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Comment on acp-2022-179

Anonymous Referee #1

Referee comment on "Contributions of primary sources to submicron organic aerosols in Delhi, India" by Sahil Bhandari et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-179-RC1, 2022

The study by Bhandari et al. builds on the group's previous and companion work measuring and characterizing temporal patterns and source apportionment of non-refractory fine particulate matter (NR-PM) in Delhi. They have developed a "time of day" PMF method which improves on the traditional PMF algorithm primarily because it does not rely on the assumption of source chemical profiles being static over time. Instead, the authors use their new method to explore the possibility of diurnal and seasonal source profile changes in typical aerosol mass spec PMF factors such as BBOA, COA, and hydrocarbon-like organic aerosol (HOA). Most of the nuts and bolts of this method is covered in their companion paper, almost to the point that the AMT manuscript is an extended "methods" section of this manuscript. Naturally, the discussion in this manuscript is thus quite reliant on the methods presented in the AMT manuscript. Ideally, this manuscript should be considered for publication after the manuscript presenting the underlying methods has been peer-reviewed. As such, I will preface the remnant of my review with the following: this review of ACP manuscript assumes that the underlying methods presented in the companion AMT manuscript do not have any major technical issues.

This manuscript focuses on how the authors interpret their results, their advantages in informing source apportionment, and policy implications. Overall, I think this is a very informative approach developed by the authors. The manuscript is very thoroughly prepared and the results are presented clearly. I have a few comments that I tihnk should improve the readability of the manuscript.

A couple of general comments first:

1. Because several different PMF runs are being compared in this manuscript, I understand why the authors decided to tag each PMF run with the SYYTTTT code. But is the "YY" component really needed? As far as I can tell, all measurements occurred in 2017. I suggest removing the YY component and hyphenating to make the code more readily graspable i.e., S-TT-TT.

2. I think the authors need to be cautious in claiming that all differences between their "time of day" PMF approach and the conventional PMF approach can be attributed SOLELY to potential changes in source chemical signatures. After all, PMF is blind to chemistry and it simply tries to find a local minima in Q/Qexp for whatever dataset is provided to it. Time-varying instrumental uncertainties, relative ionization efficiencies, etc., can also play a role in how this solution is found. In fact, for the same input dataset and same number of factors specified, multiple slightly different PMF solutions can be derived based on how the matrix is rotated ("f_peak"), or how the first numerical step is taken by the algorithm ("seed"). Attributing variations between PMF runs to source chemical signatures without exploring (or at least acknowledging) these other possible sources of variations seems risky.

Specific comments:

L104: the term "NR-PM2.5" is used here for the first time. Please define. Also, if not already specified, please specify that the DAS ACSM measured NR-PM1, not NR-PM2.5.

L118: "… its ubiquity". Please clarify what is being referred to here. Ubiquity of cooking sources? Or ubiquity of detectable COA? Its a hair-split, but good to clarify.

L135: "IGOR PET" – Igor is the software, which isn't an acronym, so shouldn't be capitalized. And PET is a super-acronym (the "P" stands for an acronym itself), which should be defined. Also, since PET is first mentioned here, the Ulbrich 2009 citation should be included here.

L164: at the end of "… course of the day", I suggested adding a clause "especially OOA factors", because the phenomena described later (reaction chemistry, gas-particle partitioning, changing meteorology, etc.) would affect the chemical signature of OOA factors more than primary factors.

L240: If I am understanding Figure 1 correctly, the sentence "clearly, POA concentrations exhibit larger variability than OOA concentrations" should end with the phrase "in winter".

Monsoon POA and OOA both seem to be similarly variable (0 to 50 ug/m3, from the looks of the y-scale).

Figure 1: I suggest a few tweaks to this plot – a) add Spearman R to each panel comparing “time of day” to “seasonal” PMF; b) adding “WINTER” and “MONSOON” to the panel headers, and adding a dividing line between (a,b) and (c,d) will make the seasonal distinction jump out better.

L291 – 304: comparisons are made to other studies that made measurements at 3m height near an arterial road, but it would be help to also include the DAS study parameters here (I know 4th floor of the building, but just to compare numbers, please include sampling inlet height off of the ground, and distance to nearest major roadway).

L334: the nomenclature “winter-to-monsoon” sounds like a ratio of winter to monsoon levels. Instead, I suggest something like “monsoon-adjusted winter BBOA”.

L345: please verify precip data shown in Fig S1. Seems like an abrupt step change at 6 am. If there is an explanation for this, please include in Fig S1 caption.

Figs 2 and 3: I don’t see why these two cannot be combined into one figure, similar to Figure 1. Irrespective of whether authors choose to combine Figs 2 and 3, please consider adding a text label to the figures clearly specifying the season.

L385: The studies cited here for “Asian cooking” references were not conducted on Indian cooking styles. Some of them were on Chinese cuisine emissions (or measurements conducted in China). I would be careful about bucketing Chinese and Indian cooking together as “Asian” here, without at least acknowledging that these two are vastly different and there just isn’t literature on the latter as there is on the former.

L390 and several other places: the m/z 55:57 signal ratio is used often to infer cooking influence, but it is not clear what is a reference value for this ratio to compare against while making this inference. For example, L416: why is the 55:57 value of 1.1 “low”? This is where those reference values from Robinson et al. and Mohr et al. would be helpful. I suggest that when the Robinson 2018 study is cited first here, also include the 55:57 ratio from that study, so as to “set the stage” for upcoming discussion on 55:57 ratios. Also, Claudia Mohr’s 2012 study (https://acp.copernicus.org/articles/12/1649/2012/) was an earlier one that used the 55:57 ratio to identify cooking emissions. It should be cited here along with Robinson, and example values of 55:57 ratios from both should be mentioned for reference.

L653: “with larger ratios of contributions of m/z 55 to m/z 57” please include a number or range here to quantify what “larger” means.