

Response to Reviewer 1

Overall comment: Yu et al reports observations of organic aerosol, both primary and secondary, collected on filters for different seasons/time periods of 2017 in Beijing, China. They report water soluble organic carbon (WSOC), its hydrophobic and hydrophilic portions, water soluble ions, total PM_{2.5}, total organic carbon (OC), and total elemental carbon (EC). Further, they report tracers associated to different sources (levoglucosan, cholesterol, phthalic acid, 4-methyl-5-nitrocatechol, 2-methylerythritol, 3-hydroxyglutaric acid, and *cis*-pinonic acid). They use the tracers to differentiate sources of OC and WSOC during the seasons via "CO-scaled" concentrations, day and night ratios, correlation coefficients with various meteorological and chemical properties of aerosol ("acidity" and liquid water content), and positive matrix factorization. They find that aqueous chemistry explains a large portion of the secondary organic carbon during most seasons except summer, where photochemistry explains an important biogenic portion. They also find differences in the sources between the seasons (biomass burning vs dust vs vegetation). Overall, the paper is important and of interest to Atmospheric Chemistry and Physics community as there is general overall uncertainty in the sources of organic aerosol in urban environments, especially during all seasons and high pollution events. The paper will be of value once the authors address the comments below.

Response: We deeply appreciate the reviewer's time and efforts devoted to improving this manuscript. We have provided our responses point by point below each comment, and have carefully revised the paper according to the reviewer's valuable suggestions.

1. Section 2.1 Sampling: Further information is needed here for the readers to have a better understanding of how the aerosol was collected—Was there a drier in-line prior to be collected on the filters? Was there a denuder to scrub gases prior to the filter to minimize gas-particle partitioning? Was there an impactor or cyclone for size selection? Further, of importance, was there any analysis of potential reactions that occurred on the filters prior to sampling?

Response: Thanks for the reviewer's comment. Our sampler included a PM_{2.5} impactor, but no in-line drier or denuder was used in this study.

Sampling of organic carbon is accompanied by both positive and negative artifacts. The positive artifact is due to the adsorption of gaseous organics to the sampling filter, and the negative artifact is caused by the evaporation of collected particulate organic carbon. To eliminate the positive artifact, a denuder can be placed upstream the sample filter to remove the gaseous organics by diffusion to the adsorbent surface (Cheng et al., 2009). The use of a denuder in the sampling system has been reported in previous studies (Eatough et al., 1993, 1999; Mader et al., 2001; Matsumoto et al., 2003; Viana et al., 2006; Cheng et al., 2009, 2010, 2012; Kristensen et al., 2016). The use of a denuder may induce a larger negative artifact, however, as the removal of gaseous organics can enhance the evaporation of particulate OC. Thus a backup filter should also be included in the sampling system (Cheng et al., 2009). Besides, the flow rate passing through the denuder was very low in most studies (Matsumoto et al., 2003; Viana et al., 2006; Cheng et al., 2009, 2010, 2012; Kristensen et al., 2016). This might be due to the significantly decreased removal efficiency of the denuder as the air flow rate increased (Cui et al., 1998; Ding et al., 2002). To collect enough samples for the accurate measurement of trace organic species, the flow rate of 1.05 m³ min⁻¹ was chosen in this study. The air flow rate of about 1.05 m³ min⁻¹ has been frequently used in the field sampling of organic aerosols (Kawamura et al., 2013; Verma et al., 2012, 2015; Li et al., 2018; Ma et al., 2018; Huang et al., 2020). At this flow rate, a denuder with a high removal efficiency is hardly commercially available.

Nevertheless, we were aware of the potential sampling artifacts and attempted to estimate the sampling artifacts of OC based on the literature results. Firstly, the adsorption behavior of OC might vary with meteorological conditions. Besides, the OC fractions with different volatility show different adsorption behavior. Cheng et al. (2015) compared the concentrations of different OC fractions (OC1, OC2, OC3, OC4) on bare quartz filters with those on denuded quartz filters in the four seasons of Beijing, and the results are summarized in Table S1. The contributions of different OC

fractions measured in this study are also listed in Table S1.

