inorganic salts due to the interaction between thermal decomposition products of inorganic salts (i.e., NH_4NO_3) and OA deposit on the vaporizer surface. The ratio of CO_2^+/NO_3^- was estimated to be ~ 2.3 % in this study through introducing the pure NH_4NO_3 particles into AMS. Based on the calibration result, the interference of artificial CO_2^+ was excluded in the series of subsequent analyses including OA quantification, elemental ratio calculation, and PMF analysis. Elemental ratios, including oxygen to carbon ratio (O:C), hydrogen to carbon ratio (H:C), and total OA versus organic carbon ratio (OA:OC), were determined by Improved-Ambient (I-A) method in this study (Canagaratna et al., 2015). All the AMS quantification and elemental ratio data used here were based on the "V" mode at a time resolution of 1 minute for the ambient dataset and 2.5 minutes for TD sampling (Fig. S1). The AMS data were analyzed with the software of Squirrel (v1.62G) and PIKA (V. 1.22G) based on Igor pro 6.37.

2.2.2 Other instruments

The mass concentration of black carbon (BC) was measured by an AE-31 Aethalometer (Magee Scientific, USA) at a time resolution of 5 minutes ($PM_{2.5}$ inlet). A scanning mobility particle sizer (SMPS, TSI, Inc.) was conducted to detect the particle number size distribution (mobility diameter: 15–600 nm) at a time resolution of 5 minutes. The total mass concentration measured by SMPS can be obtained by multiplying the integrated volume concentration (assuming the aerosol was spheric) by the corresponding density. The density of the total aerosol (1.63 ± 0.04 g m^{-3} on average) was calculated based on the main chemical components of aerosols measured by AMS and AE-31, as addressed in previous publications (Decarlo et al., 2008; Hu et al., 2017c; Kuwata et al., 2012). A good agreement of time series between mass concentrations from SMPS and AMS + AE-31 (Slope = 0.97 and R = 0.95, Fig. S2) was found, validating the robust measurement results shown here. Volatile organic compounds (VOCs) were measured by online gas chromatography with a flame ionization detector (GC-FID) at a time resolution of 1 hour (Lee et al., 2021). Regular gases of NO/NO_2, SO_2, CO, and O_3 were measured by NO_x Analyzer (42i), SO_2 Analyzer (43i), CO Analyzer (48i), and O_3 monitor (49i) from Thermo Fisher Scientific at a time resolution of 1 minute, respectively.

2.3 Source apportionments of PMF and ME-2

Sources of OA components were analyzed based on the conventional positive matrix factorization (PMF) method (Ulbrich et al., 2009) and the multi-linear engine (ME-2) approach (Canonaco et al., 2013). Unconstrained PMF analysis was conducted in PMF Evaluation Tool (PET) v2.08 described in Ulbrich et al. (2009). Compared with conventional PMF, the ME-2 approach allows introducing priori profiles of known OA factors, which can reduce the rotational ambiguity, and directly apportion towards environmentally meaningful solutions (Canonaco et al., 2013). In the ME-2 analysis of this study, we constrained the standard spectrum of BBOA from the Changdao study (Hu et al., 2013) at varied α values using SoFi v6.81. By investigating the characteristics of mass spectra and time series, as well as their correlations with external tracers (Zhang et al., 2014; Ulbrich et al., 2009), four OA factors were finally chosen as the optimum solution for both ambient and TD dataset. The details of PMF and ME-2 analysis can be found in the supporting information (S1.1).

2.4 Thermodenuder and volatility estimation

The TD used in this campaign is a commercial product from ARI and has been applied widely in laboratory experiments (Saha et al., 2015; Huffman et al., 2009b; Grieshop et al., 2009b; Lee et al., 2011; May et al., 2013c; Li et al., 2016a; Saha and Grieshop, 2016; Sato et al., 2018; Champion et al., 2019 and reference therein) and field studies (Huffman et al., 2009a; Hu et al., 2016c; Paciga et al., 2016; Xu et al., 2016; Louvaris et al., 2017b; Cao et al., 2018; Kostenidou et al., 2018; Xu et al., 2021; Zhu et al., 2021 and reference therein). The TD consists of two parts: the heated section and the denuder section. The heated section can realize step-wise heating ranging from 54 ˚C to 230 ˚C to obtain concentrations at various temperatures in combination with AMS or SMPS. The denuder section is filled with activated charcoal to absorb the gas-phase species evaporated from particles within TD. The estimated residence time through the 55 cm heating section is 20.2 seconds, which is comparable with that (21.2 seconds) used by Huffman et al. (2008; 2009a; b). The setup of the TD-AMS system in this study is shown in Fig. S1, where sampling flow was switched to TD or bypass (ambient) paths at a time resolution of 5 min before being sampled by the AMS and SMPS. The switch of ambient and TD lines was controlled by the solenoid valve. When the instruments sample the ambient or TD lines, a make-up flow line with the same flow rate (0.62 L min^{-1}) was set to refresh the air in the other sampling line by the air pump. The residence time after solenoid valves is about 3-4s. To avoid the mixing of air between the ambient and TD lines, a delay time of 30s after each switch was set in the sampling strategy of AMS and SMPS instruments. Bulk particle losses and temperatures in TD were all corrected based on the calibration results from Huffman et al. (2008) since similar TD and its setups in both studies were applied. The corrected centerline temperature is 17 % higher than the measured temperature on average. More detailed information on TD calibrations can be found in S1.2 of the supporting information.

We calculated the MFR of aerosols, which is the ratio of mass concentration through the TD path and bypass path (ambient line) at a set temperature. Then the thermogram, MFR as a function of corrected temperature, can be obtained. The thermograms of each species were depicted to semi-quantitively characterize aerosol volatility (Huffman et al., 2009b). The kinetic limitations of aerosol evaporation exist in TD (Saha et al., 2015), which were mainly influenced by the residence time and OA concentration in TD. Thus it is difficult to directly compare the thermograms in different studies with different residence times and mass loadings (Cain and Pandis, 2017; Karnezi et al., 2014). To further describe the volatility of the complex OA matrix and make it comparable, volatility distribution, expressed as volatility bin set (VBS), was estimated based on the empirical relationship between LogP_{25} and reciprocal of T_{50} developed by Faulhaber et al. (2009). T_{50} represents the temperature value where aerosol species evaporates 50 % compared to the input mass concentration under 25 ˚C (here is referred to as ambient concentration) and P_{25} represents the vapor pressure of species at 298 K. A good agreement of OA volatility distributions between this empirical method and kinetic model method was found previously (Cappa and Jimenez, 2010).

2.5 Predictions of glass transition temperature and viscosity of organic aerosols

The viscosity has been measured directly or indirectly by applying particle rebound, poke flow, and other offline measurements (Liu et al., 2017; Reid et al., 2018; Renbaum-Wolff et al., 2013). Viscosity is associated with the glass transition temperature (T_g), at which the phase transition occurs from semisolid to glassy states (Koop et al., 2011). Recently, more predictive models were developed to estimate the T_g of mixtures in field observations (Shiraiwa et al., 2017; Li et al., 2020b; Maclean et al., 2021). According to the new parameterization method introduced by Li et al. (2020b), T_g of OA ($T_{g,org}$) at dry conditions was estimated based on OA volatility distribution and the ratio of oxygen to carbon (O:C, “Aiken-Ambient” method). The T_g of OA–water mixtures ($T_{g,\omega org}$) at a given RH can be estimated using the Gordon–Taylor equation, in which the aerosol water content associated with OA can be calculated by the effective hygroscopicity parameter (κ) (Mei et al., 2013; Mikhailov et al., 2009). Finally, viscosity can then be calculated by applying the modified Vogel–Tammann–Fulcher (VTF) equation (Angell, 1991; DeRieux et al., 2018). The viscosity can affect aerosol equilibrium time (τ_{mix}), which can be estimated by the equation: $\tau_{mix} = d_p^2/(4\pi^2 D_b)$ (Seinfeld and Pandis, 2016), where d_p is the particle diameter (assuming 200 nm), and the bulk diffusion coefficient D_b is calculated from the predicted viscosity by the fractional Stokes-Einstein relation (Xu et al., 2021; Evoy et al., 2019). The detailed computational processes can be found in S1.3 of the supporting information.

2.6 Air mass trajectory analysis

Back trajectories analysis with altitude at 100 m were calculated every 6 h during this campaign using the HYSPLIT model (http://ready.arl.noaa.gov/HYSPLIT.php) developed by the Air Resources Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA) (Draxier and Hess, 1998; Cohen et al., 2015). The weather data were obtained from the Global Data Assimilation System (GDAS) archived by NOAA Air Resources Laboratory (ftp://arlftp.arlhq.noaa.gov/pub/archives/). Then the back trajectories were divided into several groups based on their similarity in spatial distribution (cluster analysis) (Zheng et al., 2010).

