08/19/2022

RC1: 'Comment on amt-2022-185', Anonymous Referee #1, 16 Jul 2022

Author statement: The authors thank the referee for reviewing this manuscript. An itemized **response** for the rebuttal can be found below for each response given in blue. The tracked-changes version can be found below for each response in *red* and the already existing text is in *italic*.

General comments:

This paper introduces a modified version of a Handix POPS and demonstrates the performance improvement in measuring the aerosol number concentration, particle size and signal time responses. The improvement is promising and very important to expand the current POPS capability. In addition, the authors presented a very useful application to monitor the particle phase transition with the high-resolution light scattering amplitude data. Thus, the reviewer recommends publication after addressing the below concerns.

1. What is the modified POPS's total weight, power requirement, and cost? What is the targeted application of the modified POPS? Is it maintained the current lightweight and low-cost sensor application?

   Response 1: The following is added to the manuscript.

Manuscript:

Section 2.1:

The schematic of the modified POPS is shown in Fig. 1. A photograph of the modified unit is provided in Fig. 2.

Discussion:

We modified the commercial version of the POPS to add an MCA card for data acquisition and to modify the flow path to facilitate integration into complex flow systems. As shown in Fig. 2, the unit was placed inside a 19" rackmount enclosure. No effort was made to minimize the height and weight of the enclosure or the power consumption of the unit. The additional power requirement beyond the factory design depends on the vacuum pump. We used an ~400W model, but smaller pumps will be sufficient. The rackmount form factor is suitable for laboratory and field applications, including measurements from mobile platforms such as vehicles or airplanes where size and weight are less critical than in balloon-borne deployments. The cost to modify the factory supplied unit was ~US $6,500, including the MCA card, enclosure, vacuum pump, needle valve, power supply, Swagelok fittings, electrical connections, and particle filters. The data presented here show that both the pulse height …

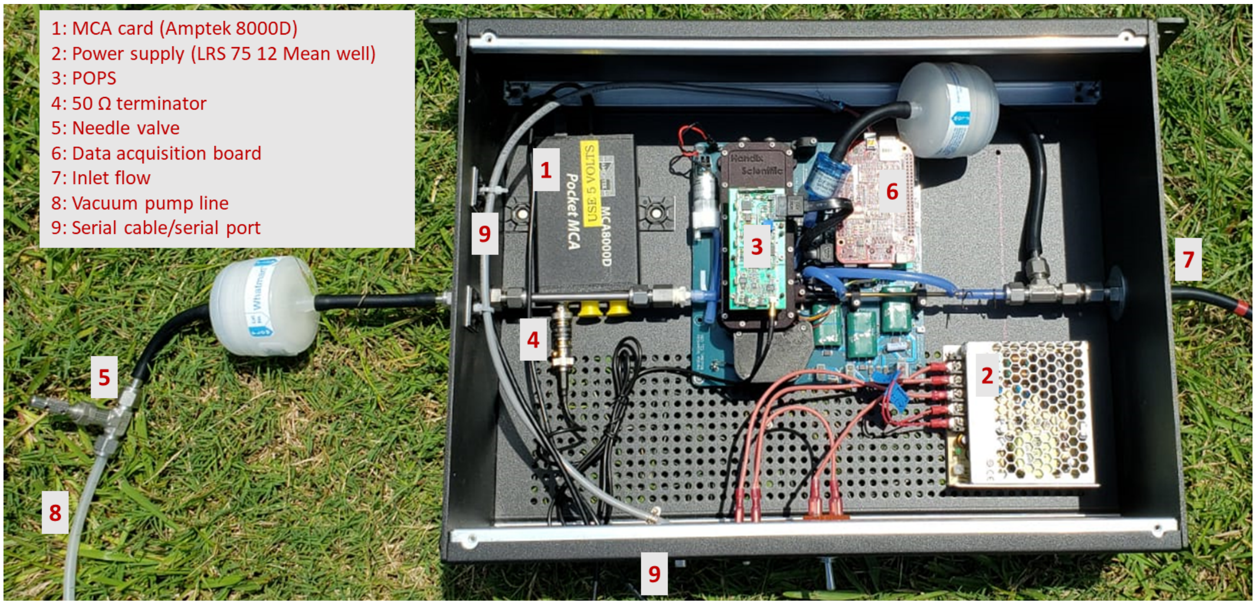

Figure 2: *Photograph of the modified POPS.*

When the modified POPS was integrated into a dual tandem DMA, how were the multiple charges treated in the data reduction?

> **Response:** The following was added to the manuscript

Manuscript: *To track the PH data across the transition from uncoalesced dimer particle to coalesced sphere, the Digitizer-PH was determined at ~±4% of the mode diameter of the dimer peak. Note that the mobility distribution of the dual tandem DMA is complex and contains multiple modes. Details are given in Petters (2018) and Rothfuss et al. (2019). The primary peak of dimer particles that formed from +1/-1 charged particles is identified by its mobility diameter of ~1.26*D_{mono}*. Near the peak of the distribution, interference from particles other than +1/-1 dimers is minimal (c.f. Figure 7 in Petters, 2018).*

How does the RH affect the light scattering calculation?

> **Response:** All experiments were performed under dry conditions. Thus an RH correction was not required.

Manuscript: no changes were made to the text.

Considering the complex light scattering responding curve, please estimate the uncertainty caused by the multiple change and RH in your results, such as in fig. 4. Additional information should be provided to quantify the sizing capability of the modified POPS.

Response: The sizing capability of the modified POPS is in principle identical to the original POPS, but with a smaller range when using the MCA card. Uncertainties due to RH and multiple charges are highly application dependent and are appropriately discussed in the context of applications where such corrections are needed. The following changes were made to the manuscript.