Table S1 The ratio of the OC concentrations on the bare quartz filters to those on the denuded quartz filters in Cheng et al. (2015), as well as the contribution of different OC fractions measured in this study.

	The ratio of OC on bare quartz filters to denuded quartz filters (Cheng et al., 2015)				The contribution of different OC fractions measured in this study			
	OC1	OC2	OC3	OC4	OC1	OC2	OC3	OC4
Winter	1.27	1.03	1.02	1.05	10.8 %	19.6 %	24.7 %	44.8 %
Spring	2.05	1.05	1.00	1.01	3.9 %	27.2 %	43.1 %	25.7 %
Summer	2.45	1.60	1.17	1.08	4.4 %	37.6 %	36.0 %	22.0 %
Autumn	2.08	1.05	0.99	1.01	7.9 %	26.5 %	40.2 %	25.3 %

McDow (1986) systematically investigated the effect of sampling procedure on the OC measurement. The adsorption of organic vapors on bare quartz filters ($C_{positive\ artifact}$) was a function of the sampling duration (t) multiplied by the face velocity (v) as follows:

$$C_{positive\ artifact} = \sum_i \rho_i \frac{1 - e^{-\varepsilon_i vt}}{\varepsilon_i vt} \quad (1)$$

where the face velocity (v , cm s^{-1}) is the ratio of the flow rate ($\text{cm}^3 \text{s}^{-1}$) to the sampling area of the filter (cm^2), ρ_i is the concentration of adsorptive vapor i (g cm^{-3}), and ε_i is a constant which can be defined as:

$$\varepsilon_i = \frac{1}{l} \left[\beta f(t) t_o \exp\left(\frac{Q_A}{RT}\right) + 1 \right] \quad (2)$$

where l is the effective filter thickness. The average thickness of the quartz filter used in this study was 463 μm . The other parameters are all constants.

Hence, it can be calculated that $\varepsilon_i > 1/l > 20 \text{ cm}^{-1}$, and $1 - e^{-\varepsilon_i vt} \approx 1$. Therefore, the positive artifact ($C_{positive\ artifact}$) is inversely proportional to the product of the sampling duration and the face velocity ($v \times t$). The face velocity of Cheng et al. (2015) was 9.8 cm s^{-1} , while that in our study was 47.3 cm s^{-1} . The sampling duration of Cheng et al. (2015) was 24 h, while that in our study was 12 h. That is to say, the positive artifact of Cheng et al. (2015) was about 2.4 times higher than that in our study.

Based on the literature results and taking into account all the factors (seasons,

OC fractions, sampling procedure), the contribution of positive artifact to the measured OC was estimated to be 2.3 %, 1.4 %, 9.9 %, and 2.2 % in winter, spring, summer and autumn respectively in this study, which is roughly acceptable.

To further estimate the impact of gas-particle partitioning and potential reactions occurring on the filters, we overlapped two quartz filters and took samples at a flow rate of 1.05 m³ min⁻¹ for a duration of 12 h. The organic tracers selected in this study were measured in both filters. The organic tracers on the backup filters typically originate from three sources: (1) adsorption of the organic vapors in the atmosphere; (2) adsorption of the semi-volatile species evaporated from the front filter; (3) secondary formation from the adsorbed organic vapors on the backup filter. Except for *cis*-pinonic acid, the tracer concentrations on the backup filter were all less than 5 % of those on the front filters, while the concentration of *cis*-pinonic acid on the backup filter was 21.6 % of that on the front filter. This result suggested that the sampling procedure in this study might bring some uncertainties for the measurement of *cis*-pinonic acid, and the sampling artifact was not significant for the other organic tracers.

References

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2. Line 121: It is unclear what the standards curves were of (the tracers reported throughout paper or other standards), and what is meant by "standard curves with five to seven concentration gradients were re-established." What was re-established?