In addition, the potential source contribution function (PSCF) (Polissar et al., 1999; Zhang et al., 2013) was also applied here to investigate the contribution of regional transport. This analysis was achieved through the “Zefir” software, which is an Igor-based package and specifically designed to achieve a comprehensive geographical origin analysis using a single statistical tool (Petit et al., 2017). Pollutant concentrations were put into “Zefir” software together with data obtained from HYSPLIT and NOAA. The geographic region was divided into 0.5×0.5° grid cells (latitude i and longitude j). If the endpoint of the trajectory falls into a grid cell, the total number of trajectories is calculated as n_{ij}, and the number of these endpoints which are higher than a threshold (75th percentile used here) is counted as m_{ij} (Liu et al., 2021). To reduce the bias of PSCF for grid cells with low values of n_{ij}, the weight function (w_{ij}) recommended by Waked et al. (2014) was applied to the PSCF.

$$W_{ij} = \begin{array}{l} 1\text{ , for } n_{ij}>0.8*\max(\log_{(n+1)}) \\ 0.725\text{, for}<0.6\max(\log_{(n+1)})*\ n_{ij}\leq 0.8*\max(\log_{(n+1)}) \\ 0.35\text{, for}<0.35\max(\log_{(n+1)})*\ n_{ij}\leq 0.6*\max(\log_{(n+1)}) \\ 0.1\text{ , } n_{ij}\leq 0.35*\max(\log_{(n+1)}) \end{array} \qquad (1)$$

3 Results and discussion

3.1 Concentrations of PM_1 chemical compositions

The time series of mass concentration from the main components in PM_1 are shown in Fig. 2. The overall PM_1 concentration ranges dramatically from 2.5 to 152.8 μg m^{-3} with an average value of 31.5 ± 22.7 μg m^{-3} (Fig. 2a, Table 1), which is generally lower than the average values in urban areas of NCP, e.g., 45 – 53 μg m^{-3} in the spring of Beijing (Hu et al., 2017a; Sun et al., 2015). During this campaign, nitrate was the dominant species, accounting for 33 % of total PM_1 mass concentration, then followed by OA (25 %), sulfate (17 %), ammonium (17 %), BC (4 %), and chloride (3 %) (Fig. 2b).

When the mass concentration of PM_1 is below 20 μg m^{-3} (Fig. 2c), OA and sulfate account for the largest proportion, about 40 % and 22 % respectively. Nitrate increases dramatically and reaches 39 % of PM_1 in the polluted periods (tinted by yellow color in Fig. 2a). The NO_2^+/NO^+ ratio of ambient nitrate in AMS (average value = 0.27) is similar to the ratio from pure ammonium nitrate calibration (average value = 0.28) (Fig. S16), suggesting that the observed nitrate here is nearly inorganic (> 90 %). An increase in inorganic nitrate at higher aerosol concentration has also been observed in other field studies including urban areas (Chen et al., 2021a; Chen et al., 2021b; Griffith et al., 2015; Li et al., 2018), downwind sites (Qin et al., 2017; Li et al., 2020a) and also other receptor sites (Hu et al., 2013; Li et al., 2014). As the air quality control policy was continuously performed in China, the emission of SO_2 decreased substantially compared to that of NO_2, and nitrate has become the dominant secondary component in the fine particles (Xie et al., 2020; Geng et al., 2017a; Li et al., 2019).

The formation of nitrate has been identified as the following two main chemical pathways: (1) homogeneous reaction of gaseous nitric acid (HNO_3) and ammonia (NH_3), in which HNO_3 is largely produced by photochemical oxidation of NO_2 by OH radicals in the daytime (Yang et al., 2022); (2) Heterogeneous hydrolysis of dinitrogen-pentoxide (N_2O_5), which is the major source of HNO_3 at night (Griffith et al., 2015); In general, abundant precursors, high pH and aerosol liquid water content (ALWC) during polluted periods might further promote these two processes (Chen et al., 2021b; Wang et al., 2020). In the remote receptor site, in addition to the chemistry process, regional transport is another important source of high nitrate mass concentration (Squizzato et al., 2012). In this study, increased nitrate can be observed when the high-speed winds from southeast to southwest dominate (above 8 km h^{-1}) (Fig. 3a1), suggesting that regional transport in winter and spring of NCP areas plays an important role in the enhanced nitrate observed here (Huang et al., 2020; Hu et al., 2016b). The sulfate, ammonium, and BC show similar patterns as nitrate in diurnal variations (Fig. 2d) and bivariate polar plots (Fig. 3), supporting the upwind anthropogenic emissions were transported to the regional receptor areas in the downwind direction. The PSCF results based on backward trajectory analysis were applied to further explore the potential transport from different regions to this receptor site. High concentrations of secondary inorganic aerosols are mainly contributed by the west and south regions of Shandong province, and the northern part of Anhui and Jiangsu province, where high anthropogenic $PM_{2.5}$ emissions exist (Lei et al., 2011; Geng et al., 2017b). The results again verify the important transport influences on aerosol chemistry from NCP urban outflows to receptor areas.

Table1. Summary of average mass concentrations of PM_1 species and OA factors (μg m^{-3}), average mixing ratios of gas-phase pollutants, and values of meteorological parameters during the entire campaign and polluted period. The polluted period definition can be found in the caption of Fig. 1.

	Dongying Spring (Average± SD)	
	Entire Study	Polluted Period
Total PM_1 (μg m^{-3})	31.5 ± 22.7	50.3 ± 5.4
Total OA (μg m^{-3})	7.9 ± 4.5	12.5 ± 4.1
BBOA	0.4 ± 0.7	0.7 ± 0.7
HOA	2.2 ± 2.0	3.5 ± 2.6
Transported-OOA	2.6 ± 2.7	5.1 ± 2.8

Background-OOA	2.6 ± 1.6	3.1 ± 1.7
Sulfate (μg m^{-3})	5.3 ± 3.9	8.0 ± 3.7
Total Nitrate (μg m^{-3})	10.6 ± 9.4	17.4 ± 8.5
Inorganic nitrate	10.0 ± 9.3	16.8 ± 8.7
Organic nitrate	0.9 ± 0.8	0.7 ± 0.9
Ammonium (μg m^{-3})	5.5 ± 4.4	8.9 ± 4.0
Chloride (μg m^{-3})	1.0 ± 1.6	1.7 ± 2.0
BC (μg m^{-3})	1.2 ± 0.8	1.8 ± 0.7
NH_3 (ppb)	26.7 ± 19.0	15.6 ± 11.6
CO (ppm)	0.5 ± 0.3	0.6 ± 0.3
NO (ppb)	0.9 ± 2.1	1.3 ± 2.5
NO_2 (ppb)	11.8 ± 7.2	16.5 ± 6.8
SO_2 (ppb)	3.1 ± 3.5	4.9 ± 3.8
O_3 (ppb)	47.1 ± 22.2	51.4 ± 25.5
Wind speed (m s^{-1})	3.1 ± 1.8	2.6 ± 1.4
Temperature (°C)	10.5 ± 7.2	15.6 ± 6.0
RH (%)	60.7 ± 19.2	57.9 ± 18.1
ALWC (μg m^{-3})	13.3 ± 18.3	23.6 ± 63.0
pH	3.9 ± 0.7	3.7 ± 0.7
Pressure (hPa)	101.4 ± 0.7	101.1 ± 0.4

Total PM_1 = OA + Sulfate + Nitrate + Ammonium + Chloride + BC

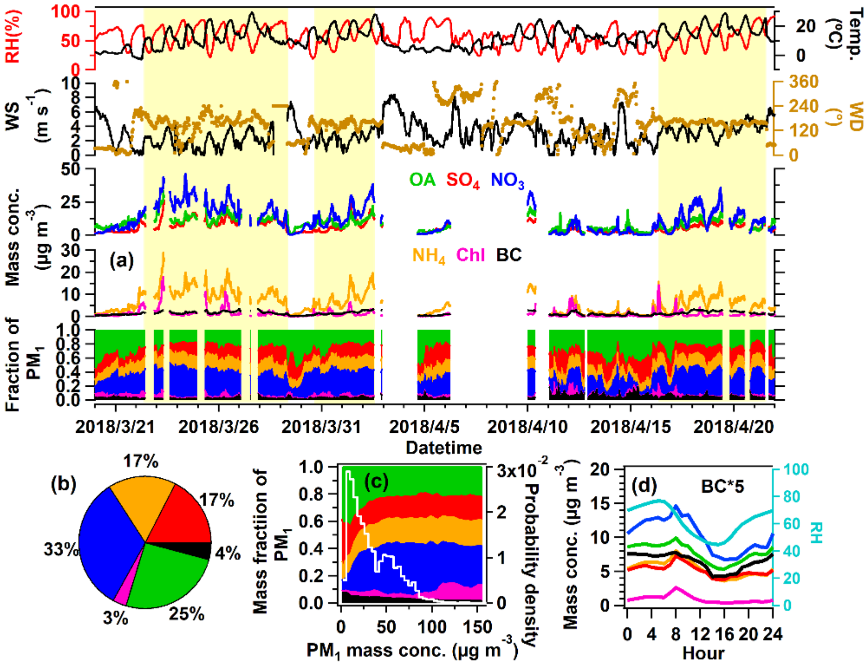

Figure 2. (a) Time series of main species in PM₁ and meteorological parameters (relative humidity, temperature, wind speed, and wind direction). The yellow background represents the polluted period in this campaign, which was selected based on mass concentration of total PM₁ above 20 μg m^{-3} and lasts for at least 2 days, accompanied by continuous south winds; (b) Average chemical compositions of PM₁; (c) Variations of main compositions (left axis) and probability density as a function of PM₁ mass concentration (right axis); (d) Average diurnal variations of PM₁ species and PBL height during the entire campaign.

