

***Interactive comment on* “Quantification of solid fuel combustion and aqueous chemistry contributions to secondary organic aerosol during wintertime haze events in Beijing” by Yandong Tong et al.**

Anonymous Referee #2

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This manuscript describes aerosol chemistry measurements that were acquired during a two and a half month period in Beijing in 2017. Aerosol chemistry was obtained using a PM₂₅ inlet Long Time of Flight Mass Spectrometer (LTOFAMS) and an Extractive Electrospray Ionization-Time of Flight Mass Spectrometer (EESI-ToF-MS). Both of these instruments provide high mass resolution measurements of aerosol chemistry and are complementary in that the EESI-TOF results in low fragmentation, allowing the detection of large molecules making it easier to determine the source and identity of different organic species.

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The study presents a combined positive matrix approach that is used to optimize the factor solutions obtained from the combined instrumentation. It equally introduces the use of z-score analysis on the mass spectral profiles of the different PMF factor solutions to determine which m/z contributed to the identification of each factor. Using this combined analysis the authors present 4 different types of primary organic aerosol and 4 types of secondary organic aerosol. The paper is relatively well written and organized. Figures are well labelled and clear, but in some cases need better referencing in the text. I recommend this manuscript for publication after some minor changes. My comments and suggestions are below.

Sample set up:

Page 4, Line 14: Were the aerosols dried prior to sampling? What was the average sample aerosol RH throughout the study? Were particle losses calculated for the PM1 to 2.5 range?

Can the authors add the size range typically measured by the EESITOF to the discussion of particle size sampling efficiency on page 5/6.

How often was the denuder regenerated during the sampling campaign?

What is the make and model of the SMPS used in this study? What is the size range measured by the SMPS. Can the authors show the aerosol size distribution measured by the SMPS and compare with that of the L-TOF (shown in Fig S2).

How did this size range change during the different sampling events “haze events” and the “clean periods” (Heating non-heating)?

What is the difference in mass between the PM25 inlet and the SMPS. How representative of the total PM2.5 mass is the PM25 LTOFAMS measurements? Are auxiliary measurements of total PM available for comparison?

Given that the smaller, locally formed aerosol particles are not efficiently sampled by the PM25 inlet, what impact does this have on the interpretation of the measurements?

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Is a standard or capture vaporizer used in conjunction with the PM25 inlet? If a standard vaporizer is used, what is the particle collection efficiency estimated to be, and is the calculated CE dependent on particle diameters (to account for the enhanced effects of particle bounce for larger particles diameters)?

High concentrations were measured during this field campaign, often resulting in the clogging of the EESI-TOF instrument. Was there any evidence to suggest that there was overloading of the LTOFAMS instrument?

The authors show several PMF solution for the LTOFAMS analysis but only show the final solution for the EESI-TOF. Can the authors state if the PMF analysis on the "unconstrained" EESI TOF was performed?, and if so which factors dominated the unconstrained PMF solution?

Aqueous phase SOA: The PMF analysis allowed the extraction of a more oxidized aqueous phase aerosol. In Fig. 3a the MOOA_AQ is shown to have highest concentrations (reaching 20 micrograms) during the haze event on the 4th and 7th, during high NO₃ contributions (and high RH and low wind seed). The characteristic enhancement of the CO₂ is illustrated in Fig. 7a during this period. However, there are several other periods during the field campaign when this MO-OOAaq species is identified (at concentrations near to 5 micrograms) (under the same conditions of RH and high NO₃ Fraction) but the enhancement of the CO₂+ signal was not observed.

Is this CO₂+ enhancement really associated with these species or is it somehow an artefact during high NO₃ concentrations?.

How did the factor mass spectral profile compare with reference mass spectra (oxalic acid, malonic acid and succinic acid (Canagaratna et al., 2015))?

The authors state that the m/z 44 artefact is very low in this instrument (4%), however, could this CO₂+ enhancement be somehow related to artefacts linked to mixtures of inorganic and organic species?

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Previous studies have shown how aerosol liquid water can promote the formation of water-soluble organic nitrogen (Yu Xu et al., 2020 Environ. Sci. Tech. <https://pubs.acs.org/doi/pdf/10.1021/acs.est.9b05849>).

What is the role of nitrogen in the formation of these aqueous organic species? Is there evidence of organonitrate species? Has aerosol acidity been evaluated during these measurements? During the intense haze episode, this MOOA species was measured continuously over a period of 3 days. In one of the cited articles (Kuang et al., 2020) it is mentioned that most of the aqSOA was formed during daytime periods with high photochemical activity and that dark aqSOA only contributed negligibly to the total OOA concentrations.

In this work, the increase in aqSOA remains constant over three days with little diurnal variation. Do you consider that this aqSOA is locally formed or influenced by regional processes? Does the aerosol size distribution provide information to determine this?

A non source-specific factor LO-OOA ns ?

Recently it has been shown that the PM₂₅ inlet AMS systems may be capable of measuring airborne bacteria (Wolf et al., 2017 Atmospheric environment, (<https://doi.org/10.1016/j.atmosenv.2017.04.001>)). In this paper, there are some characteristics of the LO-OOAs species (O:C, diurnal pattern, higher concentrations during warm period than colder periods) as the resolved bacteria-like factor in Wolf et al., 2017.

Is it possible to provide more information on the average diameter as a function of time for each of the resolved factors to help provide more information on their source and atmospheric processing prior to being sampled? At least provide the SMPS size distributions which would help illustrate regionally influenced factors and those from local processes.

Were BC measurements available for correlation with the HOA and BBOA.

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Minor remarks:

When discussing the diurnal variations better referencing to the figure is necessary.

Although the information of O/C, H/C are included in and Fig. 5 and 6, it would be useful for comparison to other studies, to have these average values as well as the N/C ratios illustrated on the factor profiles in Fig.3 and 4.

Page 13, Line 11: Were external time series available for comparison, other than CO? Can you provide the value for the "good" correlation.

Page 22, Line 44 (please include correct section no.)

Page 23, Line 7 $r^2 = 0.93$, remove = or of

Page 21, Line 25: Bertrand et al.. please include the full reference.

Page 5, Line 10 What is this diagnostic species?

Is the custom peak fitting algorithm something that could be applied to lower resolution instruments in the future?

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