Response: Thanks for the reviewer's comment. The standard curves were made by the silylation derivatives of the organic tracers, and the detailed information of these standards is listed in Table S2. "Re-established" means that we measured the derivative products of the standard solutions each time before measuring the ambient samples. We have avoided the unclear expression and revised as follows:

“The authentic standards (Table S2) were dissolved in anhydrous pyridine, and diluted to five to seven different concentrations. Then 100 μ L of the standard solutions were reacted with 200 μ L silylating reagent (BSTFA: TMCS = 99:1) at 75°C for a duration of 70 min. After cooling down to the ambient temperature, these solutions containing derivative products were diluted to 1 mL with n-hexane, and measured by GC/MS/MS right before the analysis of ambient samples. The R^2 of the derivative products were above 0.99, indicating good linearities of these standard curves. ”

3. Line 122: What authentic standards? What company/purity?

Response: The information of the authentic standards is provided in Table S2 below.

Table S2 The detailed information of the authentic standards used in this study.

Authentic standard	Molecular formula	CAS number	Company	Purity
Levogluconan	C ₆ H ₁₀ O ₅	498-07-7	Sigma-Aldrich	99 %
Cholesterol	C ₂₇ H ₄₆ O	57-88-5	Sigma-Aldrich	93 %
Phthalic acid	C ₈ H ₆ O ₄	88-99-3	Sigma-Aldrich	99 %
4-Methyl-5-nitrocatechol	C ₇ H ₇ NO ₄	68906-21-8	Toronto Research Chemicals	98 %
2-Methylerythritol	C ₅ H ₁₂ O ₄	58698-37-6	Sigma-Aldrich	90 %
3-Hydroxyglutaric acid	C ₅ H ₈ O ₅	638-18-6	Sigma-Aldrich	95 %
cis-Pinonic acid	C ₁₀ H ₁₆ O ₃	61826-55-9	Sigma-Aldrich	98 %

4. Section 2.3: Please describe or cite the PMF software used. PMF 5.0 is not enough to understand how positive matrix factorization was actually conducted.

Response: Thanks for this kind suggestion. We have added a brief introduction to the PMF software in the manuscript. Besides, we have added the detailed information on the uncertainty calculation of the input data, the selection criteria for the optimal solution, the diagnostic plots and error estimation in the supplementary material.

5. Line 140: I highly recommend the use of "aerosol acidity," as defined in this line, due to the discussion from Pye et al. 2020 (<https://acp.copernicus.org/articles/20/4809/2020/>). The ratio here does not define acidity, and is analytically challenging to say if it is defining the amount of hydronium

ions in the aerosol phase, as the hydronium ions may be a very low detection limits that cannot be quantified due to propagation of uncertainty.

Response: We deeply appreciate the reviewer's valuable suggestion. According to Pye et al. (2020), we changed the ratio of R_{AC} to the approximate value of aerosol pH (pH_F) to denote the aerosol acidity. The pH_F value was estimated using the ISORROPIA-II model. And the molality of H^+ (m_{H^+}), which was calculated by $m_{H^+} = 10^{-pH_F}$, was used instead for the correlation analysis in the revised manuscript.

6. Section 3: Though an important and valuable aspect of this whole manuscript is that the filters were collected during different seasons, I highly recommend the authors soften the language throughout that the results "reflect" a specific season or are similar or different to other studies. Since it's only for one year and approximately 2 weeks for each season. The limited data makes it hard to say how typical the results are and this should be discussed/emphasized throughout (instead of general statements that in fall this is what is observed/happens).

Response: Thanks for the reviewer's valuable suggestion. We have changed the terms of "winter/spring/summer/autumn" to "January/April/July/October" or "during the sampling periods in winter/spring/summer/autumn" throughout the revised manuscript, in order to be more specific.

7. Another area I suggest the authors be careful in their discussion of r values, as majority of the values they report lead to R^2 values less than 0.5 (thus explaining less than 50% of the variability observed).

Response: We agree with the reviewer that most of the r values led to R^2 less than 0.5, thus could only explain less than 50 % of the variability observed, and were insufficient to reach clear conclusions. Therefore, in the revised manuscript, we only explained the r results and avoided making definite conclusions based on the r values.