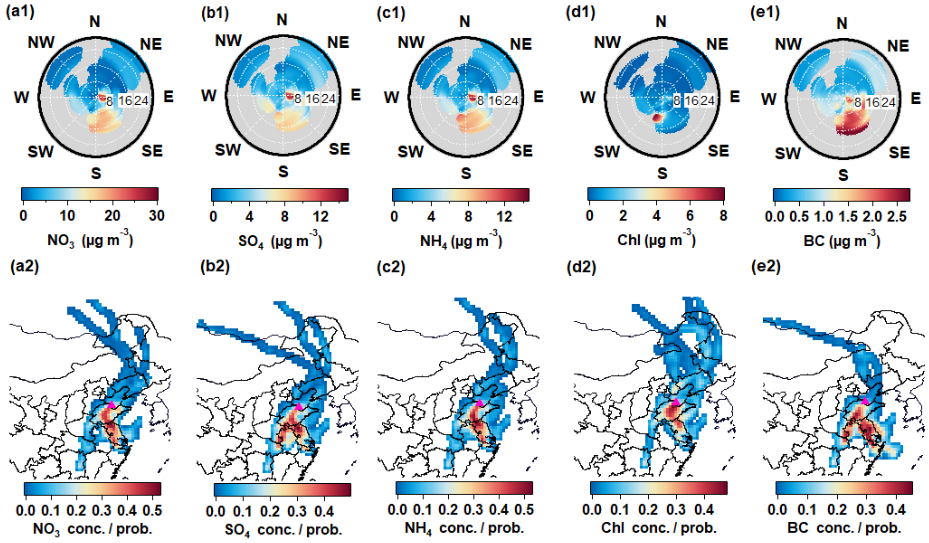

Figure 3. Bivariate polar plots and the results of potential source contribution function (PSCF) from inorganic species, including (a1, a2) NO_3; (b1, b2) SO_4; (c1, c2) NH_4; (d1, d2) Chl; (e1, e2) BC. The unit of wind speed is km h^{-1}.

3.2 Source apportionment of OA

Four OA factors were resolved in this study, including BBOA (5 %), aged-HOA (28 %), transported-OOA (33 %), and background-OOA (33 %) respectively, as shown in Fig. 4.

3.2.1 Biomass Burning OA (BBOA)

BBOA is possibly from (1) biofuel/biomass burning from domestic heating in the NCP (Duan et al., 2020; Huang et al., 2020; Elser et al., 2016); (2) The open biomass burning around observation site (Li et al., 2017a; Liang et al., 2021). It is indeed that biomass burning plumes from open fire burning were frequently seen during the campaign. As shown in Fig. S17, a positive correlation between BBOA with surrounding fire counts was found, suggesting that open biomass burning might play a more important role than domestic heating. Potassium (K^+), regarded as a good indicator of biomass burning (Aiken et al., 2010) also shows a good correlation with BBOA (R = 0.6), validating the BBOA resolved here.

$C_2H_4O_2^+$ ion is mainly produced by the pyrolysis of levoglucosan and other anhydrous sugars from thermal decomposition of cellulose and lignin (Bertrand et al., 2018), $C_2H_4O_2^+$ ion (*m/z* 60) has been considered as a tracer for BBOA (Cubison et al., 2011). As depicted in Fig. S18a, the higher f_{60} value in OA above the background level (0.3 %) is concurrent with an increased BBOA fraction. The O:C value of BBOA is 0.37, which is in the range of reported values (0.2–0.65) from primary BBOA observed in other studies (Zhou et al., 2020; Hu et al., 2016b; Xu et al., 2017; Zhang et al., 2016). According to the bivariate polar plot and PSCF result of BBOA in Fig. 5b, the higher concentration of BBOA partially comes from local emission of biomass burning southeast of the sampling site.

3.2.2 Aged hydrocarbon-like OA (aged-HOA)

HOA has been widely recognized and elaborated on in previous studies (Hao et al., 2014; Hu et al., 2016b; Aiken et al., 2009; Huang et al., 2020; Sun et al., 2014; Zhang et al., 2011; Zhou et al., 2020; Zhang et al., 2005a; Zhang et al., 2007; Ng et al., 2011b). The HOA resolved in this study is not only contributed by fragments of C_xH_y (C_nH_{2n+1} and C_nH_{2n-1}, m/z 55, 57, 69, 81, 91, etc.) resembled the reported reference spectra of HOA (Hu et al., 2013; He et al., 2011; Ng et al., 2011a; Zhang et al., 2005b; Zhu et al., 2021; Jimenez et al., 2009) but also includes a high abundance of oxygenated ions (i.e., CO_2^+). It suggests that the HOA resolved here is partially aged, and thus is entitled to aged-HOA. The diurnal variations of aged-HOA mass concentration and its fraction in total OA, show a prominent peak during the traffic rush in the morning (8:00 am), then decrease sharply with the elevated boundary layer, suggesting this factor is influenced by primary vehicle emissions and meteorological condition (Fig. 4, Fig. S19). The primary property of aged-HOA in this study was also identified by their good correlation with primary anthropogenic VOCs of benzene and ethyl toluene (with R = 0.82 and 0.8 respectively) (Hu et al., 2016a; Lee et al., 2015; Mohr et al., 2012). In addition, aged-HOA has the characterization of secondary sources. The O:C value is 0.55, which is out of the range of O:C of HOA observed in China (0.05–0.44) (Hu et al., 2013; Zhou et al., 2020; Hu et al., 2016c). The high O:C value of aged-HOA in this study may be caused by long-distance transport from surrounding urban/town areas (Fig. 1a). Based on the bivariate polar plot and PCSF analysis in Fig. 5c, the higher aged-HOA concentration is in accord with urban plumes originating from the west to south region, where Dongying city and northern Jiangsu are on this path. It generally will take 4-8 hours for the vehicle emissions to be transported from Dongying and 31-70 hours from northern Jiangsu to this site at a relatively high wind speed (7-16 km/h).

3.2.3 Transported-OOA and Background-OOA

Transported-OOA and background-OOA, representing two types of SOA with different origins, were resolved in this study. Both factors are fairly oxidized, which are characterized by the high mass abundance of oxidized m/z 44 and m/z 28 in total OA (0.23 for transported-OOA, 0.21 for background-OOA). Transported-OOA has an O:C ratio of 0.97, which is slightly lower than the 1.1 for background-OOA. Both OOA factors are in the high range compared with most OOA factors observed in urban and suburban areas (0.29–1.3) (Li et al., 2015; Xu et al., 2019) and remote areas (0.47–1.45) (Zhou et al., 2020; Zheng et al., 2017; Gong et al., 2012).

The transported-OOA shows good correlations with secondary inorganic sulfate and nitrate (R = 0.81 and 0.83 shown in Fig. S14, respectively), consistent with its secondary origins of OOA (He et al., 2011; Huang et al., 2020; Zhang et al., 2005a). The regression ratio between transported-OOA vs. sulfate + nitrate is around 0.84, within the range obtained in NCP areas (0.51–0.93) (Sun et al., 2016; Sun et al., 2012; Huang et al., 2010). Similar to secondary inorganic species, the higher mass concentration of transported-OOA is also driven by the south winds, thus named transported-OOA. The bivariate polar plots and PSCF results (Fig. 5d) show that the higher transported-OOA concentration might correspond to the transport processes from urban emissions located in the southern region of the sampling site. On the contrary, the background-OOA shows enhanced mass concentrations in all directions(Fig. 5e1) compared with transported-OOA, indicating its background origins, and representing the background concentration in NCP (2.6 ± 1.6 μg m^{-3}). In addition, in section 3.4.2, we found very low volatility of background-OOA, confirming it is very aged in the ambient air.

