Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2020-447-RC1, 2021

Atmospheric Measurement Techniques Discussions

Interactive comment on "Long-term aerosol mass concentrations in southern Finland: instrument validation, seasonal variation and trends" *by* Helmi-Marja Keskinen et al.

Anonymous Referee #1

Received and published: 12 March 2021

Summary:

The manuscript presents a long ground-based dataset of aerosol mass concentrations at the SMEAR-II site in southern Finland. Mass concentrations are measured by integrating size distributions, by SHARP, and by using a standard gravimetric impactor technique. These measurements agree reasonably well for the 13-year record. Seasonal trends are discussed. While presenting long-term measurements is certainly valuable and worthy of publication, the text lacks second-order analyses that could make the conclusions less speculative and more defendable. Thus, I recommend publication only after major revisions.

Major Critiques

1. Size distribution measurements from DMPS+APS are one of 3 techniques used here to derive mass but these measurements are not explicitly shown in the manuscript. Combining these datasets is not trivial since each technique is fundamentally different and requires a knowledge of density and shape to be properly stitched. I think a necessary addition to this paper would be to show example distributions (at a minimum) or a statistical summary of the seasonal and annual variability in distribution. This is necessary for both method verification but also would add an additional layer of complexity to the paper that might help solidify some of the seasonal explanations presented later.

2. I think there is a lot of information content from the combination of these datasets that would be interesting to probe deeper and would significantly add value to the paper. For example, how does the DMPS+APS vs impactor regression vary seasonally, which may suggest that your constant density assumption breaks down in the presence of different dominant aerosol types? How has just the coarse-mode (i.e., PM10 minus PM1) varied annually and are those concentrations changing over time?

3. The Figure 2 regression analysis is a great first step but I'm not sure that a simple correlation coefficient and slope tell the full story. For example, panel B seems to show a concentration-dependent bias in the instrument responses. I suggest at least supplementing these regression plots by plotting instrument concentration ratios vs concentration. For example, for panel B plotting [SHARP / IMPACTOR] vs IMPACTOR would highlight the relative difference in the techniques for different mass loadings and illustrate the relative accuracy of SHARP at over a range of mass loadings.

4. There is a lot of speculation in the interpretation of trends sections (3.2 and 3.3) and discussion of each technique separately is very confusing. I recommend focusing the discussion on only the standard impactor technique here since the other techniques do not offer any additional information and correlated well. Also, one of the real values in this dataset is assessing the annual trends like in Figure 4. I suggest adding analogous

plots for the other seasons, and potentially for just PM1 (all seasons) so that these long-term trends can be highlighted.

Minor Edits:

Line Number

23 The statement “Temperature had a strong influence on the measured concentrations.” is too speculative for the abstract and should be removed or the tone softened. I’m just not sure you can say this so strongly.

28 Remove “especially in winter”, it is redundant.

46 You might want to include aircraft emissions (anthropogenic) and marine sea-spray aerosol (natural) in your list of aerosol types

134 What had a “$\sim$10 min time resolution over the measured sizes”? Is the APS operated at 1Hz and averaged to 10 minutes, or are you referring to the DMPS?

139 This section was a bit confusing regarding whether you are working in aerodynamic or mobility diameter space. I think the first sentence should be flipped, “To have comparable particle size distributions, we calculated the aerodynamic diameter from the mobility diameter” should really be “To have comparable particle size distributions, we calculated the mobility diameter from aerodynamic diameter for APS measurements”.

167 This 20 ug/m3 filtering seems arbitrary. Unless there is clear evidence that the measurement was contaminated by a local source or by an instrument operational issue, how do you justify simply removing the data? Can you provide statistics on how much data was removed by this filter, and the sensitivity of your results to this filtering approach?

174 For calculating PM10 from “DMPS+APS”, how do you account for the overlap between the instruments in the 0.5-1.0um size range? Is there a stitching routine to produce a continuous size distribution? Are you choosing to use one instrument and

not the other? Is the APS data cut off above 10um so the data are directly comparable with SHARP and impactors?

178 replace “chapter” with “section”

226 The statement “In autumn, the boreal forest starts. . .” should be supported by references.

248 Why are you only showing SPRING median concentrations and fits in Figure 4? I suggest showing data and fits for each season.

252 Are there higher concentrations of SO2 in the spring compared to other seasons?

256 The sentence “After all, it is possible. . ..” Sounds quite speculative. Can you provide references to support decreases in SO2, PM2.5 over this time frame related to EU policy?

259 This is speculative as well. Does your data show that there is a springtime peak in coarse-mode aerosols that could be ascribed to the presence of pollens?

296 “Otherwise, the annual summertime maximum. . .” is entirely speculative and should not be in the conclusion unless the tone is softened.

Table 1: Why not cut the APS distributions off at 10um so all the PM10 data are directly comparable?

Figure 2: Since the impactor measurements are your direct mass measurement that other techniques are being compared to, I would remove the “SHARP vs DMPS+APS” panel.

Figure 2: It looks like the range in SHARP values is very different between panels a-b. panel a varies from roughly 2-12ug/m3 while panel b varies from roughly 0-20ug/m3. Aren’t these the same data?

Figure 3: The amount of data presented in Figure 3 is very impressive and certainly

worth publication. That said, since you have already established that there are good correlations between the different mass concentration methods, I suggest only showing impactor data, since it is the direct “standard” (panels a,c,e,h), in the main body of the paper and moving the rest to the SI.

Figure 3: I would consider adding a line representing the Laakso (2003) results for each of the PM1, PM2.5, and PM10 plots. The text continually emphasizes comparisons to this work so it would be helpful to graphically show those values in Figure 3.

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