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Atmospheric Measurement Techniques Discussions

Interactive comment on "Improved source apportionment of organic aerosols in complex urban air pollution using the multilinear engine (ME-2)" *by* Qiao Zhu et al.

Qiao Zhu et al.

huangxf@pku.edu.cn

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1. My general concern about the PMF technique on AMS OA spectra is that, is it reasonable to assume the number of factors and their profiles are always the same in different locations? Is it mathematically true that the more factors you allow PMF to resolve, the better the overall results would be? For example, in Line 133-135, the authors stated that BBOA and CCOA factors could be properly resolved by traditional PMF when the total number of factors was flexed up to 10. Also, the BBOA in Dongguan seems to be much more oxygenated than that in Qingdao, suggested by the much higher O/C ratio and OM/OC ratios. Could that be due to some factor mixing (i.e. BBOA

mixed with OOA), or is it more reasonable to not assume they are both BBOA? E.g., maybe the Dongguan-BBOA should be characterized as aged BBOA or something similar? What are the reported ranges of O/C and OM/OC ratios for BBOA in literature?

REPLY: The numbers and types of factors were determined according to the unconstrained PMF results for each case, and could vary for different cases. For both the two sites in this paper, a 6-factor solution is chosen as the final result following the procedures detailed in Zhang et al. (2007). Crippa et al. (2014) also provides some guidelines to identify HOA, COA (check f55/f57) and BBOA (f60) for ME-2, and the detailed information to identify the existence of CCOA is presented in our paper. Therefore, we have enough evidence to expect four POA factors and two OOA factors for both sites in this study, and finally we got satisfactory running results. More OA factors output by ME-2 would not produce more significant factors. On the other hand, the purpose that we allow PMF to resolve more numbers is to find "purer" or more reasonable MS profiles (e.g., with a reasonable O/C ratio) for certain factors that were not well identified previously, not to look for a better overall result of PMF. The range of O/C ratio and OM/OC ratio of BBOA reported in Canagaratna et al. (2015) is from 0.25 to 0.55, and from 1.50 to 1.88, respectively. But the fresh BBOA can be rapidly converted to OOA in less than 1 day (Bougiatioti et al., 2014), where the O/C ratio for aged BBOA could be up to 0.85 (Zheng et al., 2017). BBOA in Dongguan was apparently not fresh, considering it is an urban site and Dongguan has a warmer ambient air even in winter (17 °C in Dongguan; 9 °C in Qingdao), therefore the BBOA factor identified in Dongguan, with a strong contribution of m/z 60, has a higher O/C, indicating it is an aged and oxygenated BBOA. Following the suggestion of this reviewer, we will name this factor as Aged-BBOA in the revised paper.

2. Are there any industrial sources near the sampling sites at both cities? If so, please specify.

REPLY: There is no industrial sources around the sampling sites, which has been clarified in the text.

3. Figure 3: I highly recommend the authors reorder the source profiles and time series plots in (a) and (b) so that they follow the same order for both sites for easier cross comparison.

REPLY: We have corrected it.

4. Line 259: Please add some more details on how PAHs are derived from the OA spectra. You can also consider moving the details to Section 2.

REPLY: We have added the details about the process of PAH quantification in Section 2.4.

5. Line 269: Are the ratios of PAHs to COA at the two sites in this study comparable to each other and other cities in the literature? Is there any compositional difference in the coals used for heating (and/or cooking) in the e.g. Northern and Southern China?

REPLY: We didn't mention the ratios of PAHs to COA but the ratios of PAHs to OAs, and the ratio of PAHs to OAs (1.8%) in Qingdao was similar to that in the northern Chinese urban site of Xi'an (1.9%) (Elser et al., 2016) but was higher than that in Dongguan (0.9%) in Southern China. According to the spatial variation of heavy metal elements from coal (Tian et al., 2012), we can find that the emission compositions of coal combustion in different regions in China are quite similar. This information has been added into the revised text.

6. Line 275: correspond with -> are consistent with

REPLY: We have corrected it.

7. Line 282: pollutants -> organic aerosol pollutants

REPLY: We have corrected it.

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