

Responses to Reviewer #1

We appreciate the reviewer for his/her thorough reading and thoughtful comments and suggestions, which greatly improve the quality of the manuscript. We revised the MS accordingly. The point-to-point responses to all the comments are given below in blue.

This paper investigates the vertical distribution of organic acids with altitude in Beijing based on measurements from a tower. The results show slightly higher concentrations of oxidized species aloft, not too surprising if the source are mainly from surface emissions which would mean less aged and less oxygenated. The data analysis is very detailed, but overall the results are only mildly interesting despite the measurements being very unique. This is because really no definite results are presented. The analysis is largely based on looking at ratios of species to infer processes or sources, and the interpretation of the results are always highly qualified with words such as, might, and especially may. I wonder why there are not plots of altitude vs concentration in this paper, since that is fundamentally what the measurements were all about. It is not clear what the main contribution of the PMF source apportionment analyses is to the overall results of this paper on species vertical distributions; why was source apportionment not done at each altitude, was it because of insufficient data? The paper needs to be edited to improve the grammar. Overall, the data in this paper are interesting, but the analysis is very weak.

Response: We thank the reviewer's careful reading and important comments, based on which we tried our best to improve the quality of our manuscript.

The ratios of organic compounds can be effectively applied to assessing the photochemical aging level and relative importance of primary emissions, because the ratio values vary with different sources and photochemical aging level of aerosols. Saxena and Hildemann (1996) and Seinfeld and Pandis (2006) reported that the fresh anthropogenic organic aerosols are mainly comprised by organic compounds with low oxidation level and hydrophobicity, such as alkanes, whereas aged particles and secondary organic aerosols have more hygroscopic compounds with oxygenated polar functional groups. The proportion of hydrophobic compounds in atmospheric particles decreases progressively when moving from the urban background to continental and to remote marine locations. Simultaneously, a population of more hygroscopic particles is always present (Kanakidou et al., 2005; Seinfeld and Pandis, 2006). For instance, the relative contents of total dicarboxylic acids in aerosol organic carbon (total diacids-C/OC) in remote

marine and polar areas ($\geq 10\%$) is higher than those in urban atmospheres (1 – 3%) (Kawamura and Sakaguchi, 1999; Wang et al., 2006; Bikkina et al., 2015; Zhang et al., 2016).

Figure S3 has been redrawn for concentration variations and vertical properties of organic acids in the supporting information. Owing to the insufficient data, the PMF analysis run at each altitude existed high uncertainties, thus the PMF source apportionment analysis is to the overall results in this paper. The sample numbers will be considered to increase at each altitude. Simultaneously, more analyses will be employed to the aerosol samples to better estimate the relative contribution of sources and secondary process at different height.

This paper firstly shows the vertical distribution of organic compounds at the molecular level in megacity in China, investigates the sources and possible formation routes in clean and polluted days, and demonstrates the feedback of diacids and related compounds on the reduction of anthropogenic emissions. Such measurements in the troposphere are also critical for estimating the regional transport to air quality in Beijing and improving the simulation of aerosols in chemical transport models, which needs to be investigated in the future to give more precise conclusions. Moreover, the synchronous measurements, like Chemical Ionization Mass Spectrometer, is considered to use in combination with the sampling of diacids, oxoacids and α -dicarbonyls in gas and particle phase to better understand the gas-particle transformation of these acids at different heights.

Pg 3 lines 12-13. Needs to be edited.

Response: we have rephrased these sentences. Please see lines 12 – 15 in page 3 in the revised manuscript.

Please make clear that the high volume samplers were located at the elevation stated. That is, long sampling lines were not used in this study.

Response: The samplers were located directly at the three layers, that is, the ground surface (8 m), 120 m, and 260 m at the 325-m meteorological tower. We have rephrased the sentences in the revised manuscript. Please see lines 18 – 19 in page 4.