On average, the sum of transported-OOA and background-OOA can account for 66 % of total OA. When the total OA concentration is below 3 μg m^{-3}, background-OOA accounts for nearly 50 % of the OA mass concentration as shown in Fig. 4f. While the total OA increases from 4 to 20 μg m^{-3}, the transported OOA from urban plumes starts to dominate (from 20 % to 44 %). The dominance of OOA contribution to OA is consistent with findings in the urban outflow of Mexico City (Jimenez et al., 2009). As OA concentration enhances above 20 μg m^{-3}, the aged-HOA and BBOA fractions increase from 29 % to 53 % and 6 % to 16 %, respectively, confirming the primary anthropogenic emission is still an important contributor to the air pollution periods at a remote receptor site.

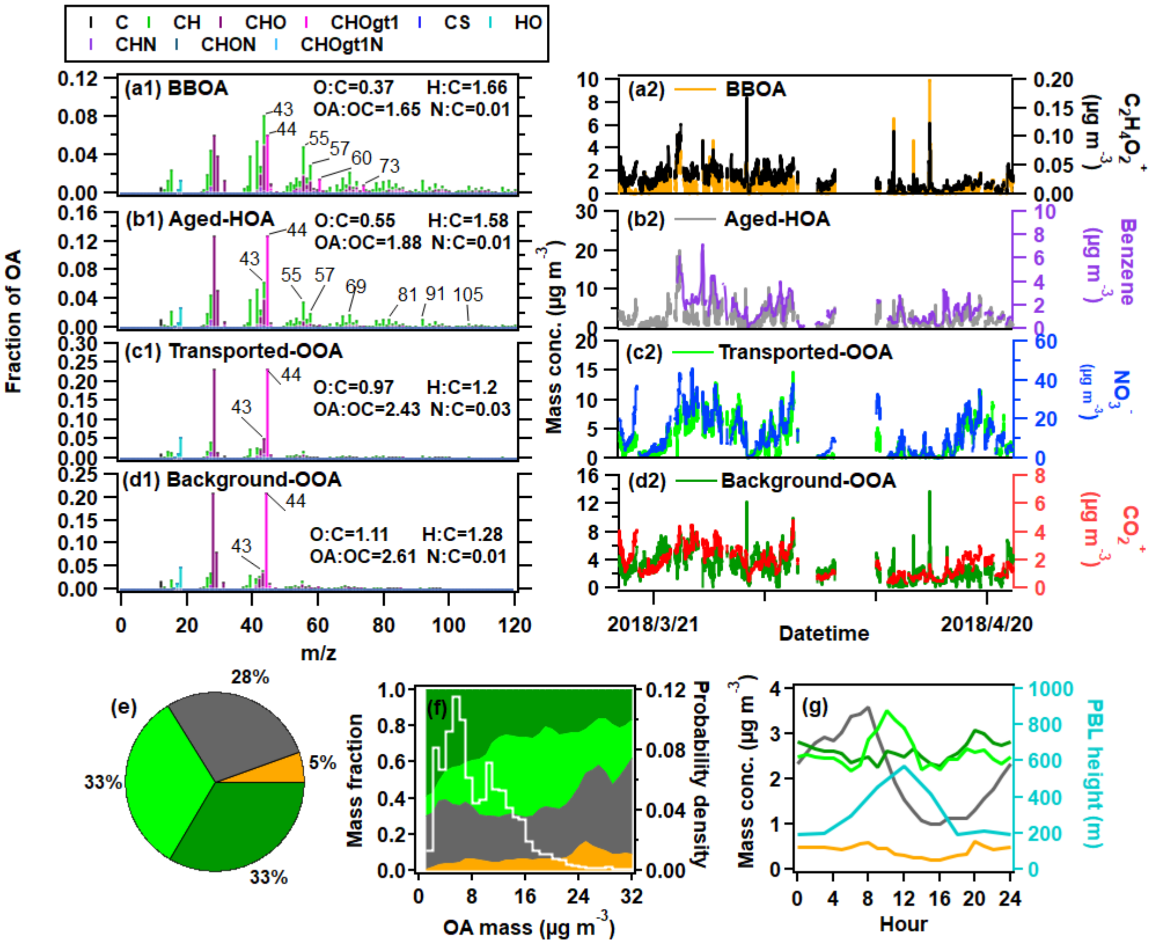

Figure 4. Mass spectra (left) and time series (right) of four OA factors: (a1, a2) BBOA, (b1, b2) aged-HOA, (c1, c2) transported-OOA, (d1, d2) background-OOA; Time series of tracers associated with each factor were also displayed. (e) Average chemical compositions of OA; (f) Variations of OA compositions as well as probability density as a function of OA mass concentration. (g) Average diurnal variations of OA factors and planetary boundary layer (PBL) height during the whole campaign.

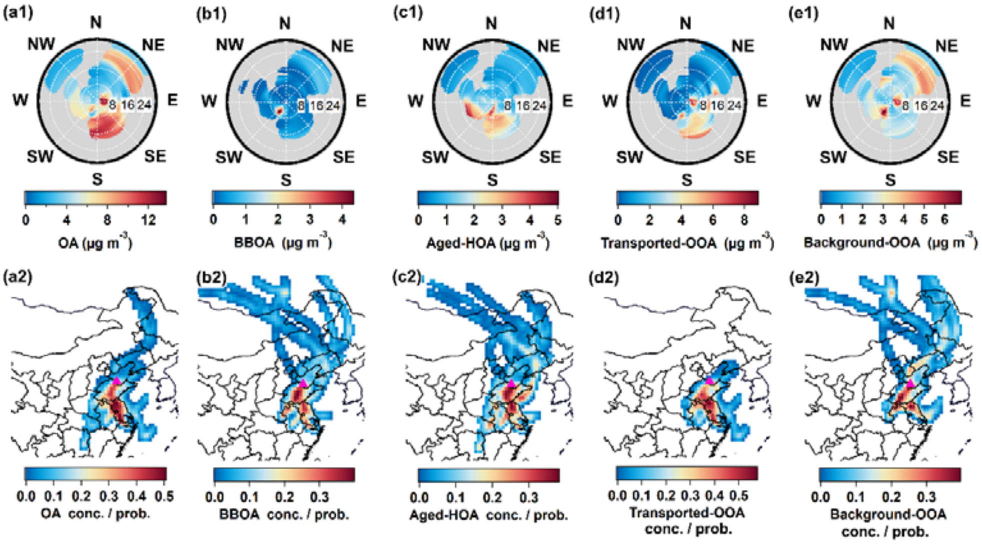

Figure 5. Bivariate polar plots and PSCF results of organic components, including (a1, a2) total OA; (b1, b2) BBOA; (c1, c2) Aged HOA; (d1, d2) transported-OOA; (e1, e2) background-OOA. The unit of wind speed is km h^{-1}.

3.4 Volatility of PM_1 species and OA factors

3.4.1 Volatility of inorganic species

Fig. 6 shows the thermograms of non-refractory aerosols as well as their main chemical components. As the TD temperature increases, the MFR of sulfate keeps constant until 115 ℃, leading to a slight increase in its fraction in total PM_1. The less steep thermogram of sulfate compared with those of other species is mainly due to the low volatility and its slight increase of collect efficiency caused by phase variation upon heating (Huffman et al., 2009a), which have been consistently observed in laboratory experiments (An et al., 2007; Huffman et al., 2009a) and field studies (Cao et al., 2018; Huffman et al., 2009a; Xu et al., 2016; Xu et al., 2021; Kang et al., 2022). When TD temperature is above 150 ℃, the MFR curve of sulfate descends sharply to 6.5 %. However, a part of sulfate remains at the highest temperature compared to that of pure $(NH_4)_2SO_4$ from our laboratory experiment, indicating that the partial sulfate signals detected by AMS might be organic sulfates or metallic sulfates (Fig. 6c). The decreased $H_2SO_4^+/SO_4^+$, HSO_3^+/SO_4^+, SO_3^+/SO_4^+ ratio and increased SO_2^+/SO_4^+ ratio as functions of TD temperature indeed were observed (Fig. S20), indicating the possible existence of organic sulfate (Chen et al., 2019). The $CH_3SO_2^+$ ion (m/z 78.99), which is considered to be a fragment ion from methanesulfonic acid (MSA) influenced by marine aerosols (Zhou et al., 2016; Huang et al., 2015; Zorn et al., 2008) and also possibly an indicator of sulfur-containing aerosols influenced by traffic and fossil carbon in urban areas (Daellenbach et al., 2017; Vlachou et al., 2019), shows higher volatility than sulfate in this study. The higher volatility of the $CH_3SO_2^+$ ion is probably influenced by its different bond types connected to the sulfate group than sulfate (Chen et al., 2019). Similarly, volatility of laboratory generated-MSA particles is higher than that of ammonium sulfate (Fig. 6c), which is consistent with the ambient results.