8. Line 195: Since the authors are comparing OC from emissions inventory to Fig. 2, I would recommend converting the emissions to OC-to-CO ratios. Also, I would

recommend adding these ratios, if possible, to Fig. 2, for direct comparisons with observations.

Response: We are deeply grateful for the reviewer’s valuable suggestion. As suggested by the reviewer, we have added the emissions of OC from the Fire Inventory to Figure 2 in the revised manuscript. As a tracer for biomass burning, levoglucosan in the atmosphere may derive from both residential biofuel burning and open biomass burning. To determine whether open biomass burning was the dominant type of biomass burning in Beijing, the seasonal variation of the CO-scaled concentration of levoglucosan was compared with that of the OC emission amount from the Fire Inventory in this study. The CO-scaled concentration of levoglucosan showed a totally different seasonal variation trend from that of the OC emission from open biomass burning, therefore, we speculated that open biomass burning was not the dominant category of biomass burning in Beijing. After careful consideration, we thought that it was not necessary to convert the emissions to OC-to-CO ratios. Instead, the absolute emission amounts of OC from the Fire Inventory were added.

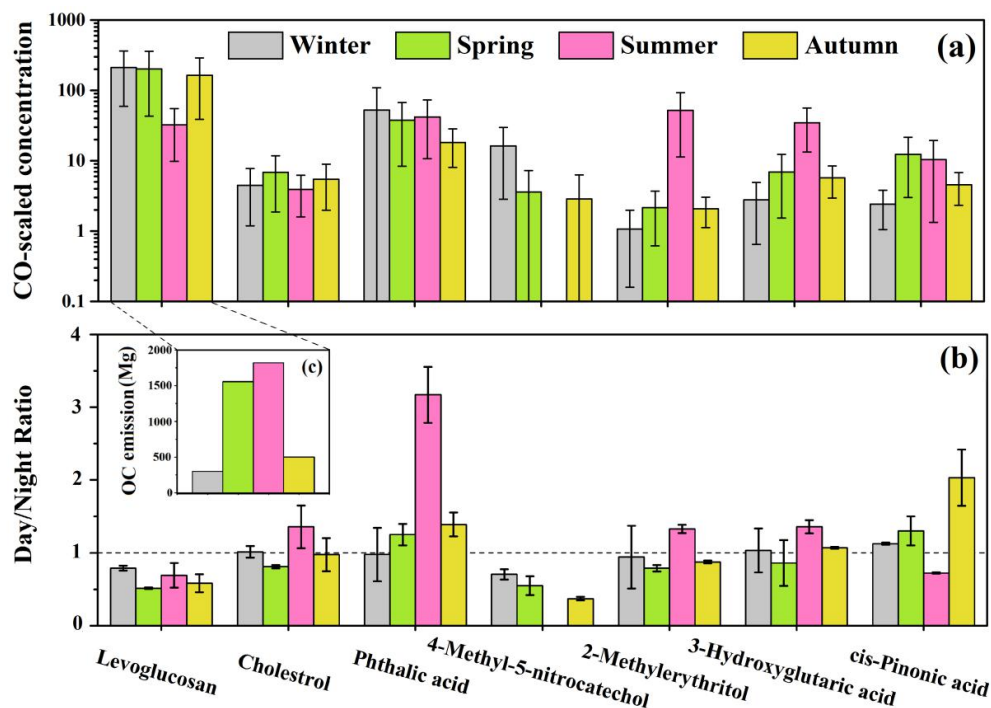


Figure 2. (a) The CO-scaled concentration of the identified organic tracers; (b) The day to night ratios of the measured concentrations of the organic tracers; (c) The OC emission amounts from open biomass burning provided by the Fire Inventory (FINN)

in Beijing during the sampling periods in four seasons of 2017.

9. Line 199: Is it possible to get emission inventory values of residential biofuel combustion and coal combustion to compare with the OC from open biomass burning?

Response: The total emission amounts of OC from the residential sources in January, April, July and October of 2016 can be obtained from the multi-resolution emission inventory for China (MEIC, <http://www.MEICmodel.org>). However, the residential OC from MEIC is a total amount of biofuel combustion, coal combustion, other fossil fuel combustion, etc. Unfortunately, the detailed OC emission amount from each residential source is not available. Therefore, the respective OC emission inventory values of residential biofuel combustion and coal combustion were not provided in the manuscript.