What type of filter was used, quartz, Teflon? Were the samples gas denuded (apparently not). There should be a discussion on possible artifacts associated with this sampling system, such as loss of the small acids due to evaporation, etc. Maybe this is why no light organic acids (eg, formate, acetate) are reported?

Response: We thank for the reviewer's suggestion. PM_{2.5} samples were collected on quartz-fiber filters (Pallflex). Please see lines 17 – 18 in page 4, where contains the filter information. Aerosol sampling onto quartz-fiber filters is accompanied by positive (e.g., adsorption of organic vapors) and/or negative (e.g., volatilization of organic aerosols after sample collection) artifacts. The collected particulate matter may chemically react with vapor components that pass through the filter during sampling to change the chemical composition of the deposit. This can result in positive or negative artifacts, especially if individual chemical compounds or chemical functionalities are being measured (Bennett and Stockburger, 1994). These sampling artifacts complicate the organic aerosol measurements, and it is difficult to separate the different competing artifact processes although this has been the object of many studies.

The positive artifact, as indicated by field blanks and backup filters, is believed to exceed the negative artifact for most samples (ten Brink et al., 2004; Watson et al., 2009). Without blank or backup filter subtraction, the artifact inflates organic carbon concentrations (Chow et al., 2010). Composition of the adsorbed/desorbed material, its exchange between gas and particle phases, the degree to which filters become saturated, and how the sign and amount of artifact differ among filter media and sampling environments have been studied, but these issues are not well understood (ten Brink et al., 2004; Watson et al., 2009; Vecchi et al., 2009; Arp et al., 2007; Zhu et al., 2012). In studies where the investigators attempted to measure the sampling error, the magnitudes of the determined positive or negative artifacts have varied considerably (Zhu et al., 2012; Liu et al., 2014a; Cheng et al., 2010a; Cheng et al., 2010b). This is probably due, in part, to differences in the organic molecular composition (hence in the relative volatilities and sorptivities) and in the concentrations that occurred at the different sampling locations. Multiple component sampling systems employing vapor phase separation devices such as denuders require a thorough evaluation to demonstrate efficient removal of the vapor at the operating conditions. This has been done for specific PAH compounds (Liu et al., 2014b; Schauer et al., 2003; Tsapakis and Stephanou, 2003; Liu et al., 2006). However, the application of their multiple component system to the determination of total OC aerosol for mixtures of atmospheric organic compounds presents a formidable challenge. The use of paired, parallel samplers (denuder difference method) is subject to the propagation of errors through subtraction, which degrades the precision. More development work with control experiments needs to be done on a wide range of organic compounds and concentrations before the sampling approach becomes credible for extended field studies.

Based on PM₁₀ aerosols collected using a high-volume sampler with two quartz fiber filters,

Ray and McDow (2005) found that dicarboxylic acids are affected by significant sampling artifact errors at concentrations below thresholds ($0.5 \text{ ng m}^{-3} \leq \text{artifact thresholds} \leq 12 \text{ ng m}^{-3}$), but sampling artifacts are considerably lower at high concentrations expected when their primary emission sources and/or secondary formation are most important. Sihabut et al. (2005) also reported that the amount of dicarboxylic acids ($\text{C}_3 - \text{C}_9$) collected on the backup filter was consistently a small fraction of the amount collected on the front filter.

Due to the large contribution of primary emissions and secondary formation pathways to organic aerosols in Beijing, the concentrations of dicarboxylic acids detected in our study were larger than the artifact thresholds determined by Ray and McDow (2005). So far, most studies use single filter to collect organic and inorganic compounds in particulate aerosols (Wang et al., 2012; Zhang et al., 2015; Huang et al., 2014; Shi et al., 2018; Lyu et al., 2019; Mikhailov et al., 2017; Cheng et al., 2014; Ding et al., 2016; Cheng et al., 2016; Andreae et al., 2012; Saturno et al., 2018; Hsieh et al., 2008; Sanderson and Farant, 2005; Yue et al., 2019; Wang et al., 2016; Wang et al., 2017; An et al., 2019; Elser et al., 2016; Li et al., 2018; Kang et al., 2018; Liang et al., 2020; Zhang et al., 2016; Zhang et al., 2017; Wu et al., 2019; Zotter et al., 2014).

