Responses to Reviewer #1

We appreciate the thoughtful comments and suggestions from the reviewers, which greatly improved the quality of our manuscript. The point-by-point responses to these comments are listed below with referee's comments in black and our responses in blue.

Reviewer #1 (Formal Review for Author (shown to author)):

The manuscript by Xu et al. investigated molecular markers of primary biomass burning and biological aerosols during different seasons in Beijing, with focuses on size distribution and seasonal variation. Four sets of ambient aerosol samples were collected for each season using a nine-stage cascade impactor sampler, leading to a total of sixteen sets of samples for the entire measurement period. The samples were analyzed for anhydrosugars, sugar alcohols and sugars. Based on the measurement results, the authors discussed the abundances, seasonal variations and size distributions of these compounds, then estimated the contributions of biomass burning, plant debris and fungal spores to OC. In principle, the topic of this manuscript falls within the scope of Atmospheric Chemistry and Physics. However, I could not support its publication due to the following concerns.

Response:

We appreciated the valuable comments from the reviewer. Organic molecular characterization of urban aerosols has been conducted comprehensively during the past decade. However, very limited studies have been conducted for size-segregated aerosol samples. Thus, we believe that our detailed characterization of size distributions of saccharides in urban Beijing provides useful information on the biomass burning and fungal spore tracers and their patterns of size distributions and GMDs for the first time, which is worth publication in ACP.

1、There have been many previous studies investigating the concentrations of saccharides in Beijing aerosol (e.g., Liang et al., Chemosphere, 2016, 150, 365-377). Although these studies typically relied on $PM_{2.5}$ and/or PM_{10} rather than size resolved samples, they generally covered much more sampling days than the present study, and therefore much more representative when discussing the abundances and seasonal variations of saccharides as well as when estimating the contributions of biomass burning, plant debris and fungal spores to OC.

Response:

We appreciated the valuable comments from the reviewer. We know that there are excellent studies focusing on the concentrations of saccharides in Beijing aerosol, while there is still a

lack of knowledge on the size distribution of these organic species. In fact, most of the previous studies use high performance anion-exchange chromatography (HPAEC) (e.g. Liang et al., 2016). Here, we measured anhydrosugars, primary saccharides and sugar alcohols using GC/MS in this study. Generally, many studies were carried out based on dozens of samples by GC/MS (Fu et al., 2008; Li et al., 2018; Wan et al., 2019; Wang et al., 2006). Though we could not take into full consideration the sampling days, each set of our samples analyzed in our study are representative and adequate when discussing the abundances and seasonal variations of saccharides as well as when estimating the contributions of biomass burning, plant debris and fungal spores to OC. Each sample set corresponded to specific meteorological condition, which are listed in Table S1.

Compared to the results from previous studies, are there any new findings in Sections 3.1, 3.2 and 3.4? Maybe the authors should focus on the size distributions of saccharides, which may be able to differ the present study from previous ones.

Response:

In Section 3.1, we give a detailed description about the L/M, M/G, L/OC, and L/EC ratios according to particle size. New findings were as followed:

(1) Higher L/M and M/G values were observed in the fine mode, which ascribed to the difference of burning substrates. Hardwood was potentially the burning substrates in the fine mode while mixture impact of hardwood and softwood burning accounted for the relatively lower L/M ratios in the coarse mode. Higher M/G ratios in the fine mode implied the increasing contributions from crop straws burning, especially in haze days.

(2) Dust storms could induce a high L /EC ratio in the coarse mode because of coarse particles brought by dust storms and/or road dust resuspension. While decreased concentrations of EC in the coarse mode in dust storms implying that EC may derived of local emissions rather than long distance transportation of dust.

In Section 3.4, we calculated the contributions of OC from BB, plant debris and fungal spores in terms of particle size. The contributions of BB-OC were different in the fine and coarse mode. BB-OC dominated in the fine mode (>90%) and the contribution of BB-OC in the size range of 2.1–9.0 μm were with a proportion over 60%. While in a larger size range, the relative contribution were below 60% in most periods. Plant debris-derived OC and fungal spores-derived OC mostly existed in the coarse mode. However, both of them were present in the fine mode, especially in spring and summer. Though the contribution of plant debris and

fungal spores in the whole range were insignificant in cold seasons, their relative contribution in the coarse mode were comparatively high.

2、It is completely unclear how the haze, non-haze and dust-storm periods (which were frequently mentioned throughout the manuscript) were identified.

Response:

Thanks for the reviewer for pointing out the missing data. Such information on the meteorological parameters during each sampling period is added in Table S1. The two sets (11–19 April and 4–5 May) were collected in dust storm days, the following sets (30 June–2 July, 12–14 July, 25–27 October, 6–8 November, 27–29 December and 25–27 January) were affected by haze. And the rest sets were for non-haze days.

3、Considering the formation and evolution processes of haze events in Beijing (which could be fast; e.g., Sun et al., Sci. Rep., 2016, 6, 27151), it is questionable whether the so-called haze samples were representative (recalling that only four sets of samples were collected for each season).

