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Comment on amt-2022-206

Anonymous Referee #1

Referee comment on "The AERosol and TRACe gas Collector (AERTRACC): an online-measurement-controlled sampler for source-resolved emission analysis" by Julia Pikmann et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2022-206-RC1>, 2022

The submitted manuscript by Pikmann et al introduces a new, automated offline sampler for both aerosols and gases called the AERTRACC, which is part of the mobile laboratory platform MoLA. The stated aim in its design is to allow for more efficient source separation when sampling complex source environments, specifically for the analysis of organic aerosol (OA) and their precursors. The manuscript describes the hardware and control software in detail, and then demonstrates its operation with a (static) single source experiment. That includes an in-depth analysis of gas and particulate phase tracers detected by their analyzer of choice (an TD-CIMS) and the source-specific contrast ratios observed. The authors conclude with a detailed discussion of the optimal choice of input parameters to control the sampler, in order to increase source contrasts for this particular case. So while there is no data shown from actual mobile sampling, the reader is reasonably reassured that it will in fact work for that scenario.

The overwhelming majority of offline samplers operate on some type of predefined time-grid, which leads to different airmasses and hence sources and chemistries being integrated into one sample. For mobile sources where air mass variability is higher this can limit the usefulness of offline analysis significantly. Hence, on airborne platforms, most offline samplers (such as whole air samplers) are operated based on some type of expert assessment of airmass change, which in (some, rare) cases is automated. While aircraft payloads are highly variable from mission to mission, that is obviously not the case for the MoLA platform, which has a well-defined instrument payload. Therefore, it makes sense to not only build a "smart" offline sampler for MoLA, but to automate it based on all the possible ancillary measurements available. So while this type of sampling has been done before, this paper shows a technically proficient, well-executed implementation of it for a platform that will certainly make good use of it in the future. So this is well within the scope of and of general interest to the AMT audience. However, I have some concerns about the way the work is framed and about the details of the analysis, which are described in the following:

- Unless I am completely misunderstanding the work presented, the key aspect of this sampler is that it switches between “source” and “background” conditions many, many times over the course of the experiment. This is quite different from most offline samplers where only one continuous sample is taken. I would recommend making this much clearer in the description, and possibly illustrating this by e.g. showing the actual sample times on top of Figure S5. It would also be good if the authors discussed what the minimum sample time (per valve switch) actually is, and how this could possibly impact the overall performance once MoLA is actually moving and hence the source sector segments become smaller.
- Related to the last item, can the authors please discuss what the limitations are on leaving the “source” channel idle for long periods between source interceptions? My specific concern is that any type of filter substrate has issues with loss of volatiles (e.g. Heim et al, 2020), and while during active sampling these “losses” will likely end up in the TDTs, once the line is idle they will simply evaporate and possibly diffuse/deposit to the “background” channel. And there is also, specifically for CIMS measurements, obviously the oligomerization problem (e.g. Lopez-Hilfiker et al, 2015), which should at least be acknowledged.
- I do not think that the current introduction does the best job in framing the significance and applicability of AERTRACC. First of all, I think the sampler needs to be separated more clearly from the specific application (in this case, molecular OA composition). So by way of example, the current AERTRACC, with a simple change in filter media would surely be of high interest to single-particle TEM folks, a field where optimizing contrast for the source-specific aerosol types of interest (and not necessarily organic ones) can be extremely challenging. So framing AERTRACC as an efficient way of solving the contrast issue for any type of off-line analyzers will better illustrate its potential.
- More importantly, this statement (L79-80) “Currently, no instrument offers detailed chemical analysis of aerosols in real-time for the analysis of individual sources (Parshintsev and Hyötyläinen, 2015)” which is the linchpin of the current introduction, is simply incorrect. There are real-time, 1 Hz capable molecular OA detectors, such as the EESI-ToF (Lopez-Hilfiker et al, 2019, Pagonis et al, 2021) or the CHARON-PTR-MS (Eichler et al, 2015, Piel et al, 2019). None of them are perfect (no instrument is), but the point is that molecular identification of OA from mobile sources can be accomplished in many different ways, and AERTRACC+TD-CIMS is just one of many, and not really the main reason (in my view) for AERTRACC.
- What both the introduction and the rest of the paper do not discuss are detection limits, which are typically one of the main design criteria for samplers. This needs to be addressed, since currently there is not context whatsoever on what a reasonable total sample time would be (and hence how practical the use of AERTRACC is for typical urban source applications). Based on the description, under typical polluted urban conditions ($\sim 10 \mu\text{g m}^{-3}$ OA), it would take about 2 hours to sample 1 μg of material, which given typical DLs for organic compounds in the FIGAREO ($\sim 0.5 \text{ ng sm}^3$ from filter blanks) seems sensible. But that seems like a long sampling time for a local source, so a discussion of the possible tradeoffs would be appreciated.
- The (nicely done) PMF analysis of the AMS measurements seems a bit underutilized at the moment. Its sole purpose currently is to show the wind dependence for the OA sources, which could certainly be showcased with less work. Obviously it is an ancillary measurement, and not part of AERTRACC, but Figure S1 shows a lot of high-time resolution, high contrast data that could be certainly utilized to assess what ideally AERTRACC can achieve. So I would encourage the authors to add the PMF factors to Figure 5 (and Tables S3-S5), and the sampling periods to Figure S1, which should make current trends clearer, at the very least. In that context (assuming the AMS is a regular instrument on MoLA), the authors could also consider adding some discussion of to what extent the ancillary variables used in the last section could be augmented by using the f43, f55, f57, f60 from the AMS real time feed.

Minor comments:

- The manuscript often uses "aerosol" when it really means "aerosol type", suggest rephrasing
- L29: "Within multiphase processes aerosol interacts with atmospheric gases forming new substances". While this is certainly true, in many (most) cases the chemistry happens in the gas phase and the products end up in the particle phase by simple partitioning, so consider rephrasing.
- L36: Seasalt (as well as dust) is by far the largest primary aerosol type by mass, consider revising
- It's a minor point, but I find the distinction between the FIGAREO-CIMS and the "TD-HR-ToF-CIMS" not helpful and mostly distracting. It's the same instrument, and per the AERTRACC description the authors are even using the FIGAREO inlet for their filter desorption. So it is the same instrument, and the authors are just using it in offline mode, no need to suggest that there is a difference beyond that.
- L105: "non-refractory chemical composition of submicron particles". This sounds as if the AMS is part of the standard MoLA suite, but that's not what the Table reflects. Please make it consistent.
- Just a general comment: it seems that the flows are simply checked before startup and then left to their own devices. Given that a typical sample day is probably 4-6 h before the TDTs and filter holders are taken out, relying on the needle valves alone seems fine, but there could still be some variations that could potentially lead to turbulence while valve switching. So some monitoring equipment might be wise...
- Section 2.3: It should be stated in what language the software was developed (Igor Pro, I assume) and also what type of license is used for it.
- Section S1: What organic density is used? 1.4 g/cc or the variable one based on Kuwata et al (2012). For source studies the latter seems like the better choice

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