As for monocarboxylic acids, formic (HCOOH) and acetic (CH_3COOH) acids are among the most abundant and ubiquitous trace gases in the atmosphere (Khare et al., 1999; Talbot et al., 1988), and they have also been detected in remote, rural, polar, marine and urban aerosols, clouds and rain water (Chang et al., 2019; Andreae et al., 1988; Stavrou et al., 2012; Paulot et al., 2011; Chattopadhyay et al., 2015; Chameides and Davis, 1983; Herndon et al., 2007; Zhang et al., 2011; Trentmann et al., 2005; Liu et al., 2012; Liang et al., 2020; Kesselmeier et al., 1998; Wang et al., 2007; Willey and Wilson, 1993; Avery et al., 2001; Xu et al., 2009; Kumar et al., 1996).

Our further work will focus on calculating the concentration thresholds of organic compounds in different locations and conditions and will try to find out more suitable sampling techniques to better understand the relationship between observed thresholds to adsorptive vapor concentrations and filter adsorption capacity for better evaluating the contribution of water-soluble organic acids to organic aerosol.

Page 7 lines 13 to 14. Stating that there is no direct analytical route to measure SOC may be strictly true, but there are ways that get pretty close, such as AMS measurements with PMF analysis. The statements and method to determine SOC in this paper are like reading a paper about 10 to 15 years old. Note the dates for the references for the method are 1995 and 1999.

The reason this method is rarely used is that it is highly inaccurate. Limitations with the method need to be discussed.

Response: Several indirect methods are widely employed to estimate the concentrations of primary and secondary organic aerosols, which can be categorized as the EC tracer method (Turpin and Huntzicker, 1995; Castro et al., 1999; Lim and Turpin, 2002; Chu, 2005; Yu et al., 2009), receptor modeling (Zheng et al., 2002; Na et al., 2004; Schauer et al., 2007) and chemical transport modeling (Pandis et al., 1992; Johnson et al., 2006b; Johnson et al., 2006a). Among these, the EC tracer method has received the widest application due to its simplicity and reliance on ambient measurements (Chu, 2005; Yu et al., 2009; Sun et al., 2011; Ji et al., 2016; Amato et al., 2011; Bougiatioti et al., 2013; Day et al., 2015; Grivas et al., 2012; Harrison and Yin, 2008; Kim et al., 2012; Kumar et al., 2012; Lin et al., 2009; Lonati et al., 2007; Samara et al., 2014; Saylor et al., 2006; Seguel A et al., 2009; Takegawa et al., 2006; Weber et al., 2007; Wu and Yu, 2016; Zhang et al., 2005; Zhang et al., 2014; Zhao et al., 2013; Zhou et al., 2012; Pio et al., 2011).

The EC tracer method is used here to derive POC and SOC empirically. The assumptions and methodology of EC tracer method are described in detail elsewhere (Castro et al., 1999; Turpin and Huntzicker, 1991, 1995; Yu et al., 2007). Briefly, total OC is defined as the sum of POC and SOC. By this method, the POC concentration can be defined as

$$\text{POC} = (\text{OC}/\text{EC})_{\text{pri}} \times \text{EC} + c, \quad (1)$$

where $(\text{OC}/\text{EC})_{\text{pri}}$ is the estimated primary OC/EC ratio and c is to account for non-combustion sources contributing to POC (Turpin and Huntzicker, 1995; Castro et al., 1999). The SOC concentration can be estimated as