Response:

In many previous studies, total suspended particles (TSP) samples were collected by a high-volume sampler, with an operating flow rate of 1.00 m^3/min, approximately (Chen et al., 2013; Li et al., 2018; Wan et al., 2019). While in this study, all samples were collected using a nine-stage cascade impactor sampler (Andersen, U.S.A.) at a flow rate of 25.8 L/min from April 2017 to January 2018. Compared to the high-volume sampler, the flow rate of nine-stage cascade impactor sampler is much lower. If the sampling durations were too short, the circumstance of the concentrations of size-resolved samples below the detection line will occur. As a result, we had to prolong the sampling time to guarantee the validity of samples, especially for the non-haze days. Former studies found that the formation and evolution processes of haze events could be fast, sometimes happened even less in one day (Sun et al., 2014; Yang et al., 2015). To completely encompass the durations of the rapid formation of haze events and the evolution of secondary organic aerosols (which not discussed in this manuscript), we considered 2 to 3 days as a reasonable sampling period. In addition, such sampling period is necessary to collect enough particles for organic analysis.

4、Please clarify why dust storm is a major source of OC in coarse particles. This point was presented as a conclusion but was not clearly explained in the manuscript.

Response:

Thanks for the suggestion. In this manuscript, we presented "Dust storm is a major source of organic compounds in the coarse particles, which induce a remarkable size shift to the coarse mode". This description may be a little inappropriate. We corrected the conclusion in the revised manuscript as "Dust storm greatly enhance organic aerosol concentrations and induce a remarkable size shift towards coarse sizes (see Page 15, Lines 9–10). There are several reasons for this conclusion. Firstly, in general, the concentrations of most primary saccharides sugar alcohols in each impactor stage during dust storms (17–19 April and 4–5 May) were higher than those of non-haze days (19–21 April and 5–6 May), especially for the coarse particle fraction (Figure 6–7). As for anhydrosugars and related sugars, their concentrations in the coarse mode in dust storm were higher, too. Such phenomenon could be probably attributed to strongly windblown mass coarse dust derived from large scale resuspension of dust from crustal, soil, roads or other unpaved areas, along with long-range transport of particles from north and northwest desert regions. Previous studies found that elevated concentrations of trehalose, mannitol and arabitol are generally related to resuspended soil and the outflow of dust storms (Liang et al., 2013; Rogge et al., 2007). Secondly, the GMDs of the total size range and the coarse mode particles in dust storm were larger than non-haze and haze days (Table 4–S5). Some species, such as arabitol, mannitol and inositol, their GMDs associated with coarse particles in dust storm presented a significantly increase, again suggesting the effect of dust storms on the aerosol particle size. Wang et al. (2013) also found that dust storms could act as a major source of coarse particulate matter.

5、A minor point. Page 6, Line 28. Please check the two ratios cited here.

Response:

Thanks. We have corrected the mistake in the revised manuscript (see Page 6, Line 25–26). The revised content is as followed:

"The M/G ratios during all the periods were in a range of 1.35–2.08 with an average 1.70 (Fig. 2b). The M/G ratios maximized in autumn (1.68–1.97, 1.88) and minimized in summer (1.35–1.82, 1.59)."

Table S1. Information on the weather conditions during each of the sampling periods from April 2017 to January 2018.

Year	Season	Sampling period	Duration (min)	T (°C)[a]	RH (%)[b]	WS[c]	$PM_{2.5}$	PM_{10}	Weather conditions
	2017	spring	17–19 Apr.	2880	18.2	32.3	5	174	124	dust storm
			19–21 Apr.	2887	14.7	46.2	3	78.3	84.6	non-haze
			4–5 May	1364	20.1	33.4	6	501	656	dust storm
			5–6 May	1954	17.6	25.2	5	131	125	non-haze
		summer	30 Jun.–2 Jul.	2862	29.3	70.5	2	143	103	haze
			12–14 Jul.	2854	31.5	76.7	1	89.2	75.9	haze
			14–16 Jul.	2900	29.0	58.3	2	65.5	62.1	non-haze
			21–23 Jul.	2843	25.2	64.2	3	42.7	36.3	non-haze
		Autumn	11–13 Oct.	2877	12.3	64.2	2	38.1	32.4	non-haze
			16–18 Oct.	2900	13.1	69.1	2	78.0	68.7	non-haze
			25–27 Oct.	2865	11.8	81.3	1	183	120	haze
			6–8 Nov.	2887	9.36	72.4	2	146	91.7	haze
	2017-2018	Winter	27–29 Dec.	2781	-2.34	75.2	1	137	140	haze
			2–4 Jan.	2757	-2.68	32.4	3	33.7	32.1	non-haze
			25–27 Jan.	2858	-8.13	41.6	1	82.2	59.6	haze
			29–31 Jan.	2835	-1.95	22.4	2	47.5	49.5	non-haze

[a]temperature (T); [b]relative humidity (RH); [c]wind scale (WS).

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