10. Line 207: It is unclear how aromatics form SOA to impact WSOC during winter, as the photochemistry is greatly reduced. Could the authors provide more discussion concerning this?

Response: We deeply appreciate the reviewer's valuable suggestion. The following discussion has been added in the revised manuscript.

Although the concentrations of oxidants were usually lower in winter due to the weaker solar radiation, a previous observation found that the $\cdot\text{OH}$ concentration in Beijing was significantly higher than that in New York, Birmingham and Tokyo, and was nearly 1 order of magnitude higher than that predicted by global models in northern China in winter (Tan et al., 2018). Zhang et al. (2019) indicated that HONO, which was mainly from the heterogeneous reactions of NO_2 and traffic emissions, was the major precursor of $\cdot\text{OH}$ in winter. According to the WRF-Chem model simulation, HONO resulted in a significant enhancement ($5\text{-}25 \mu\text{g m}^{-3}$) of SOA formation (most of which were from the aromatic precursors) during a haze episode in winter in the Beijing-Tianjin-Hebei region (Zhang et al., 2019). Besides, some recent studies suggested that the brown carbon-derived singlet molecular oxygen ($^1\text{O}_2^*$) in aerosol

liquid water could react rapidly with the electron-rich organics such as PAHs, thus facilitate the aromatic SOA formation (Kaur et al., 2019; Manfrin et al., 2019). This process might be more significant in winter, when the concentration of HULIS was much higher than that in other seasons. Therefore, both the enhanced levels of oxidants including $\cdot\text{OH}$ and $^1\text{O}_2^*$ and the higher concentrations of aromatic precursors in winter contributed to the enhanced aromatic SOA formation during the study period in winter.

References

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11. Line 249: It is surprising that the authors are saying that gas-phase photooxidation was not the dominant formation mechanism of secondary organic carbon. I can see maybe WSOC, but seeing all secondary organic carbon is a big statement. Especially, since the authors go on in line 254-55 to say photochemistry plays a role.

Response: We deeply appreciate the reviewer's valuable comment. We agree with the reviewer that it was not proper to conclude that "gas-phase photooxidation was not the dominant formation mechanism of SOC" merely based on the result that "WSOC/OC did not show any significant positive correlation with O_3 concentrations". The corresponding statement has been deleted in the revised manuscript. Besides, "SOC" has been corrected to "SOC in WSOC" to be more specific.

12. Line 339: Source 3 did not show the highest contribution in winter...highest contribution of what?

Response: We feel sorry for the unclear expression. This sentence has been revised as: “The primary emission strength of coal combustion was the strongest in winter among four seasons, since the domestic heating activities required extra amounts of coal combustion in this season. However, the contribution of Factor 3 to WSOC during the study period in winter was not the highest among four seasons, implying that there could be other sources beyond coal combustion included in Factor 3.

13. Line 416-419: I would recommend caution here, as other hypothesis have been stated for reasons in differences between chamber SOA and ambient SOA, including losses of vapors to the walls and autoxidation (which has been shut down in chamber experiments due to too high NO_x levels and/or too high aerosol loadings).

Response: We deeply appreciate the reviewer’s valuable suggestion. This statement has been revised as: “Previous observation suggested that a large fraction of ambient SOA was more oxidized than those generated in the dry smog chambers, where SOA could only be produced through gas-phase oxidation (Aiken et al., 2008). There have been some hypothesis for the difference between the chamber SOA and ambient SOA, such as the losses of vapors to the walls, and the autoxidation due to the uncertainties in chamber radical environment (McVay et al., 2016; Thornton et al., 2020). Besides, the results of this study also indicated that the aqueous-phase processing, which can produce more hydrophilic SOA, may be one of the reasons for the discrepancy in the oxidation degrees of ambient SOA and chamber SOA (Ervens et al., 2011).”

References

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14. Table 1: I would recommend somehow highlighting which values show statistical

differences between day and night and between seasons. Also, I would recommend including average CO mixing ratios.