To quantitively compare the thermograms of different species, T_{50}, the temperature when MFR is equal to 0.5 is used here (Faulhaber et al., 2009; Cappa, 2010b). To obtain the T_{50} more accurately, the thermograms were fitted by Hill's Equation (S1.2 in supporting information) (Emanuelsson et al., 2013; Kolesar et al., 2015). Compared to sulfate (T_{50} = 159 ℃), the T_{50} values of nitrate and chloride are 57 ℃ and 61 ℃ (Figs. 6d and e), respectively, supporting their semi-volatile properties in the ambient air (Cao et al., 2018; Xu et al., 2016; Xu et al., 2021). The evaporated loss of ambient nitrate has also been frequently observed in the offline membrane samples, in which 60–80 % of nitrate can evaporate in the summer (Chow et al., 2005; Hering and Cass, 1999). The similar phenomenon of NH_4NO_3 evaporation (30–40 %) has been found in the field-deployed oxidation flow tube, the temperature inside of the reactor is about 10 ℃ higher than that in the ambient air due to the heated cabinet of the airplane (Nault et al. 2018). Despite the quick evaporations of ambient nitrate and chloride, their T_{50} values are still higher than those of pure ammonium salts (i.e., NH_4NO_3 and NH_4Cl) generated by an atomizer in the laboratory experiments (T_{50} = 37 ℃ and 54 ℃, respectively). When TD temperature is above 150 ℃, compared to the complete evaporations of pure NH_4NO_3 and NH_4Cl (Fig. 6), ~ 5 % of nitrate and ~ 10 % of chloride in ambient air remained. The fractions of nitrate and chloride in total PM_1 species also increase from 5 % and 1 % at middle temperatures to 14 % and 3 % at higher temperatures (> 180 ℃). These increases have also been observed in other field studies conducted with the TD-AMS system (Huffman et al., 2009a; Xu et al., 2019). For nitrate, the high remaining fraction is probably caused by the partial contribution from organic nitrate or non-refractory inorganic salts (e.g., $NaNO_3$). This deduction is supported by the continuously decreased NO_2^+/NO^+ ratio (0.06 finally), consistent with similar low values in organic nitrate ($\approx$ 0.1, a factor of 2.75 lower than the ratio of 0.28 in NH_4NO_3) (Day et al., 2022; Fry et al., 2013) and $NaNO_3$ ($\approx$ 0.006, a factor of 50 lower) (Hu et al., 2017c). The remaining of chloride at the highest temperature might indicate the presence of metallic chlorides, such as $ZnCl_2$, $PbCl_2$, and KCl, which are usually emitted from combustion sources at high temperatures (Huffman et al., 2008; Xu et al., 2019; Aiken et al., 2009; Moffet et al., 2008). The positive correlation between Cl^- vs. Pb^{2+} (R=0.51), as well as Cl^- vs. K^+ (R=0.54) supports this probability (Fig. S21).

Quick evaporation of ammonium species than cation species during heating was observed, leading to acidified aerosols (measured NH_4/predicted NH_4 < 1) (Fig. 6f). This is mainly due to the decomposition of ammonium salts and yield of gas-phase NH_3 upon heating (Hu et al., 2017b). Similar behavior of acidified aerosol in the TD line was exhibited in other field studies (Huffman et al., 2009a; Denkenberger et al., 2007).

In summary, the volatility of inorganic species rank in the sequence of nitrate > chloride > ammonia > sulfate in this study. Compared to their pure ammonium salts, all ambient inorganic aerosols have lower volatilities and part of them remain at high temperatures, indicating the existence of low volatile metallic or organic-related species in AMS detected signals. These results support the complex property of ambient aerosols.

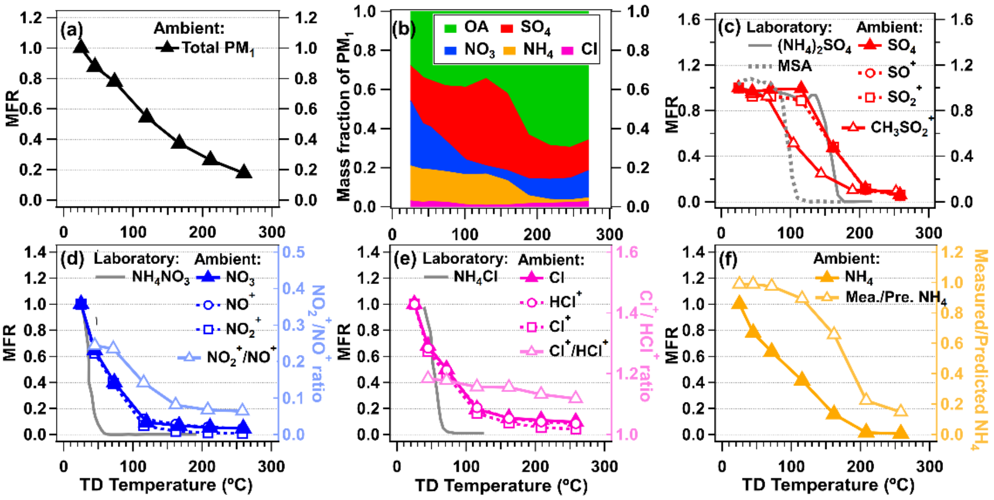

Figure 6. (a) Mass fraction remaining (MFR) of total NR-PM$_1$; (b) Fraction of main components in NR-PM$_1$ as a function of TD temperature; (c) Variations in the average MFR of the inorganic species with their corresponding constituent ions as functions of TD temperature: (c) sulfate; (d) nitrate; (e) chloride; (f) ammonium. In addition, MFRs of pure ammonium salts of sulfate, nitrate, and chloride, as well as methanesulfonic acid (MSA) measured in this study were shown for comparison. Particle acidity calculated as the measured NH_4 vs predicated NH_4 assuming a neutral balance from aforementioned cations as a function of TD temperature is also shown in the right axis of panel (f).

3.4.2 Volatility of OA and different OA factors

The thermograms of total OA and OA factors are shown in Fig. 7. The T_{50} of total OA is about 132 °C, which is higher than that of nitrate ($T_{50} = 57$ °C), while lower than that of sulfate ($T_{50} = 159$ °C). Total OA thermograms in Riverside, LA and Mexico City are also shown in Fig. 7a. Thermograms of these three studies should be comparable since they have a similar residence time of TD (about 21s) and OA concentration (7.9–19.9 μg m^{-3} in average) (Huffman et al., 2009a; Docherty et al., 2011; Aiken et al., 2009), as well as the similar TD setup. During these two campaigns in Riverside, LA and Mexico City, the T_{50} of OA (104 °C and 106 °C respectively) are lower than that obtained in Dongying ($T_{50} = 132$ °C). The slightly lower volatility of OA in Dongying is reasonable since the OA is more oxidized (O:C = 0.85) than those from Riverside, LA and Mexico City (O:C = 0.45 and 0.54 respectively) (Aiken et al., 2008; Docherty et al., 2011).

To further compare the volatility results among more studies, the volatility distribution of OA was estimated based on the empirical method of Faulhaber et al. (2009). As is shown in Fig. 8a, the volatility of OA is distributed in the range of $C^* = 10^{-9}$–10^{2} μg m^{-3} with two prominent peaks around $C^* = 10^2$ and 10^{-4}–10^{-3}μg m^{-3}. If we categorize the entire distribution into different groups based on volatility introduced by Seinfeld and Pandis (2016), the OA in this study is composed of 52 % extremely low volatile organic compounds (ELVOCs, $C^* \leqslant 10^{-4}$ μg m^{-3}), 26 % low volatile organic compounds (LVOCs, $10^{-4} < C^* \leqslant 10^{-1}$ μg m^{-3}) and 22 % semi-volatile organic compounds (SVOCs, $10^{-1} < C^* \leqslant 10^{2}$ μg m^{-3}). The SVOCs fraction in this study is lower than the values from urban areas (37–67 %), however, is higher than that of OA strongly influenced by biogenic emissions (11–16 %), as shown in Fig. 9a. The former is mainly due to the OA in this study being more oxidized than those from urban areas where fresh primary emissions impact strongly. The lower SVOCs fraction of OA in forest areas is reasonable since the OA is mainly composed of the high fraction of biogenic SOA, which is strongly influenced by isoprene and monoterpene emissions. The autoxidation and oligomerization during biogenic SOA formation leading to lower volatility have also been reported (Ehn et al., 2014; Bianchi et al., 2017; Lopez-Hilfiker et al., 2019).

