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Atmospheric
Measurement
Techniques
Discussions

Interactive comment on "Multi-Year ACSM measurements at the Central European Research Station Melpitz (Germany) Part I: Instrument Robustness, Quality Assurance, and Impact of Upper Size Cut-Off Diameter" *by* Laurent Poulain et al.

Anonymous Referee #2

Received and published: 3 March 2020

The paper by Poulain et al. reports on long-term ACSM measurements at an established central European location and discuss different aspects of measurement quality assurance. The paper is well developed with data thoroughly analysed and as such is worth publishing in AMT. However, there are several issues to be resolved before it can be accepted for publication. Last but not least English of the paper should be improved following numerous suggestions. Special attention should be given to Figure captions which are often inconsistent and awkwardly worded.

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Major comments

Particle mass closure is already a routine requirement requested by reviewers when considering different aspects of AMS/ACSM performance. But what is missing in the abstract and little attention is given in the paper, is the impact of RH and inlets (impactor type or cyclones) affecting comparability. There is no question of the usefulness to cross-checking ACSM with online PM mass or offline PM speciation, but this paper should take a much more comprehensive approach to sampling inlets as crucial factors for maintaining stability and consistency of online chemical speciation monitors, more specifically isokinetic sampling and wet/dry cut-off effects in size selective inlets.

Page 3. Line 26. Volume cannot be converted to mass without the use of average density which is derived from ACSM/AMS measurements making the MPSS derived mass and ACSM-MAAP mass the dependent variables. Instead, ACSM/AMS and MAAP mass can be correctly converted to volume, because individual species mass and density is known (with some exception of organics and black carbon perhaps) making no prior reference to MPSS and keeping both variables independent of each other. All of the above assumes fully internally mixed aerosol which may not always be the case necessitating AMS and BC size distribution.

Page 10. Line 22. I do not follow this reasoning. Sulfuric acid is a stronger acid and ammonia is preferentially neutralizing stronger acid when compared to nitric acid. Therefore, it needs to be checked for degree of neutralization taking into account that ammonium ion has to balance sulfate first and only then the nitrate. Small amounts of organosulfate or organonitrate do not violate the above general pattern at significant sulfate and nitrate concentrations.

Line 30. Estimation of the ratio can only be done after validating ACSM/PM1 agreement which should be done by an independent variable. Otherwise what is the reasoning of using ACSM/PM1or2.5 comparison for sulfate uncovering discrepancies, but assuming that ACSM OM measurements are fine and ready for deriving OM/OC ratio? Later the authors do comparison by retrieving OC from f44, but that is rather arbitrary given little confidence with regard to AMS measurements done elsewhere and with different instrument. In any event the authors should not start OM comparison going straight to discussing OM:OC ratios before all other technical matters were discussed. And I doubt that ACSM/PM1 comparison can inform about the OM:OC ratio, only AMS/PM1 can, because only AMS can provide OC mass after laborious data processing. Page 11. Line 2. Very high ratio of 2.74 (or even higher observed by Ripoll and Minguillon) suggests that ACSM is probably overestimating OM as well as in the case of sulfate or nitrate with the remainder attributed to OM:OC ratio. Can the authors support such a high oxidation ratio from the literature? I would be very curious to see those studies from the ambient atmosphere as even theoretically the ratio should not exceed ~3 (CO2 would have 3.66 and it is a gas). It is irrelevant that seasonal OM:OC ratios make sense if they are made from averaging unrealistic values. If so, the authors should not talk about OM:OC ratios before examining slopes carefully and discussing what is and is not theoretically possible. Perhaps consideration of CDCE by neglecting OM is a likely source of overestimation too.

Minor comments

Page 2. Line 7. Spell MPSS for those who read abstract only.

Line 25. . . .composition

Page 3. Line 8. Ovadnevaite et al. have published a rare study that reports...long-term... Do authors suggest in the following sentence that several years (e.g. three) is not sufficiently long period, but their own five year long period is sufficiently long? Please reword or clarify.

Line 18. within the scope, not focus

Line 33. TEOM-FDMS never provides PM1 mass and, even worse, separation in size typically occurs at ambient RH while the mass measurement occurs after drying at

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room temperature.

Page 4. Line 9. the impact, not effect.

Line 18. Is the RH actively monitored or indirectly maintained? Is the aerosol flow divided/split isokinetically or randomly? Significant losses can occur if split randomly due to different instrument flow rates.

Page 5. Line 14. ACSM data capture was 80% during deployment at Melpitz.

Line 18. CDCE is Composition Dependent Collection Efficiency as used in the original Middlebrook et al paper. It is also important to note that CDCE algorithm does not take OM into account which can be a potential error source.

Line 23. . . .collect particles with size selective PM2.5 and PM10 inlets on preheated...

Line 24. Samples were collected on a daily basis...

Page 6. Line 15. It is probably meant by "within Planetary Boundary Layer", but I do not quite understand what is meant by "above 500m" and why.