Response: We deeply appreciate the reviewer's suggestion. The average values which showed statistical differences among seasons and between day and night have been highlighted as follows, and the average CO concentrations have also been included in Table 1 in the revised manuscript.

Table 1 The average concentrations and standard deviations of the identified carbonaceous species in PM_{2.5} during the sampling periods in four seasons.

Compounds ($\mu\text{g m}^{-3}$)	Winter			Spring			Summer			Autumn		
	Daytime	Nighttime	Mean	Daytime	Nighttime	Mean	Daytime	Nighttime	Mean	Daytime	Nighttime	Mean
CO	1.7 ± 1.7	2.2 ± 2.8	1.9 ± 2.3 ^a	0.6 ± 0.4*	0.8 ± 0.4*	0.7 ± 0.4 ^b	1.1 ± 0.4	0.9 ± 0.2	1.0 ± 0.3 ^b	1.1 ± 0.4*	1.4 ± 0.4*	1.2 ± 0.4 ^b
PM _{2.5}	120 ± 107	147 ± 154	133 ± 131 ^a	60.6 ± 36.2	64.5 ± 34.8	62.5 ± 34.9 ^b	59.8 ± 28.6	51.9 ± 20.6	55.8 ± 24.8 ^b	75.2 ± 58.1	81.1 ± 50.8	78.2 ± 53.7 ^b
OC	20.1 ± 19.2	21.0 ± 24.8	20.6 ± 21.9 ^a	7.9 ± 2.6	9.5 ± 3.4	8.7 ± 3.1 ^b	8.7 ± 3.4*	6.8 ± 4.3*	7.8 ± 3.9 ^b	9.4 ± 3.8	10.1 ± 3.7	9.7 ± 3.7 ^b
EC	3.9 ± 3.1	4.7 ± 5.8	4.3 ± 4.6 ^a	1.9 ± 1.1*	2.7 ± 1.4*	2.3 ± 1.3 ^{b, c}	1.4 ± 1.0	1.3 ± 1.0	1.3 ± 1.0 ^c	2.4 ± 1.4*	3.4 ± 1.7*	2.9 ± 1.6 ^b
OC/EC	4.6 ± 1.1	4.3 ± 1.3	4.5 ± 1.2 ^{a, b}	5.2 ± 2.1	4.4 ± 2.5	4.8 ± 2.3 ^{a, b}	6.7 ± 3.9	5.3 ± 4.2	6.1 ± 4.1 ^a	4.4 ± 1.6*	3.3 ± 0.9*	3.8 ± 1.4 ^b
WSOC	11.4 ± 11.3	12.0 ± 16.4	11.7 ± 13.9 ^a	4.1 ± 2.0	4.7 ± 2.6	4.4 ± 2.3 ^b	5.3 ± 2.1*	4.0 ± 2.7*	4.7 ± 2.5 ^b	4.7 ± 3.0	4.9 ± 2.8	4.8 ± 2.8 ^b