Manuscript: *Atomized particles were dried to < 25% relative humidity (RH) using 4 or 5 silica-gel driers in series. At RH < 25% the water content for organic particles such as sucrose is negligible (Power et al., 2013). Ammonium sulfate is effloresced at RH < 40% and thus contains no water.*

Specific comments:

Line 63, What time and bin resolution do the DMA-OPC experiments need? How does the 10 Hz data improve the result compared with the 1 Hz data?

Response Line 63: Precise requirements will depend on the application. Based on the results shown in Figure 5 of the revised manuscript, a resolution of 500 bins is sufficient to resolve the transfer function of the DMA, where the mobility diameter is translated into an optical signal. The MCA data is useful here since it doesn't saturate at high concentration. Ten Hz data acquisition in DMA-OPC experiments may be useful to resolve concentration changes for fast mobility scans. However, the exact scan rate where this benefit will occur was not the focus of our work and needs to be determined in future studies.

Line 97, What is the uncertainty caused by the sheath flow variation?

Response Line 97: The purpose of the sheath flow is to prevent the optics from accumulating particles. We believe that it is small/negligible. First, the sheath flow is also not actively controlled in the regular POPS. It is simply taken from the ambient line. Second, concentration is determined from the sample flow rate, and thus does not depend on the sheath flow. Third, concentrations between the POPS and a CPC agree for sufficiently large sizes. Finally, we did test the setup with sample flow (not reported here), and found no change in concentration or sizing performance.

Figure 2, At what locations does the author monitor the phase transition temperature? Will the particle temperature decrease when transferring from SMPS to POPS?

Response Figure 2: The following was added to the manuscript.

Manuscript: ... *particles are passed through a coalescence chamber which exposes the particles for a fixed time to elevated temperature conditions, which is measured at the outside of the metal chamber. Upon exiting the chamber, the sample is cooled to ambient conditions. This arrests the* *sintering process, allowing for the observation of partially coalesced particles. If the temperature is sufficiently high, the particles coalesce and transform from a touching bisphere dimer particle into a spherical particle.*

Line 155, Can the author distinguish the multiple charged particles with a POPS? If so, what is the size resolution of the modified POPS?

> **Response Line 155:** The multiple charges can be distinguished with POPS, as is discussed in Section 3.1. We estimate that relative changes in scattering intensity of ~2-3% can be resolved. The corresponding size resolution will depend on the refractive index, particle shape, and particle size/mass. The sizing capability of the modified POPS is in principle identical to the original POPS, but with a smaller range when using the MCA card.

Manuscript: *no changes were made to the text.*

Line 157, Do we expect the refractive index to change during this particle transition? If so, how can it affect the mode diameter determination?

> **Response Line 157:** We do not know if the refractive changes during the particle phase transition. However, the mode diameter is determined from the count vs. mobility distribution, like it is done for standard scanning mobility particle sizer measurements. Therefore, in this context the refractive index is not important.

Line 160, The viscosity of what chemical compound? Under what temperature?

Can the author quantify the enhancement?

Response Line 160: Manuscript: *The mapping between transition temperature and viscosity depends on particle size, surface tension, and residence time in the coalescence chamber (Rothfuss and Petters, 2016). The effect of surface tension on the mapping is small (Marsh et al, 2018). Here, the transition temperature corresponds to the condition when viscosity is ~10^7 Pa s.*

Line 265, does it suggest that the MCA modification narrows the size detection range of POPS? If it is true, that will limit the application of POPS. Is there any solution for that?

> **Response Line 265:** The MCA addition is not limiting the POPS application as the POPS serial and Digitizer-PH are able to capture lower and higher size detections while the MCA is attached (see Figure 5). It is in principle possible to use the MCA card output to measure the concentration and scale the Digitizer-PH size distribution accordingly. This requires the assumption that the coincidence only minimally affects the size distribution and the assumption that the concentration dynamics of < 200 nm particle is the same as for > 200 nm particles. This is also mentioned in the manuscript:

Manuscript: *Importantly, the addition of the MCA still preserves the Digitizer-PH output. Thus, the combination of the two may be used to reconstruct the size distribution from the Digitizer-PH using post-processing while scaling the concentration using the measured MCA-PH data.*

Fig 8. Does the author have a plot of the temperature scans of MCA-PH response? Is it different or similar to digitizer-PH?

> **Response Fig 8:** The selected particle size was too small to resolve changes in the MCA-PH response.

The nominal 166 nm is based on the electrical mobility size, right? If so, what is the corresponding optical size of the particles? Line 275, at 72 C, the mobility size is around 210 nm, similar to the doubly charged particles. How does the author separate the coalescence and the doubly-charged particles?

> **Response:** We do not explicitly map mobility and optical size in this paper as this mapping is sensitive to the refractive index. Based on the calibration shown in Figure 5, this size is at the limit of detection of the instrument. Particles with 166 mobility diameter are also charged and filtered by the electrostatic precipitator. The size of doubly charged particles is 261 nm, which is much larger than the dimer size. Furthermore, doubly charged particles transmitted by the size-selecting DMAs carry charge and thus are also filtered by the electrostatic precipitator. The following is added to the manuscript to refer the reader for further details:

Manuscript: *Note that the mobility distribution of the dual tandem DMA is complex and contains multiple modes. Details are given in Petters (2018) and Rothfuss et al. (2019). The primary peak of dimer particles that formed from +1/-1 charged particles is identified by its mobility diameter of* $\sim 1.26D_{mono}$*. Near the peak of the distribution, interference from particles other than +1/-1 dimers is minimal (c.f. Figure 7 in Petters, 2018).*