

***Interactive comment on* “Seasonal analysis of submicron aerosol in Old Delhi using high resolution aerosol mass spectrometry: Chemical characterisation, source apportionment and new marker identification” by James M. Cash et al.**

Anonymous Referee #2

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General Comments:

The manuscript by Cash et al. describes a chemical characterization and source apportionment work using high resolution aerosol mass spectrometry (HR-AMS) data collected in one of the most polluted and populated areas in Old Delhi, India in 2018. It covered 3 time periods: pre-monsoon (around 1 month), monsoon (around 15 days), and post-monsoon (around 6 weeks). The study identified two traffic-related factors (NHOA and HOA), two burning-related factors (SFOA and SVBBOA), a cooking factor (COA), and two SOA factors (LVOOA and SVOOA) using positive matrix factorization

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(PMF). In the end, the Authors suggest developing air quality policies by mitigating emissions from crop residue burning, open waste burning, and traffic activities. Cash et al. were able to reveal the mystery of high chloride concentration using HR-AMS, which has strong links with the burning sources, especially in the post-monsoon period. In addition, with the rather well-resolved PMF solution, it is certainly a valuable reference for AMS users in the future (e.g., the mass spectra of NHOA, SFOA, SVB-BOA). Collecting and analyzing such a dataset from a high temperature, high relative humidity environment remains challenging, but the authors were able to justify their results using both independent and internal tracers. The multilinear regression analysis is a useful tool for this dataset considering there is no reference HR mass spectrum available for a unique environment, like old Delhi. I believe the contribution of this work is significant and meets the scope of the ACP. Overall, the English are perfect, but the whole manuscript is not so well-organized and wordy, which makes it difficult to follow. For this reason, I will suggest accepting this manuscript only after re-organizing of the text and considering some major issues listed below.

Major issues:

Section 3.3. focus too much on the technical details in justifying the PMF factors using some independent or internal tracers, which is important but not the scope of this study. Thus, I will only summarize it within few paragraphs, and move some figures and texts into SI. I suggest focusing more on the discussions of each source you retrieved from PMF. The separation between section 3 and 4 and the long text makes the storyline discrete and difficult to follow. In addition, it also makes figures often not the closest to the texts that explain the figures, which makes it difficult to read. Please condense the captions you have in this manuscript, especially for the figures in the SI.

Technical comment:

Have you tried to run the bootstrap analysis to see if you get a rather robust result? If it is just a single PMF run, I would argue you might suffer from rotational ambiguity. How

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did you cope with that? Have you ever tried to use factor profiles from “7f_ac_S1_C1” as a-prior information to run PMF but with rather loose constraints?

Detailed comments:

1. Line 73: Add (Lalchandani et al., 2021; Tobler et al., 2020) to your citation.
2. Finger 1: Combine Fig1, Fig.2, and Fig 5 to give a better overview of the chemical composition as well as the data availability
3. Line 129-131: Provide RF, RIEs somewhere, also mention how many times you have calibrated the instrument.
4. Line 124: I will try to mention how many data points were eventually considered and used to run PMF.
5. Line 134-136: Not convinced about how you decide the CE=1 for the Diwali period. Why keep the gradient of 0.8 instead of 0.9, which you obtained from pre- and post-monsoon campaigns?
6. Line 263-264: “The pollution rose suggests that most of the organic mass is from the east and southeast with high peaks ($> 140 \mu\text{g m}^{-3}$) originating from the west and north-west.” I will add “(Fig. 4(e))” at the end of the sentence. But I cannot draw the same conclusion as you do, I can only see that these high peaks were not originated from east south, northeast, and southwest, the width of the other direction at >140 looks quite similar to me. I will suggest to re-do the plot with more bins of concentration >140 to see if you can see some patterns. Otherwise, I will not make such a statement.
7. Line 264: “Its polar graph also shows some extreme values existing from the south-east.” Add “(Fig. S8)” at the end of the sentence to direct readers. Please read through your manuscript again to guide the reader properly.
8. Figure 4: Why there are negative concentration in (b), (d), and (e), what happened to these data points? 1. These plots did not seem to have many fractions for the smallest

concentration bin; 2. I am surprised that you still end up some noisy data points in such a polluted area like India, please explain in more details if you do believe these negative points are real.

