Anonymous Referee #1

The authors have made significant additions to the manuscript in the areas recommended, and I think it is now publishable. I think though that some of the new text should get a second look for clarity/grammar before finalizing. I highlight a few sentences here that could be improved:

Response: Authors are thankful to the reviewers for their valuable comments and suggestions.

Line 60 suggestion: "On a global scale, one-fourth of anthropogenic aerosol (mass?) is contributed by China, approximately 70% of which was emitted from coal burning"

Response: Modified. Please see lines 60-61 in the revised MS.

"Globally, significant anthropogenic and carbonaceous aerosols are contributed by China (Cooke et al., 1999, Wang et al., 2007)."

Cooke, W.F., Liousse, C., Cachier, H., and Feichter, J.: Construction of a 1° X 1° fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. J. Geophys. Res. Atmos., 104, 22137–22162, https://doi.org/10.1029/1999JD900187, 1999.

Wang, G., Kawamura, K., Zhao, X., Li, Q., Dai, Z., and Niu, H.: Identification, abundance, and seasonal variation of anthropogenic organic aerosols from a mega-city in China. Atmos. Environ., 41, 407–416, https://doi.org/10.1016/j.atmosenv.2006.07.033, 2007.

Line 65 suggestion: "OAs are composed of a complex…"

Response: Modified. Please see lines 65-66 in the revised manuscript.

"Organic aerosols (OAs) are composed of a complex mixture of diverse molecules (Xu et al., 2011)."

Line 363: suggestion "suggested it had sources in addition to fungal spores…"

Response: Modified. Please see lines 391-392 in revised manuscript.

"In contrast, the higher concentration of mannitol than arabitol suggested it had sources in addition to fungal spores in the Mangshan forest site."

Line 585: "contributions of 36%" specify contributions of SCs if that's meant

Response: Corrected. Please see line 619-621 in the revised MS.

"PMF results concluded the contributions of 36% from vegetation (21% vegetation factor and 15% pollen factor) and 37% from microbial and fungal species (21% microbial soil dust and 16% fungal factor) of total measured SCs."

Anonymous Referee #2

Verma et al. reports measurements of sugars collected on filters for about 3 months to determine the potential sources of the sugars. Though the authors addressed many of the comments from both reviewers, there are still concerns in regards to the methods and the interpretation of the data. Some of these concerns were brought up in the first set of reviews but were not addressed at all.

Response: Authors are thankful to the reviewer's valuable comments and suggestions, which help to upgrade the quality of the manuscript. We made significant changes in the manuscript acording to the reviewer's suggestions.

Major

(1) It is not clear if they sampled any aerosol size < 100 um vs having a cut-off.

Response: In this sampling, we used a high-volume air sampler (Kimoto-AS810A) to collect total suspended particles (TSP) without cut-off device. We added information in the revised manuscript. Please see lines 146-147 in revised MS.

(2) Since there is no denuder, what is the impact of condensation of semi-volatile gases onto the filters in the analysis? At least levoglucosan is known to partition between the gas and particle phase. This should be addressed as either a potential uncertainty/limitation if unknown or at least discussed if it is known the potential interference/impact it may have on the reported mass.

Response: In this work, no denuder was used to remove semi-volatile species. The removal of the gaseous species in the denuder distorts the gas-particle equilibrium and leads to the dissociation of the particulate phase during the sampling. This phenomenon is particularly true for volatile organic species. It leads to significant errors in determining gas-to-particle partitioning of organic aerosols due to underestimating the particle phase (Dhawan and Biswas et al., 2019). In this study, we reported nonvolatile sugar compounds. However, the levoglucosan partition between the gas and particle phases, but their concentration was low. The sampling time was rather short due to the day and night sampling. Therefore, we believe that the uncertainty due to the gas phases in the particulate species concentration might be insignificant. Sentences are added, please see lines 147-152 in the revised MS.

"In the sampling, no denuder was applied to remove semi-volatile gases because the filter samples were used to analyze nonvolatile sugar compounds. However, the levoglucosan partition between the gas and particle phases, but their concentration was low. The sampling time was rather short due to the day and night sampling.

Therefore, the uncertainty due to the gas phases in the particulate species concentration might be insignificant."

(3) Though the author cite a reference on how OC, WSOC, and Ca^{2+} was measured/determined, the authors should still add a brief description so that readers do not have to dig through other papers. This can either be added to the main paper or SI, but a brief description in how these were measured would be beneficial for the overall paper.