Page 7. Line 2. ...mass concentration comparison is suggesting overestimation by ACSM when compared to offline PM1.

Line 6. influenced by three specific periods in January 2015 and February 2017 (...).

Line 10. The overestimation can be caused by either size selective inlet or(and) sodium sulfate mainly residing in coarse particles. However, I am not sure that is good enough explanation of very contrasting comparison between PM1 and PM2.5. Judging from slopes (1.45 for PM1 and 0.68 for PM2.5) that is suggesting 100% difference between PM1 and PM2.5 - that is massive and not necessarily related to ACSM.

Line 18. That is correct theoretical explanation, but given huge discrepancy during "outlier" days the organic fragment signals should be of very significant magnitude to inorganic ones. Do authors have hints from the multiple campaigns when high resolution AMS was deployed at the site?

Line 27. coincides, not corresponds.

Line 31. PM1 cannot contribute to PM2.5 it is an inherent part of it. Is it not simply a ratio?

Page 8. Line 1. Consider that wet PM1 size segregation will inevitably pass less submicron particles. However, if that is the plausible culprit then ACSM should perfectly compare with PM2.5 which even in wet cut-off conditions should well correspond to dry ACSM PM1.

Line 5. The effect may still be there, but masked by the evaporation effect and NaNO3 effect on filters. Is this sentence out of place as it is becomes confusing whether the following sentences regard to sulfate or nitrate?

Line 19. Please explain what air mass density means.

Line 20. This confirms the predominantly shallow PBL...

Line 28. ...to slightly overestimate... The overestimation is very small and within the error margin compared to massive overestimation in Minguillon et al. I see this as a contrast from previous studies, not similarity.

Page 9. Line 5. I see more evidence in Figure 5. First, a lot of nitrate evaporation is visible at low ambient temperature as many of the points are clearly above ratio of 1.0. That is not surprising considering evaporation from particles already collected on the filter. However, the effect in summer seems to be larger, but concentrations are much lower in summer suggesting that evaporation can be near constant (as long as filter temperature is maintained similar during all seasons which needs to be confirmed). As more nitrate loaded particles are deposited on filter during winter they become buried under new layers of particles before being significantly evaporated. During summer there is little nitrate on particles and less particles altogether resulting in more absolute evaporative losses until particles buried under the new layers of particles.

Page 10. Line 20. ...with off-line measurements fall somewhere in between the two previously discussed ions.

Page 12. Line 10. in another location.

Line 25. The first comparison is correct by comparing mass with mass, but the second comparison is not, because particle mass derived from PNSD and MPSS will rely on ACSM for estimating average density making the two variables dependent. Therefore, only volume, not mass comparison of ACSM-MAAP and PNSD can be done correctly.

Page 13. Line 3. Therefore, it is not possible to conclude whether....depends on the location or the presence of larger coarse mode.

Line 9. . . .cannot typically account for the entire mass

Line 12. It is inappropriate to arbitrarily choose specific OM:OC ratio when the above paragraphs discussed wildly different ratios.

Line 32. Incorrect method. See above.

Page 14. Line 24. This artefact may plausibly explain the seasonality of the mass closure (it should be volume closure instead).

Page 15. Line 1. By the same principle ACSM size range is 1um/1.6=0.625um (dvac/density=dmob) and discrepancies between size ranges of ACSM and MPSS should be irrelevant. Please reconsider and reflect in conclusions.

Line 31. Can this be called an artefact? It depends whether organo-nitrate belongs to organics class or nitrate. Nitrate functional group is still a nitrate even if bound to organic species. Quite contrary, off-line inorganic and offline OC/EC analysis completely misses nitrate bound to organics making the use of the term “artefact” justified. Ideally, one would want a clear distinction of organo-nitrate compound which would be out of reach by off-line inorganic techniques unless specifically measured for OrgNO3.

Figure 1. I recommend changing PM to ACSM-MAAP or ACSM+eBC for consistency

with further Figures.

Figure 2. ...and solid black lines represent regression fit by least orthogonal distance (y=a+bx)

Figure 3. change "data coverage" to "data capture".

Figure 4. ...for days where sulfate concentration difference between PM2.5 and PM1 exceeded 1ug/m3: (a) overpassing trajectory density; (b) potential source contribution function??; (c) time series of sulfate concentration difference, PBL height above 2000m, precipitation events exceeding 1mm/h and PBL formation above the station altitude.

Figure 6. ...and off-line OC PM1 (a); seasonal variability....(b) and the entire time series (c).

Figure 7. Awkward Figure caption. Please rewrite according to suggested above.

Figure 9. Incorrect graph as it should be volume comparison. Awkward Figure caption. Please rewrite according to suggested above.

Figure S1. Make PM axis consistent with further Figures

Figure S2. ...to daily PM2.5 mass.

Figure S5. ...corrected for organo-nitrate contribution during days with PM1:PM2.5 < 0.6.

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