WSOC/OC	0.53 ± 0.08	0.51 ± 0.08	0.52 ± 0.08 ^b	0.50 ± 0.10	0.47 ± 0.14	0.49 ± 0.12 ^b	0.62 ± 0.11	0.59 ± 0.10	0.60 ± 0.11 ^a	0.47 ± 0.12	0.46 ± 0.12	0.46 ± 0.12 ^b
MH-WSOC	7.9 ± 7.6	8.0 ± 10.3	8.0 ± 8.9 ^a	2.8 ± 1.3	2.9 ± 1.6	2.9 ± 1.5 ^b	4.1 ± 1.2	3.4 ± 1.6	3.8 ± 1.5 ^b	2.9 ± 1.6	2.9 ± 1.3	2.9 ± 1.5 ^b
SH-WSOC	3.2 ± 3.8	4.0 ± 6.1	3.6 ± 5.0 ^a	1.3 ± 0.9	1.8 ± 1.1	1.6 ± 1.0 ^b	1.2 ± 1.0*	0.7 ± 1.1*	1.0 ± 1.1 ^b	1.8 ± 1.4	2.0 ± 1.5	1.9 ± 1.4 ^b
Organic tracers (ng m^{-3})												
Levoglucosan	307 ± 300	388 ± 394	349 ± 348 ^a	100 ± 87.8*	194 ± 175*	147 ± 144 ^b	23.6 ± 11.0	34.2 ± 24.2	28.9 ± 19.3 ^c	136 ± 102*	234 ± 125*	185 ± 123 ^b
Cholesterol	5.0 ± 3.0	4.9 ± 3.3	4.9 ± 3.1 ^{a, b}	3.9 ± 1.9	4.8 ± 2.5	4.3 ± 2.3 ^b	4.1 ± 2.4	3.0 ± 1.1	3.6 ± 1.9 ^b	6.1 ± 4.4	6.3 ± 3.1	6.2 ± 3.8 ^a
Phthalic acid	88.7 ± 84.8	90.8 ± 121	89.8 ± 103 ^a	27.3 ± 20.8	21.9 ± 14.0	24.6 ± 17.7 ^b	55.9 ± 22.0*	17.6 ± 9.1*	36.8 ± 25.5 ^b	27.6 ± 21.8	19.9 ± 13.3	23.8 ± 18.2 ^b
4-Methyl-5-nitrocatechol	24.7 ± 26.4	35.2 ± 41.0	30.1 ± 34.5 ^a	1.8 ± 1.9	3.3 ± 2.7	2.6 ± 2.4 ^b	0.1 ± 0.3	0.0 ± 0.0	0.1 ± 0.2 ^b	1.6 ± 1.2*	4.4 ± 3.6*	3.0 ± 3.0 ^b
2-Methylerythritol	2.1 ± 2.3	2.2 ± 3.5	2.2 ± 2.9 ^b	1.2 ± 0.6	1.5 ± 0.8	1.4 ± 0.7 ^b	55.4 ± 48.5	41.6 ± 34.6	48.5 ± 42.0 ^a	2.3 ± 1.1	2.6 ± 1.2	2.5 ± 1.1 ^b
3-Hydroxyglutaric acid	4.4 ± 3.9	4.2 ± 5.0	4.3 ± 4.5 ^b	4.2 ± 2.8	4.9 ± 5.1	4.6 ± 4.0 ^b	37.1 ± 22.7*	27.3 ± 18.5*	32.2 ± 20.9 ^a	7.5 ± 4.6	7.0 ± 4.4	7.2 ± 4.5 ^b
<i>cis</i> -Pinonic acid	3.3 ± 2.4	3.0 ± 2.1	3.2 ± 2.2 ^c	9.0 ± 6.0*	6.9 ± 3.6*	7.9 ± 5.0 ^a	7.3 ± 4.2	10.1 ± 6.0	8.7 ± 5.3 ^a	7.3 ± 3.0*	3.6 ± 0.8*	5.5 ± 2.9 ^b