Response: According to the reviewer's suggestions, we added a brief description of methods for the determination of OC, WSOC, and inorganic ions as new section 2.4 in the materials and methods. Please see lines 196-208 in the revised MS.

2.4. Chemical analyses of organic carbon, water-soluble organic carbon and inorganic ions

The data set and methods for the determination of organic carbon (OC), water-soluble organic carbon (WSOC) and inorganic ion (Ca^{2+}) were reported in He et al. (2015). Briefly, the concentrations of OC were measured using a semi-continuous OC/EC analyzer (Sunset Laboratory Inc., Portland, OR, USA). A punch of the filter (Φ14 mm) was placed in a quartz boat inside the thermal desorption chamber of the analyzer, and then stepwise heating (IMPROVE) was applied. The oven temperature was programmed as follows: under He, every 2 minutes, the oven temperature was increased starting from 250°C for 2 min, at 450°C for 2 min, and at 550°C for 2 min. After that, 550°C was maintained for two minutes under He mixed with 10% O_2, then at 700°C for 2 min and at 870°C for 3.5 min. NDIR detector was used to determine CO_2 generated in the above process (Wang et al., 2005). The carbon content of the sample that evolves to CO_2 between 250 and 700°C was defined as OC.

Aliquots of the filter samples (3.14 cm^2) were extracted with Milli Q water for the water-soluble inorganic ion and WSOC measurements. After extraction, one part was used for the analyses of inorganic ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, Ca^{2+}, K^+ and Mg^{2+}) using an ion chromatography (IC) system (761 Compact IC, Metrohm, Switzerland). Cations on a Shodex YK-421 column with 4mM H_3PO_4 as eluent and anions were separated on a Shodex SI-90 4E column with 1.8mM Na_2CO_3 and 1.7mM $NaHCO_3$ as eluent. The injection loop volume was 200 µl. Both cations and anions were quantified against a standard calibration curve. Another part of the filtered water extract was acidified with 1.2 M HCl and purged with pure air to remove dissolved inorganic carbon and volatile organics. Then WSOC was measured with a carbon analyzer (Shimadzu, TOC-5000). Procedural blanks were carried out in parallel with real samples to account for any contamination (He et al., 2015).

(4) It is still not clear how PMF was determined. What software was used? Were the solutions constrained? How were the solutions selected? At minimum, a time series of the solutions should be added.

Response: For the analysis of source apportionment, Positive matrix factorization (PMF) software version 5.0 (Environmental Protection Agency, USA) was used. The information is added in the revised MS, please see lines 424-425.

The additional information about the PMF are added as text in the revised manuscript and the time series plots of the solutions also added as Figure S-3 in suplementory. Please see lines 445 – 450 in the revised MS.

“The time series plots of the factors solutions determined by PMF were similar to the temporal plots of the concentration of sugar species of the factor composition (Figure S-3). The numbers of factors were reduced if the pair of factors was strongly correlated. The composition of each factor was also checked; none of the pair of factors were found with similar composition. We also investigated the change in factor profile with positive and negative values of f_{peak} for the chosen solution in the PMF analysis.”