$$\text{SOC} = \text{OC} - \text{POC} \quad (2)$$

Previous studies often summarized the linear least-squares fit results of OC vs. EC from the lowest 5% and 10% OC/EC values to estimate $(\text{OC}/\text{EC})_{\text{pri}}$ (Sun et al., 2011; Miyazaki et al., 2006; Chu, 2005; Day et al., 2015; Ji et al., 2016), which is seasonally-dependent (Yuan et al., 2006). Therefore, the $(\text{OC}/\text{EC})_{\text{pri}}$ should be calculated at each sampling height (8 m, 120 m and 260 m) in our study due to the different influence strength of primary sources and secondary formation pathways. But it is worth mentioning that the number of PM_{2.5} samples collected at each sampling height were less than 30 in our study, which means that there is no statistical significance for the $(\text{OC}/\text{EC})_{\text{pri}}$ estimation from the lowest 5% and 10% OC/EC values. The $(\text{OC}/\text{EC})_{\text{pri}}$ is 0.21, 0.14 and 0.24 calculated from the lowest 20% OC/EC values at the ground surface, 120 m and 260 m, respectively (Fig. 1). Based on these $(\text{OC}/\text{EC})_{\text{pri}}$ values, the possible

uncertainties ($\pm 138\%$ – $\pm 207\%$) largely vary for the estimated SOC. Meanwhile, these $(OC/EC)_{pri}$ ratios are not comparable to those (1.9 – 2.8) based on the lowest 5% OC/EC values in four seasons in Beijing, China (Ho et al., 2007; Ji et al., 2016).

The direct emission from vegetation, the major non-combustion source, contributed 3.8% to OC in total suspended particles in summer in Beijing (Li et al., 2018), so OC emitted from non-combustion sources is assumed to be negligible in the approach used here. Using the minimum OC to EC ratio, $(OC/EC)_{min}$, to substitute for $(OC/EC)_{pri}$, the SOC and POC can therefore be estimated (Yu et al., 2009; Cabada et al., 2004; Castro et al., 1999). The $(OC/EC)_{min}$ ratios were 5.0, 2.6 and 2.5 at the ground level, 120 m and 260 m in summer in Beijing, respectively. The sampling site is a typical urban location largely influenced by traffic and cooking emissions in Beijing, especially at the ground surface (Zhou et al., 2018b; Zhou et al., 2018a). Additionally, cooking emission also correspond to high OC/EC ratios (Samara et al., 2014). Thus, the $(OC/EC)_{min}$ ratio at the ground surface (5.0) was larger than those at 120 m (2.6) and 260 m (2.5) in our study and those (1.9 – 2.8) detected in other sampling sites in Beijing (Ho et al., 2007; Ji et al., 2016). It is rational to use the minimum OC/EC ratio to estimate the SOC and POC concentrations in this paper.

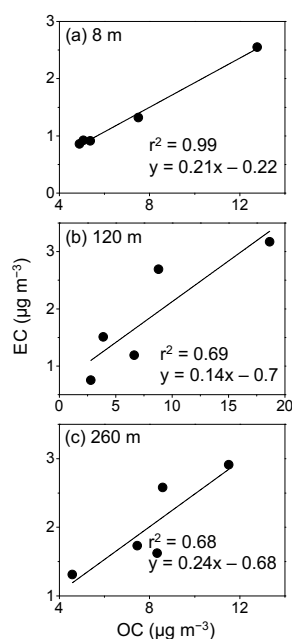


Figure 1. The linear least-squares analysis fitting results grouped by the lowest 20% of OC/EC ratios at the ground level, 120 m and 260 m

Based on AMS online data, the PMF result does get closer to the true SOC value, but this method is not widely used and is beyond the scope of our study. Because our group focus on the offline analyses of aerosol chemistry at the molecular level.

The 1995 and 1999 references cited for the EC tracer method in our study are the most classic literatures, which have been widely cited. Please see the lines 18 – 19 in page 7, where we have updated the references.

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