To further investigate the OA volatilities from different sources, the fractions of OA factors as a function of TD temperature and their thermograms are shown in Figs. 7b–f. In general, aged-HOA, whose MFR is also characterized by its internal tracers ($C_xH_y^+$), is the most volatile component before 120 °C among all OA factors. During this campaign, half of aged-HOA evaporates at ~ 92 °C (T_{50}), which is similar to that in Riverside, LA (T_{50} = 92 °C) and slightly higher than that in Mexico City (T_{50} = 78 °C). While the temperature is above 100 °C, the MFR of aged-HOA in this study is much higher than those in the other two campaigns (Huffman et al., 2009a), consistent with that HOA was oxidized during the transport process leading to a lower volatility. As shown in the plots of volatility distributions in Figs. 8b and 9b, the non-volatile ELVOCs and LVOCs fraction of aged-HOA is around 60 % in this study, which is higher than the HOA obtained in other areas (21–40 %), however, similar to the HOA value obtained in Athens during wintertime (72 %, Fig. 9b), which was also conducted at an urban background site. This suggests the aging processes greatly decrease the POA volatility. For the laboratory-generated POA from diesel/gasoline emissions, their non-volatile fractions only account for 3–27 % of OA, indicating much higher volatility of POA than that of ambient HOA. The lower volatility supports that ambient HOA resolved in ambient air is a complex mixture and might be partially oxidized (Cappa and Jimenez, 2010). We examined if low volatile PAH compounds might contribute to the low volatility of HOA, whereas negligible PAH ions contribution to OA was found. This suggests the PAH is not the main reason leading to the high non-volatile contribution of aged-HOA.

The thermogram of BBOA is shown in Fig. 7d. The T_{50} of BBOA in this study is 81 °C, which is slightly lower than that of HOA (92 °C) in this study. The BBOA shows a much lower residual (< 5 %) than that of aged-HOA when TD temperature is above 150 °C, consistent with the fact that BBOA observed here is fresher and more local than aged-HOA. Although BBOA is the most volatile factor resolved in this study, it still shows the highest non-volatile fraction (72 %) compared with the results of BBOA in other studies (31–49 %) (Fig. 9c). The diversity of BBOA volatility distributions might be caused by the different burning materials and conditions, as well as the influence of aging processes (Huffman et al., 2009b; May et al., 2013c; Xu et al., 2021). It is also supported by the fact that ambient BBOA show lower volatility than that of laboratory-generated BBOA from different combustion conditions (Grieshop et al., 2009a; Grieshop et al., 2009b; May et al., 2013c), emphasizing the aging impact on ambient BBOA volatility.

The thermograms of transported-OOA and background-OOA are shown in Figs. 7e and f. Obvious lower volatility of background-OOA than that of transported-OOA was found, which is consistent with the slightly higher O:C value of the latter (1:1 vs 0.97). The thermogram of the CO_2^+ ion, a reliable signature of SOA from thermal decarboxylation of organic acids in AMS (Aiken et al., 2009; Ng et al., 2010), more resembled the thermogram of background-OOA, which is consistent with the finding that highly oxidized OA is mainly composed of organic acids (Chen et al., 2020). We compared the volatilities of the two OOA factors resolved in this study to those reported in other ambient campaigns in Fig. 9d. It was found that the non-volatile fractions of transported-OOA and background-OOA (both are 82 %) are only lower than that of LV-OOA obtained in Mexico City (98 %) (Cappa and Jimenez, 2010), however, are higher than those from the majority of other urban OOA factors (27–71 %). This is consistent with the fact that in addition to POA the aging process can substantially decrease the SOA volatility in ambient air as well. Meanwhile, we found the SOA volatility in this study is comparable to that of isoprene-expoxydiol derived SOA (IEPOX-SOA, ELVOCs + LVOCs = 87–98 %), a large fraction of which is composed of oligomers (Hu et al., 2016c; Krechmer et al., 2015), suggesting the probable existence of oligomers in the two very aged SOA resolved here. In addition, we investigated the OA spectrum as a function of TD temperature, as shown in Fig. S22. When the TD temperature is above 200 ℃, the fraction signals of some ions at m/z >300 were enhanced, implying the existence of oligomers.

By summarizing all the average volatilities of OA and OA factors in previous and this studies (average log C* ranged from –5.34 μg m^{-3} to –0.015 μg m^{-3}), the scatter plot of O:C vs average log C* is shown in Fig. 10. In general, the average log C* exhibits slight dependence on the ratio of O:C (slope = –0.18), which do not show a clear trend as found in the laboratory SOA (Jimenez et al., 2009). The difference is possibly due to that oligomerization and decomposition might result in different volatilities, however, with similar O:C values (Hildebrandt et al., 2010; Kroll and Seinfeld, 2008). E.g., previous studies have shown that the OA, oligomers, in particular, can decompose to more volatile components or form new/less non-volatile species upon heating (Cappa and Jimenez, 2010). Thus, the fraction of oligomers in OA did not show the same trend with O:C ratios during the heating process (Zhao et al., 2020). In addition, the ambient RH made the relationship between OA volatility and its O:C more complicated. On the one hand, the aqueous-phase reaction has been supposed to be an important pathway for the formation of low volatility and higher molecular weight compounds (e.g., oligomer) (Zhou et al., 2016). While on the other hand, the volatility of OA increases after hydrolysis (Emanuelsson et al., 2013; Claflin and Ziemann, 2019). Therefore, the results here suggest that the ambient O:C ratio may not be a robust indicator of OA volatility among different studies based on heating detection techniques (Huffman et al., 2009a; Denkenberger et al., 2007; Cerully et al., 2015; Zhao et al., 2020).

Further, we investigated the evaporation loss of OA due to dilution according to the theoretical equation (Cappa and Jimenez, 2010), which is mainly dependent upon its non-volatile/volatile fraction.

$$E_{\mathrm{loss}}=100\%\left[1-\frac{C_{\mathrm{OA}}(\mathrm{DF})}{C_{\mathrm{OA}}(0)/\mathrm{DF}}\right] \tag{2}$$

Where DF is the dilution factor, here we assume a 30 times dilution. C_{OA}(DF) is the re-equilibrated C_{OA} after dilution, C_{OA}(0) is the OA mass before dilution, and E_{loss} is the relative mass loss due to evaporation of semivolatile components. The higher amounts of non-volatile species exist, the lower evaporation losses to OA dilution. Finally, the evaporation loss of dilution follows reversely the order of the fraction of non-volatile species (f_{nv}), aged-HOA (f_{nv} = 0.63) > BBOA (f_{nv} = 0.72) > transported-OOA (f_{nv} = 0.82) ≈ background-OOA (f_{nv} = 0.82), as shown in Fig. 8f. After a factor of 10 dilution, aged-HOA would evaporate 30 % at ambient temperature, which suggests that even the HOA is pretty aged in this study and it can still be oxidized continuously, thus contributes the SOA formation during the long-distance transport. This suggests a high SOA formation potential in the outflow of NCP urban emissions.

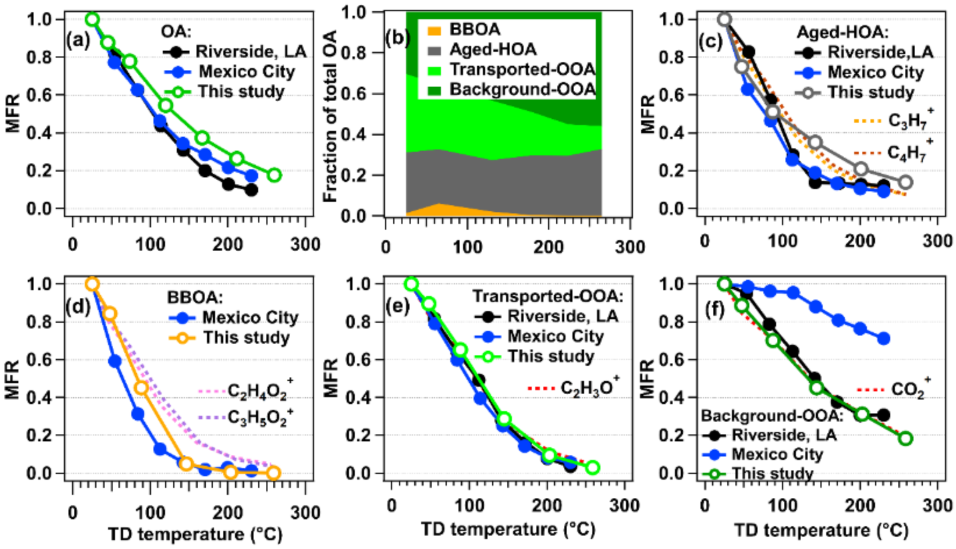

Figure 7. (a) Thermogram of total OA; (b) Fraction of four factors in total OA as functions of TD temperature; MFRs of (c) aged-HOA, (d) BBOA, (e) transported-OOA, and (f) background-OOA. For comparison, the MFR curves observed in other campaigns, such as Riverside, Los Angeles, USA, and Mexico City, Mexico (Huffman et al., 2009a) are also shown together due to their similar residence time in TD, which are marked by black and blue solid circles, respectively.