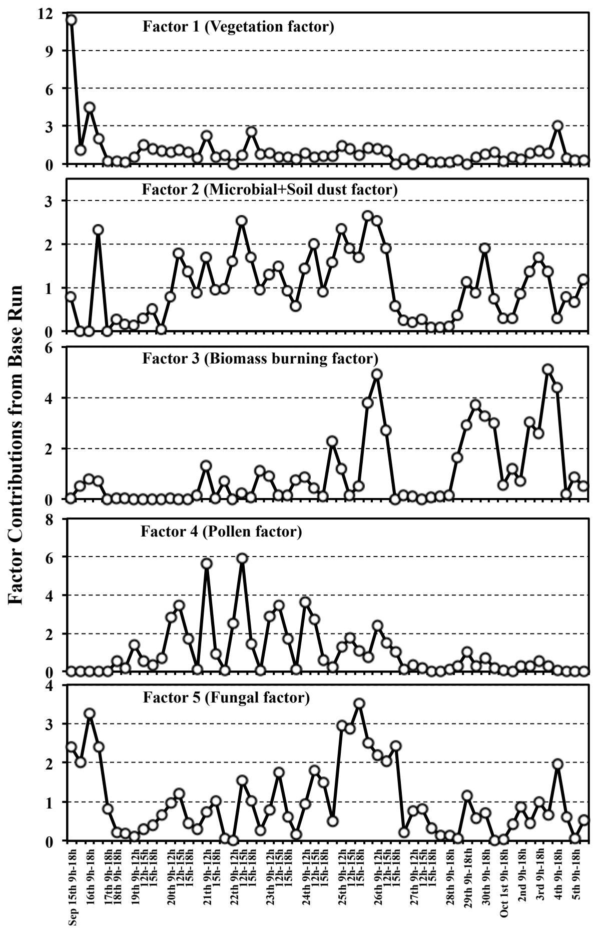

Figure S-3. The time series plots of the factors solutions determined by PMF

(5) As previously mentioned, looking at the average values, the standard deviation, and the number of measurements, many of the "day" "night" differences are not significantly different (student t-test, 95% confidence interval). This impacts the narrative throughout Sect. 3.1.

Response: We discussed the diurnal variations on the basis of difference in the concentrations of sugar species during day and night time, however, we did not observe statistically significant differences (student t-test, 95% confidence interval, $p > 0.05$) in their atmospheric abundances. Therefore, we added few lines in the revied MS. Please see lines 239 and 242.

“The overall concentrations of SCs varied from 30.8–875 ng m^{-3} (avg. 325 ng m^{-3}), which was higher in the daytime (315 ng m^{-3}) and lower at nighttime (276 ng m^{-3}), however, we did not observe statistically significant differences (student t-test, 95% confidence interval, $p > 0.05$) in their atmospheric abundances.”

(6) It is still unclear how biomass burning/levoglucosan is "higher" during night vs day (though as mentioned above, there is no statistical difference using a student t-

test) as the wind is coming from the "forested" site instead of Beijing. Where is this source of biomass burning coming from?

Response: We have mentioned in the text that the local biomass burning are prime sources for nighttime levoglucosan contribution in Mangshan aerosol. The nighttime samples were collected from 18:00h to 09:00h, including peak hours of BB for domestic purposes. Therefore, it is reasonable to detect higher abundances of BB tracers in the nighttime than daytime. However, northeast wind (99.5%) was dominated at night, coming from the forested site, but they carry relatively clean air masses; therefore, it has no significant levoglucosan contribution at nighttime samples from the forest site. In contrast, the daytime wind carries levoglucosan from Beijing City to the sampling site, which contribut levoglucosan during daytime aerosol samples. Please see sub-section 3.1.1 in the revised MS.

We agreed with the reviewers that day/night differences in the atmospheric abundance of some sugar species are not statistically significant. Therefore we added a line in section 3.1. Please see lines 241-242 in the revised MS.

"However, we did not observe statistically significant differences (student t-test, 95% confidence interval, $p > 0.05$) in their atmospheric abundances."

Minor

(1) It is beneficial to type exactly what you have changed in the responses to reviewers instead of asking them to dig through the document to find the changes. Also, it is unclear what, if any, sentences/sections were removed as only additions are highlighted.

Response: We apologize if reviewers feel some difficulties to read response letter. According to the reviewer's comment, we have included all the changes made in the revised manuscript into the response letter.

(2) Line 62 - 66: Sources of OA are misrepresented here, as majority of the OA originate from the photoxidation of gases into SOA (e.g., Jimenez et al, Science, 2009).

Response: Modified. Please see lines 61-65 in the revised manuscript.

"Beijing is one of the largest polluted cities in East Asia; its air quality deteriorates seriously due to massive emissions of anthropogenic aerosols from vehicles and industries (Cao et al., 2014; Qiao et al., 2018; Tao et al., 2017; Wei et al., 2018; Yu et al., 2013)."

(3) Line 170: Was the mass spec a quad or TOF? What was the resolution?
Response: The quadrupole mass spectrometer with resolution of 1000 was used in this study.

(4) Fig. 5: I would recommend making the y-axis label that corresponds to Ca^{2+} the same color as it can be hard to interpret which axis corresponds to which data. Also, Ca^{2+} is not included in the caption.
Response: We change the color of y-axis, and information added in the figure caption.

"Y-axis shows temporal variations in the concentrations (μg m^{-3}) of Ca^{2+}."