^{a, b, c} We performed one-way analysis of variance (ANOVA) to evaluate whether these mean values showed statistically significant differences ($p < 0.05$) between two seasons. If two seasonal average values have one or more same superscripts, it means that they did not show significant differences ($p > 0.05$). In contrast, if two average values do not have any same superscript, it means that they showed significant differences ($p < 0.05$). For example, the PM_{2.5} concentration was significantly higher in winter than in other seasons, but it did not show significant difference between spring and summer, spring and autumn, or summer and autumn. Besides, the EC concentration did not show significant difference between spring and summer, as well as between spring and autumn, but it showed significant difference between summer and autumn.

* We also performed paired t test to evaluate whether daytime and nighttime values showed statistically significant differences ($p < 0.05$). The values with * as their superscripts showed statistically significant differences between day and night.

15. Fig. 1: I would recommend including a line that shows the average and standard deviation for the WSOC/OC values. Currently, eyeballing the values in Fig. 1, they look fairly similar in all seasons.

Response: We deeply appreciate this nice suggestion. As suggested by the reviewer, we have added four lines which represented the average WSOC/OC values in each season, and four shaded areas that showed the standard deviations of WSOC/OC values.

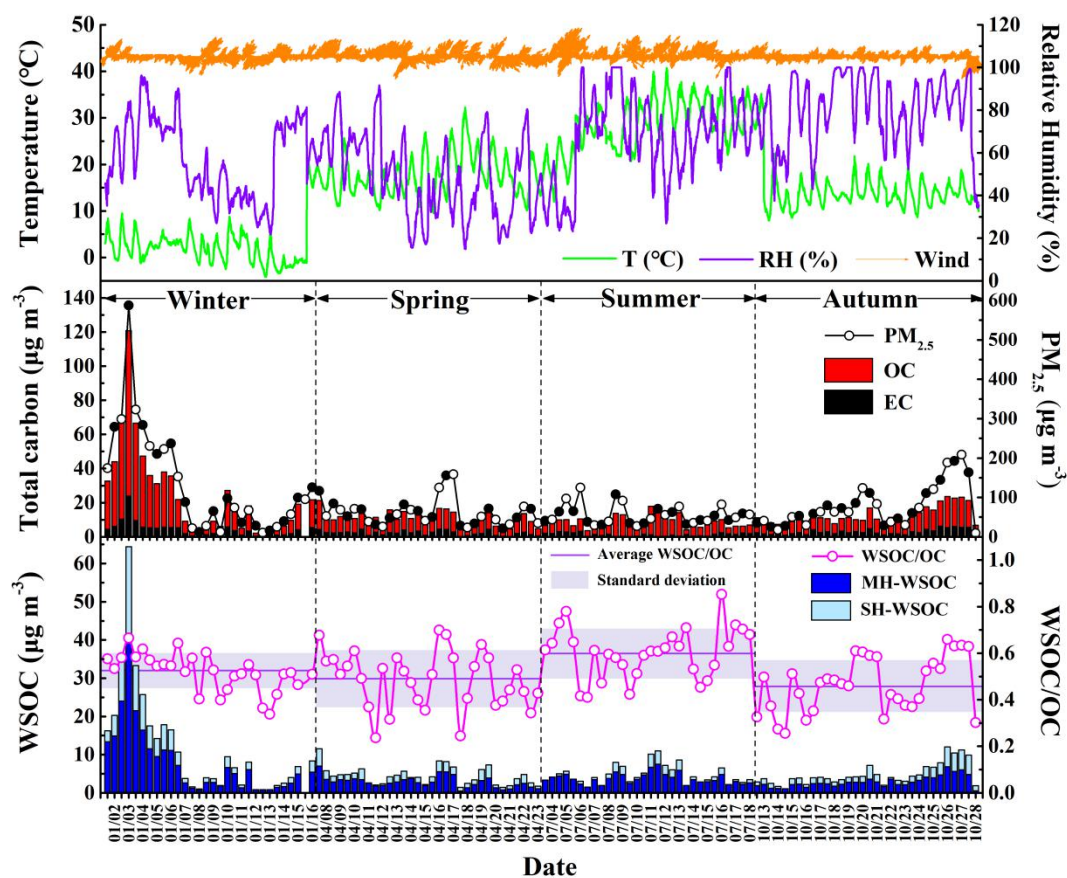


Figure 1. Temporal variations of meteorological parameters, the mass concentrations of PM_{2.5}, OC, EC, WSOC and WSOC/OC ratio in Beijing during the sampling periods in four seasons of 2017.

16. Minor: Line 30: replace “takes up” with “composes”;
Line 117: replace “entirely dryness” with either "entirely dry" or "entire dryness";
Line 121: replace “T The” with “The”;
Line 248: replace “did not appear any” with “did not have any”;
Line 280: replace “association” with “correlation”;
Line 294: replace “appeared” with “showed”;
Line 340: replace “except” with “beyond”;
Line 377: remove “Nevertheless”;
Line 399: remove “of” after Both;
Line 408: believe “C” is missing after “SO”;
Line 427: replace “was in consistent” with “was consistent”.

Response: We deeply appreciate the reviewer’s careful and detailed comments. We have corrected these errors in the revised manuscript. Thank you very much!