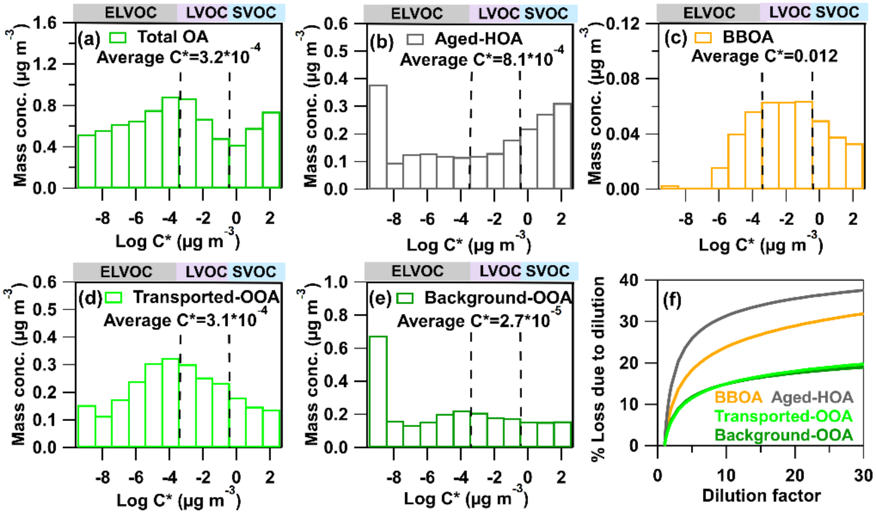

Figure 8. Volatility distributions of (a) total OA, (b) aged-HOA, (c) BBOA, (d) transported-OOA, (e) background-OOA. The unit of average C* is µg m^{-3}. (f) The additional mass losses (E_{loss}) of four OA factors as functions of the dilution factor.

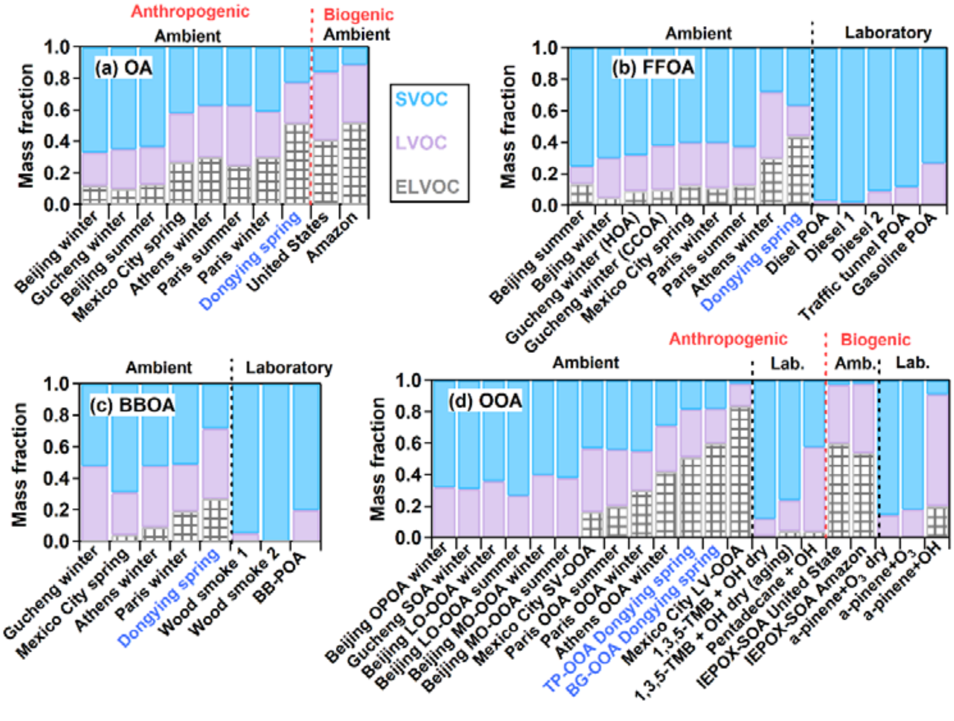

Figure 9. Fractional contributions of various types of OA for different studies, including studies in Dongying (this study), Beijing and Gucheng (Xu et al., 2021), Paris (Paciga et al., 2016), Athens (Louvaris et al., 2017b), Mexico City (Cappa and Jimenez, 2010), southeastern of United State and Amazon (Hu et al., 2016c) and several laboratory experiments (Grieshop et al., 2009b; Grieshop et al., 2009c; Li et al., 2016a; May et al., 2013a; May et al., 2013b; May et al., 2013c; Sato et al., 2018; Sato et al., 2019; Ylisirniö et al., 2020). Detailed data can be found in Table S2–S5. In panel (b), FFOA (fossil fuel-related OA) represents a sum of CCOA (coal combustion OA) and HOA. In panel (d) LO-OOA, MO-OOA, SV-OOA, and LV-OOA represent less oxidized OOA, more oxidized OOA, semi-volatile OOA, and low volatile OOA, respectively. TP-OOA and BG-OOA represent transported-OOA and background-OOA resolved in this study. TMB and IEPOX-SOA represent trimethylbenzene and Isoprene-epoxydiols-derived secondary organic aerosol, respectively. Lab. and Amb. represent studies from laboratories and field measurements of ambient air.

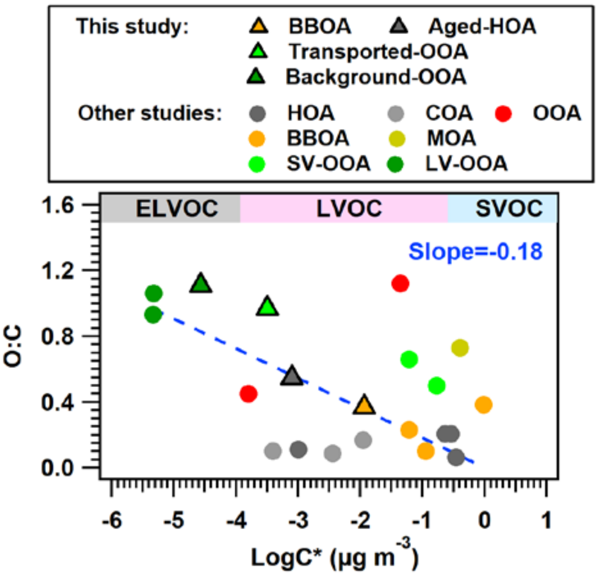

Figure 10. Scatter plot of O:C and average log C* from this and previous studies. MOA represents marine OA. The detailed values in this plot are exhibited in Table S2–S5.

3.5 Viscosity of OA

During this campaign, the predicted glass transmission temperature of OA ($T_{g,org}$) under dry conditions is 332.7 K, which is in the range of ambient $T_{g,org}$ values estimated by Li et al. (2020b), and is much higher than the values (288.9–291.5 K) estimated in the urban area of Beijing and at the rural site of Gucheng (Xu et al., 2021). It is consistent with the lower volatility of OA in this study, where ~ 78 % (shown in Fig. 9 above) of OA are non-volatile compounds compared with 30–40 % in Beijing and Gucheng (Xu et al., 2021).

The diurnal variations of OA viscosity as well as corresponding temperature and RH in previous and these studies are shown in Figs. 11a and b. According to the viscosity categories (Reid et al., 2018), the ambient OA can exist in three phase states, liquid ($\eta < 10^2$ Pa s), semisolid ($10^2 \leq \eta \leq 10^{12}$ Pa s), and amorphous solid ($\eta > 10^{12}$ Pa s). When $T_{g,org}$ is higher than the ambient temperature, aerosol occurs as a solid phase, otherwise behaving as semisolid or liquid (Koop et al., 2011; DeRieux et al., 2018). The viscosity of ambient OA in this study generally ranges from 10^{-2} to more than 10^{12} pa s, which shows that the OA phase exists mainly as semi-solid (60 %), then followed by liquid (21 %, under high RH) and solid phases (19 %, low RH and high O:C). As shown in Figs. 11a and b, the average daily fluctuation of RH could inversely affect the viscosity of OA, which stayed low (5×10^2–10^6 pa s) during the nighttime and then increased during daytime, thereby reaching a solid phase state (> 10^{12} pa s) between 14:00–16:00. The variation of OA viscosity is similar to the diurnal variation of O:C. It is expected since higher O:C due to photochemistry during the daytime leads to aerosol less volatile, which can result in a higher $T_{g,org}$, thus higher viscosity (Shiraiwa et al., 2011; Mikhailov et al., 2009).

For fixed volatility and hygroscopicity (κ) of OA, ambient RH is the key parameter to determine the viscosity. We calculated the RH-dependent viscosity based on the volatilities of different OA factors in Fig. 11. Compared with the deployed field observations and laboratory experimental results, the predicted viscosities of two OOA factors are generally within the range of viscosity values from ambient OOA summarized in Li et al. (2020b), and in the higher ranges of these from laboratory-generated SOA summarized in DeRieux et al. (2018). It is reasonable since the laboratory-generated SOA usually has a lower viscosity due to a lower degree of oxidation (Aiken et al., 2008; Chen et al., 2015). Consistent with the inverse order of volatility, OOA factors in this study undergo liquifying transition when the RH is above 67 % for transported OOA and 73 % for background OOA. The viscosity curves of aged-HOA and BBOA as functions of RH are more viscous, which is consistent with what has been found in other urban HOA (as the grey color tinted in Fig. 11c). Aged-HOA and BBOA in this study are more viscous on account of low f_{44} relative to the OOA factors, that is low hygroscopicity (DeRieux et al., 2018). It suggests the phase transition RH would be higher when higher fractions of anthropogenic HOA and BBOA were involved in the particle.

