

## General comments

This manuscript reports a compositional analysis of PM<sub>2.5</sub> by offline filter measurement on a day-night basis and NR-PM<sub>1</sub> by an in-situ HR-TOF-AMS near the HSC in May 2015. Integrated source apportionment analysis for OC was conducted incorporating AMS-PMF, MM-PMF and CMB methods. The three source apportionment models are in agreement that ~50% of OC was formed from primary fossil fuel combustion. SOA was further apportioned to anthropogenic and biogenic sources by combing results from MM-PMF and CMB. The comprehensive nature of this work in data and source apportionment analysis bring to the literature a valuable case study report. This work nicely contributes to our improved understanding of SOA sources. I have the following specific points that need clarification/more discussion in their next revision.

### Major comments:

1. The offline and online measurements overlapped between 13-27 May. Some comparison of the two measurements would be useful as data cross checking and verification. Such comparisons are currently lacking. For example, what's the percentage of PM<sub>1</sub> contributed to total PM<sub>2.5</sub> and how the two mass concentrations correlated with each other? This is important not only for data validation, but also for the latter comparison between AMS-PMF and MM-PMF and CMB.

2. There are inconsistencies among the source apportionment results by the different approaches. AMS-PMF resolved a cooking influenced factor but no BB related OA, and the authors proposed the loss of the m/z 60 signatures during transport as possible reason. This reason is not supported by their data, as MM-PMF resolved both BB and cooking sources and BB even contributed more than cooking emission (11% vs 1%). It is odd that the authors didn't include cooking emission profiles in CMB. Previous studies have successfully apportioned PM<sub>2.5</sub> to cooking emission (6%) in *Fraser et al.* [2003], as mentioned in the introduction part.

3. The AMS-PMF resolved a CI-SV-OOA factor that contributed to 31% of ambient OC (Table 5), but such a considerable contribution seems inconsistent with the minor cooking contribution estimated from MM-PMF (1%). This inconsistency calls into question whether naming the SV-OOA factor as "cooking-influenced" is appropriate, as it may imply this factor is largely influenced by cooking emissions.

4. The identification of different vehicle emission factors, i.e. diesel engines, gasoline engines and non-tailpipe vehicle emissions, is not very convincing, considering alkanes, PAHs are not unique source tracers to vehicle emissions. It is unusual that norhopane and hopane are separated into two different factors. The two species are usually highly correlated with each other. What's the correlation between the two species in this study? Also, a high amount of EC was present in low-NOx anthropogenic SOA factor. Does this indicate the mixing of primary sources in this factor?

5. In the CMB model (section 2.5), were EC and levoglucosan included in the calculation? If not, which species was/were mainly responsible for determining the contributions from diesel engines and biomass burning? Also, it would be better if the statistical performance of the CMB results can be reported (e.g.,  $\chi^2$ , calculated vs. modelled species concentrations, species source contributions).

6. The sample size of MM-PMF for PM<sub>2.5</sub> OC is 46 in the study, but in the introduction section it is suggested the sample size should be 60–200 (P.2 line 39–40). It seems, because of this reason, the MM-PMF performance is not very robust as shown in Table S2 (some key species like cis-pinonic acid and phthalic acid are not well modelled) and Table S5 (only 54% of bootstrap BB factor was mapped). The uncertain BB factor contribution in MM-PMF would also weaken the reliability of BB SOA estimation in this study. Please comment on how the small sample size affect the MM-PMF results in this work.

7. Fig. 8 shows that the non-tailpipe vehicle emissions (marked largely by 17 $\alpha$ (H),21 $\beta$ (H)-hopane only) made a notable contribution after 20 May (except 10 May daytime), is there any reason for it?

8. Is there any reason to select 17 $\beta$ (H)-21 $\alpha$ (H)-30-norhopane over 17 $\alpha$ (H),21 $\beta$ (H)-30-norhopane as the vehicle emissions tracer in CMB and MM-PMF? The former species should be less abundant in ambient and source PM, and therefore is less commonly used in receptor models (e.g., Yu et al., 2011, *Analytical and Bioanalytical Chemistry*, 401, 3125-3139).

9. When the source apportionment results from AMS-PMF, MM-PMF and CMB were integrated to obtain insights in SOA contributions (section 3.5), it seems the conclusion that anthropogenic SOA is the dominant contributor is largely drawn from the MM-PMF results, while the CMB and AMS-PMF results do not converge to the same conclusion (e.g., P.14 line 17: CMB is unable to provide a reliable estimation of total anthropogenic SOA, ... ; and line 20–21: AMS-PMF is unable to distinguish between anthropogenic and biogenic origins of SOA, ...). However, the MM-PMF results may have large uncertainties, especially considering the small sample size and poor stability (e.g., only 67% of bootstrap high-NO<sub>x</sub> anthropogenic SOA factor can be mapped as shown in Table S5), making such a conclusion less convincing. Additional supportive evidence/argument for this point is recommended.

#### Minor comments:

1. Line 4-5 in p7: “organic carbon”, “elemental carbon” and “Organic matter” should be removed as the abbreviations have been introduced before. Check throughout the context to avoid redundant words.
2. Include the measured OC concentrations in Figure 8, which can help visualize directly how MM-PMF predicts the measured concentrations.
3. Line 16 in p1: Change “fine” to “ambient”.
4. Line 25 in p1 and line 11 in p2: “VOC” should be “VOCs”.
5. Line 21 in p2: “for” should be “from”.
6. Line 13-16 in p6: the correlations are R or R<sup>2</sup>?
7. Figure S10, correct the typo mistake for the y-axis.
8. P.5 line 36: Bituminous coal source profile was said to be included in CMB, but it is not reported in the result section (3.4.1), please check.
9. P.8 line 25–26: The order of HOA, CI-SV-OOA and LV-OOA should be reversed to be consistent with the statement in the next sentence, as well as the abstract (line 20–22).
10. p.10 line 13: Full stop missing in “... comparing this study to Buzcu et al., (2006).”
11. P.11 line 34–36: Yan et al. (2008) did not report isophthalic acid in aged BB plumes, please consider citing another reference.
12. P.13 line 8 & 9: section 3.5.2 → section 3.4.2
13. P.13 line 33–34 argued that isoprene-derived SOA contribution estimated by MM-PMF is more reliable than that by CMB, is there any further support/reference for this argument? Uncertainties associated with ambient measurements and temporal correlation between species in PMF should not be neglected.
14. Space missing: after Table S2 in P. 10 line 28; between “models reveals” in P.14 line 25; after NR-PM1 in P.33 Table 2 title.
15. P.14 line 32: ..., from BB (5%), ...
16. P.15 line 5: ... to organic aerosol by can be estimated by...
17. Missing close parenthesis for graph (a) and (b) in Fig. S3.
18. Misaligned unit in the y-axis title of Fig. S10.