

Atmos. Meas. Tech. Discuss., referee comment RC1
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Comment on amt-2021-174

Anonymous Referee #1

Referee comment on "On the use of reference mass spectra for reducing uncertainty in source apportionment of solid-fuel burning in ambient organic aerosol" by Chunshui Lin et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2021-174-RC1>, 2021

This study presents laboratory studies testing various solid fuels and stoves as well as ambient measurements investigating solid fuel emissions using the limits approach within ME2. The findings in this research will help improving the source apportionment of OA by applying the limits approach within ME2 analysis.

The paper, which fits well within the scope of AMT, is recommended to be published after working on the following main comments.

The introduction is well elaborated with adequate use of references. However, it may be improved by describing more Dublin, previous solid fuel studies and the importance of the present work.

I think This work needs details about the ME2 analysis and the selection of the optimal solution, perhaps explain it in the supplement material. The authors mention ME2 does a better job compared to PMF and also that the limits approach improves the ME-2 source apportionment. However, there are no details on the factor selection. Also, the R2 is not the best statistical parameter to use when comparing mass spectra due to the large contribution of the important m/z for example m/z 55 or m/z 57. A more suitable parameter should be used, for example uncentered R2 when analysing mass spectra.

The authors mention the technical details of the lab experiments are in a previous publication but perhaps a quick overview should be covered here, i.e. the sampling times/conditions, maybe diagrams, photos, etc.

Line 17. Perhaps change ACSM by aerosol mass spectrometers. Mass spectra are used for source apportionment with all the AMS instruments not only ACSM.

Line 19. When the authors mention stoves, I think this refers only to solid fuel burning used for cooking, is that correct? If that is the case how to differentiate between solid fuel and biomass burning? Or solid fuel used for other activities apart from cooking?

Line 23 how can the authors attribute a >100% variation in the m/z? where in the manuscript is that mentioned?

Line 29 What do the authors refer as "despite their small uses"? are you talking about the ME2 approaches or the solid fuels?

Line 44 What is the purpose of the reference "Chen et al., 2020"? I think the references of Canonaco and Paatero you cover the ME2 and PMF nicely.

Line 45 I would not be as strong in the statement to say that "ME2 is a significant source of uncertainty" at least not using references 9 years old or more. You might want either to rephrase the line mentioning something like "ME2 can be a significant ..." or find more recent references.

Line 70 It would be good to see a diagram and/or photos of the stoves.

Line 82 Was the ACSM calibrated? If so, please describe it.

Line 92 It might be a good idea to add a map, perhaps in the supplement, indicating the location of the sampling site and showing the area of Dublin for readers that are not familiar with the location. Perhaps to extend a bit more in the site description.

Line 97 Please add the website link and date of last access.

Line 104 I kind of follow the description of the PMF model equation because I am familiar with the topic. However, for someone else might be challenging to understand it as it is. Please rephrase it, for instance what does "p" represent? Or use the other equation showing the summatory and the use of I, j, p, etc.

Line 112 It seems that there have been a couple of solid fuel studies in Dublin. Those can

be used in the introduction to inform the reader about the current findings and identifying the needs of the current study.

Line 114 I think Paatero 1999 (<https://doi.org/10.1080/10618600.1999.10474853>) or Canonaco et al 2013 would be more suitable references here.

Line 115 Again, I think here the authors make a strong statement that ME2 is better than PMF by involving mass spec from previous studies and it is assuming the PMF solution will be with mixed inaccurate factors. I think the authors should rephrase this paragraph mentioning the caveats of using inaccurate target profiles and also that PMF does a good job on performing source apportionment and is only when the target profiles are mixed when the use of ME2 shows an improved performance if the user does an extensive and objective analysis.

Line 121 This is why the user should do an intensive analysis with ME2 testing different α -values and/or different mass spec target profiles.

Line 125 is this "SoFi" the same SoFi version 6.F1 mentioned in line 101? Or did the authors used different versions?

127 What is a bootstrap-based resampling strategy? Could you explain it in a couple of lines?

134 What I understand here is that the stoves are used for heating and not for cooking. Please, explain this in the introduction so the reader could follow the manuscript nicely.

Line 165 Apart from m/z 60, m/z 73 is also a solid fuel marker. The authors might want to consider include it into the analysis.

Line 166 Are $f_{m/z}$ calculated the same as f_{60} and f_{44} in line 170? If not, maybe use different acronyms.

Line 169 I think Ng et al (2011) ([doi:10.5194/acp-11-6465-2011](https://doi.org/10.5194/acp-11-6465-2011)) would be a more appropriate reference here to avoid confusion.

Figure S1 in supplement. The Relative difference refers to $f_{m/z}$ difference or m/z

difference? The authors might want to explain figure S1 in supplement, for instance who is y and who is x based on equation in line 166.

Line 189 What are these "PAH-related fragments", do you mention them previously?

Line 203 How could the authors state that solid fuel burning emissions are the dominant source due to OA and BC similar diurnal trend? At this stage is why we use PMF/ME2, to identify potential sources and do not to conclude from OA diurnal cycles.

Line 204 SOA can be transported from long distances and/or to increase concentrations due to drop in the boundary layer height and not necessarily to represent the contribution of heating emissions.

Line 195 300 $\mu\text{g m}^{-3}$ are high aerosol concentrations it would be interesting to know the type of monitoring site, the type of city Dublin is, I guess Nov-Jan is considered to be Winter and this makes sense for analysing solid fuel OA concentrations but it would be useful to explain all this maybe in the introduction. Also, it would be interesting to analyse the large peaks and see if they can be explain i.e by looking at the meteorology or special events. Did the authors look at the boundary layer height for this manuscript?

Line 211 why only wood, peat, and smoky coal?

Line 216 Could the authors explicitly mention the number of factors identified? What about cooking OA? How the solution with an additional factor looked like? Maybe add details about how the solution was obtained would be added in the supplement.

Line 218 I think the methods section there should definitely be a more detailed description of the city/sampling site. Is it correct to associate HOA to oil heating? Would not be that it is actually from traffic emissions? Section 3.2.2 is about ambient OA, Is Dublin a remote site where there is no important traffic emissions or cooking activities?

Line 328 If I'm correct, in this study a 5-factor solution is the chosen one, did Lin et al (2018) also selected a 5-factor solution? Are the same 5 factors identified in both studies? I would like to see in the supplement how the 5-factor solution was selected in this study, how the 6-factor solution looks like? Is there another BBOA factor that might hold the OA concentrations that are not attributed to the target profiles used in the constraint?

Line 230 There is no description of bootstrap in Method section.

253 It is not clear to see the OOA spikes during evening and night time in Figure 3c, maybe show a diurnal plot instead. Moreover, The evening OOA spikes would be related also to boundary layer. It is not clear to me how to relate the evening peaks with the results of using the dilutor.

Line 268 What do the authors mean by "their high emission factors"?

Line 266 If there were found high concentrations of 300 $\mu\text{g}\cdot\text{m}^{-3}$ and ~50% is from solid fuels, does this mean ~150 $\mu\text{g}\cdot\text{m}^{-3}$ of solid fuel OA is produced from 10% of the population only?

Line 287 What about m/z 73?