Furthermore, we estimated the mixing time τ_{mix} (right axis in Fig. 11a) to roughly clarify the time scale of organic molecules diffusion in 200 nm OA (Seinfeld and Pandis, 2016; Li and Shiraiwa, 2019), which can be compared with other studies conveniently. The results suggest that the mixing time of organic molecules in 200 nm OA varies dramatically from minutes at night to years in the afternoon. This large diurnal variation of OA mixing time might have significant impacts on aerosol evolution, such as gas-particle partitioning, multiphase chemical reactions, and nucleation, associated with the atmospheric fate of aerosols (Price et al., 2016; Shiraiwa et al., 2011; Gržinić et al., 2015). The higher mixing time during the day indicates that the kinetic limitation of diffusion after aging processes may impede gas-particle partitioning, thus SOA formation and growth in ambient air (Xu et al., 2021; Galeazzo et al., 2021; Li and Shiraiwa, 2019). This result emphasizes the necessity of considering kinetical limit processes in the model, otherwise, the LVOC in the particle phase might be overpredicted (Renbaum-Wolff et al., 2013).

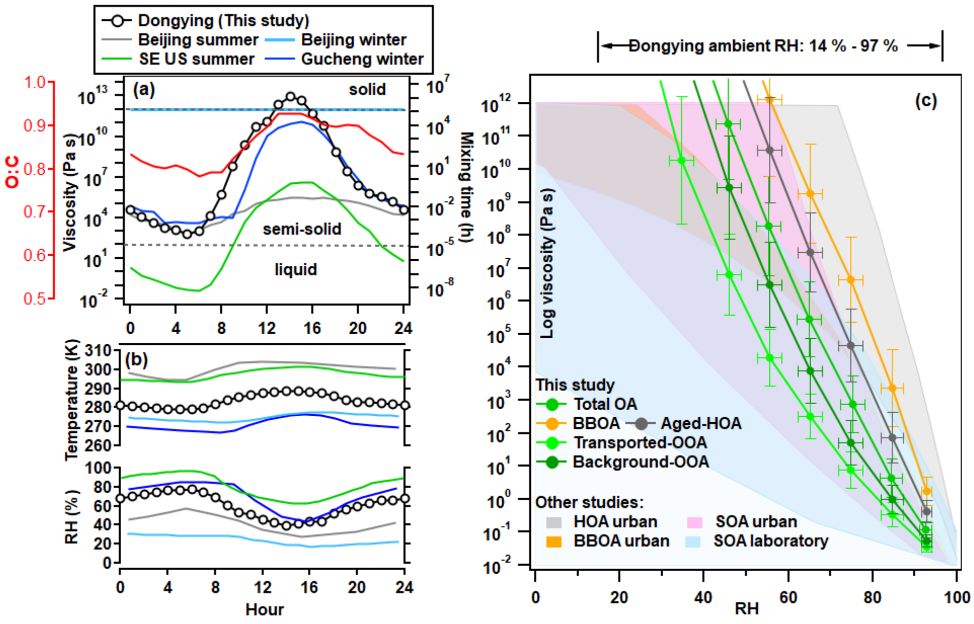

Figure 11. Average diurnal variations of (a) estimated viscosity of total OA, (b) ambient RH and temperature in different studies; In addition to this study, Beijing and Gucheng reported by Xu et al. (2021) and southeastern United States (SE US) reported by Li et al. (2020b) were also shown. The mixing time assuming the particle size is 200 nm was also displayed in the right axis of panel (a) here. (c) Predicted viscosity of total OA, aged-HOA, BBOA, transported-OOA, and background-OOA as a function of RH. The ambient and laboratory viscosity results (Li et al., 2020b; DeRieux et al., 2018) were also tinted in the background as a reference.

4 Conclusion

To characterize the aging impact on sources, volatility, and viscosity of OA during the spring of 2018, a TD-AMS system was deployed in Dongying, a regional receptor site in North China Plain. The average PM_1 concentration is 31.5 ± 22.7 $\mu g\ m^{-3}$, of which secondary aerosols (inorganic + organic species) dominate in the entire campaign, accounting for nearly 84 % on average, which suggests the aging process plays an important role in the NCP urban outflows. Secondary inorganic aerosols occupy 67 % of PM_1, which are mainly affected by south winds from urban areas in the NCP. Among secondary inorganic aerosols, nitrate accounts for the most (33 % of total PM_1), suggesting that nitrate and its precursor (NO_2) have become the primary pollutants in the NCP area, which shall be considered in the regional pollution control in China.

Similar to previous studies (Xu et al., 2016; Cao et al., 2018; Huffman et al., 2009a; Xu et al., 2019), OA, as an integration of different types of organic species, is moderately volatile in this study, where nitrate is the most volatile species and sulfate is the least. Ambient inorganic aerosols all show higher residuals upon heating at higher TD temperatures than their laboratory-derived pure standards, indicating the universal existence of low volatile metallic or organic-related species in ambient aerosols. This supports the complexity of ambient aerosols. Source apportionment of OA resolved four OA factors, BBOA (5 %), aged-HOA (28 %), transported-OOA (33 %), and background-OOA (33 %). BBOA, which is considered as emitted from the open fire nearby, is the most volatile in this study (average C* = 0.012 μg m^{-3}). Aged-HOA is characterized by high proportions of $C_xH_y^+$ ions and has good correlations with primary VOC species (benzene and ethyl toluene), as well as a pronounced peak of diurnal variations in the morning, suggesting that it is mainly transported from vehicle emissions from surrounding towns. The volatility of aged-HOA has been proven to be impacted by the aging process, which has lower volatility (average C* = 8.1×10^{-4} μg m^{-3}) than those of urban areas in NCP (Beijing), however still shows high SOA formation potential. In this study, total OOA (transported-OOA + background-OOA) accounts for the largest fractions of the OA (66 %) due to long-distance aging and fewer effects from local emissions than those of urban areas. Generally, transported-OOA (average C* = 3.7×10^{-4} μg m^{-3}) shows good agreements with secondary inorganic species, supporting their similar origins. Background-OOA shows the background concentration of OA in the outflow of the Chinese continent, which is around 2.6 μg m^{-3} on average. The background-OA is the least volatile factor in this study (average C* = 2.7×10^{-5} μg m^{-3}), implying the existence of a large fraction of oligomers in this type of SOA and shall be further investigated. The results suggest the aging processes during the transport of air plumes can significantly enhance the SOA formation and thus, decrease the OA volatility from different sources.

The glass transition temperature ($T_{g,org}$) and RH-dependent viscosity (η) of OA were predicted based on volatility distribution. The $T_{g,org}$ of total OA under dry conditions is 332.7 K, which is in the range of values from other campaigns, however much higher than those in urban NCP due to the low volatility of OA in our study (72 % of OA are non-volatile species). The viscosity of OA varies from 10^{-2} to more than 10^{12} Pa s, which mainly exist as semisolid particles. The solid phase usually occurs in the afternoon time when the oxidation of OA is higher and RH is low. When the ambient RH is above 80 %, OA exists as liquid aerosols. Phase transition RH would be higher when higher fractions of anthropogenic HOA and BBOA are involved in the particle. The dynamic range of OA viscosity results in a wide variation in the mixing time. The mixing time of organic molecules in 200 nm OA ranges from 10^{-4} to 10^{6} h, suggesting a large timescale to reach equilibrium. This emphasizes the necessity to consider dynamic kinetic limits of OA viscosity when modeling OA evolutions. Note that the OA viscosity estimated here is mainly derived based on a method from the volatility of pure organic compounds. there are also other factors that can influence OA viscosity in reality, e.g., OA functional group or molecular structure and organic/inorganic mixing state (Li et al., 2020b; Ditto et al., 2019), which shall be further investigated in the future. Overall, our results reveal how the aging process in the continental outflow can significantly influence the source and physiochemistry properties of OA, which shall be considered in the modeling and policy-making work regarding to evaluating its environment impact in the future.

Data availability. The data shown in the paper are available on request from the corresponding author (weiweihu@gig.ac.cn)

Author contributions. WH, WS, YS, designed the research. MZ, ZF, WD, XY, BY, SH, and HF conducted the field measurements. TF, YW, WH, YL, LH, CW, WC, MZ, ZF, and HF analyzed the data. TT and WC conducted the laboratory experiment on volatility. AP and FC supported the OA source analysis. WH and WC supported the viscosity analysis. TF and YW wrote the paper. WH, WC, CW, XH, LH, MS, XW, and AP reviewed and commented on the paper.

Competing interests. The authors declare that they have no conflict of interest.

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