9. Figure 6: Please explain what the uncalibrated PAH concentration is in the caption.

10. Line 299-300: “The PAH time series in Figure 6 shows similar quantities during the pre-monsoon and monsoon periods and the consistent low levels suggest common sources such as traffic, solid fuel burning or cooking activities.” Similar to what? OrgNO? Is the second part of the sentence not finished yet? Or did you mean because the low level of PAH, suggested the sources of PAH are traffic, solid fuel burning, or cooking activities? If so, please add a citation at the end.

11. Line 300-302 “There is however a large increase in PAHs during the post-monsoon period when the burning of the rice crop residue begins,” how do you know it started in the post-monsoon, please provide a citation here.

12. Figure 8: label the Diwali period.

13. Line 345: “The PMF results from the AMS unit-mass resolution flux measurements (Ben Langford pers. commun.) show two traffic factors which peak at 9 a.m. and 11 a.m., which supports this.” There are some other source apportionment studies in India having HOA factor, I suggest not citing an unpublished manuscript.

14. Figure 9: It is difficult to locate the “Lower panel” when it is a text not a “(b)” in the caption especially when the caption is so long. In addition, for the lower panel graph, I would change the y-axis to mass concentration, and keep the percentage of each factor inside the bar, it makes readers easier to compare the relative concentration of each factor for a different time period. Besides, I think you don’t gain extra information here by adding inorganic into this graph when you already have Figure 2.

15. Line 364: “This may suggest atmospheric processing is occurring, for example oxidation reactions.” Please explain what oxidation reactions occurred and why it caused

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the larger spread of the mass of NHOA in different wind directions. Or did you just simply imply the NHOA was rather fast oxidized that cannot transport in a long-range?

16. Line 437-438: “The polar graph also shows maximum concentrations coincide with a south-easterly direction. The timing of these peaks match with maxima seen in polar graphs for PAH, SFOA, and NHOA, which suggests a common source.” Please mention which figure it is since you have two polar graphs and two pollution rose figures.

17. Line 458: “UnSubPAHs” is not defined in the previous text.

18. Line 836-837: “One study indicates that this behaviour results in larger amounts of municipal waste being burnt in the morning compared to the evening (Nagpure et al., 2015).” One could add is that the temperature in India is not low, thus the evening peak is not so pronounced.

19. Line 925: “PCDBs” and “PCDFs” did not mention previously.

Comments for SI:

1. Line numbers are missing

2. The second paragraph on P3: I understand previous studies used the terminology of “background concentration” to define the intercept of the multilinear regression, but I found it was difficult to read directly from Fig. S2-S5 without any other clarification. It is quite miss leading for me, I thought it was an averaged background level in a given time period of one of the tracers, I wonder why it was different over different PMF runs on the same dataset. I will change this terminology or at least descript it in the caption to make the graph easier to read.

3. Last paragraph on P3: use a table instead to explain the name of each solution, it is very difficult to read and understand in the text. Also, in this way, the captions in Fig. S2-S5 could be shortened.

4. Title on P7: “S2. Method for determining the Inorganic-Organic PMF solution”

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should be S3

5. Figure S8 on P13 and Figure S10 on P15: add the units for conc. and (a), (b), (c), (d), (e), etc. in the graph. Consider using a color code or anything else to differentiate the wind speed but not with closed circles. It is just not easy to read about the overlapped points, which are true for most of your data points.

6. The first paragraph on P12: “Error! Not a valid bookmark self-reference”, please revise.

7. Figure S7 on P19: Could you also label where are the two cremation sites are if you are going to mention them in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1009>, 2020.

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