“Evaluation of a Low-Cost Dryer for a Low-Cost Optical Particle Counter”

Response to Report #1

I commend the authors for the substantial improvements and clarifications they have made in this revision. I think the potential utility of their system is highlighted and the experiments and data are generally clearly articulated. Most of my concerns have been addressed, but there remain some issues… In my view, the limitations of the system as it was operated in this study are not clearly enough stated using the data they have and insights from other work. It is important to both show that this is a contribution but also be very clear about its limitations. The most important limitation in my mind is that it is clear that the aerosol is being heated far more than it should be, and that this is likely causing excess water, and perhaps more importantly, any semivolatile components (ammonium nitrate and organics, most notably) to evaporate. The revised manuscript makes it clear this is an issue, but several times calls it ‘over drying’ (it’s not drying, but drying and evaporation) and does little to say why/by how much. For example, the supplement contains data (Fig S3) that demonstrate this issue (extensive evaporation of ammonium nitrate particles in the heater and in the IADS), but the manuscript simply mentions the figure (line 387) without discussing it and its implications. Fig S3 shows a ~5-fold under prediction with the dryer and ammonium nitrate (Fig S3), which will introduce huge bias if this or organics are dominant components of PM. In fact, in the phase I field measurements you specifically note that high concentrations of ammonium nitrate (which is hydroscopic, but also semivolatile) are expected. The degradation in PM2.5 performance with the heater is likely due to this. My feeling is that this issue needs to be addressed directly.

We thank the reviewer for his comments and the time spent in undergoing the review process. In the new version of the manuscript, we have discussed Fig. S3 and added its implications. The text is as follows:

“An experiment with NH_4NO_3 particles is shown in Fig. S3, in which different IADS temperatures were manually set (20 °C, 35 °C, 50 °C and 65 °C). This experiment clearly shows the impact of temperature in the loss of semi-volatiles due to evaporation and, therefore, the detrimental effect that the use of high temperatures in a heated inlet can have on the mass concentration when species with high volatility like NH_4NO_3 are present in the sample. In this sense, the presented design of a low-cost dryer is behaving as a thermodenuder, i.e., a device that is used to study the volatility fraction of aerosol particles (Huffman et al., 2008). Studies using thermodenuders have shown that temperatures of 83 to 88 °C can cause 50 % of the organic aerosol mass to evaporate (Paciga et al., 2016). For the specific case of nitrate, much lower temperatures are needed to reduce the mass by 50 %, as it is shown in the results of Huffman et al. (2009) where 50 % of the nitrate during a field campaign was evaporated at 54 °C.”

Regarding the field measurements, we agree with the reviewer and think that the degradation of the PM2.5 performance during the field measurements is likely due to the evaporation of the semi-volatile species, as already stated in the previous version of the manuscript: *“... the PM2.5 is frequently underestimated. This underestimation occurs probably due to two reasons: (1) most of the semi-volatile organic compounds belong to the PM2.5 fraction and the dryer could be evaporating them.”* Additionally, we have added the following sentence:

“It is likely that reason (1) prevails over (2), as a significant amount of ammonium sulphate and, especially, ammonium nitrate, is expected in the PM2.5 fraction.”

In my first review I pointed out an example of the use of a heated tube as a thermodenuder, which is essentially what this is, and how it may influence aerosol mass transmitted to the sensor. Considering the high temperature (nearly 70 deg. C) reached in the heater, all results should be viewed in this context (how much bias may be introduced), rather than just including a caveat that this is an issue. However, no mention of this literature or the data that was collected showing this to be an issue is included. Another paper, which shows similar data for nitrate is (Huffman et al. 2008), or for lab-generated ammonium nitrate see Fig 4a here: (Huffman et al. 2009).

We have included a comparison of the low-cost dryer with a thermodenuder and the mentioned literature following the discussion of Fig. S3.

In a related issue, my suspicion is that the results in Fig. 4 a and 4b are being misinterpreted, which are as much showing evaporation of particles and not removal of water. The fact that the Fidas and the undried sensor in Fig. 4a seem to agree suggests that the particles formed during the humidification stage have not taken up excess water (otherwise you'd see a big difference) and what you're seeing when the heater turns at 65 minutes on is evaporation of semivolatile material in the particles. This is confirmed in 4b, where you see that heating to 70 deg. C, much more than needed to remove water, is greatly reducing the Fidas signal and making it align with your heated sensor data.

After further inspection of the literature, we agree with the reviewer on the fact that the ultrasonic humidifier is also generating particles that have their origin in the minerals present in the tap water. These impurities are mainly calcium, magnesium and sodium among other compounds. That means that what we probably measured was the evaporation of water that condensed on the impurities, which had acted as condensation nuclei. In the laboratory experiments carried out by Lau et al. (2021), a diffusion dryer is used to dry the water droplets before sample collection which means that he was also concerned that the impurities were not fully dry and that a determined content of water remains absorbed in the residuals.

For clarification in the manuscript we have included the following sentences:

"As shown in Fig. 4a during the experiment with the IADS in automatic mode, once the air humidifier was on, tiny water droplets containing impurities were generated. The water droplets evaporated quickly and, as a consequence, the RH started to increase, leaving the solid impurities with associated water as suspended particles in the air."

"…a remarkable increase in the PM2.5 concentration was observed, possibly due to the water uptake of the impurities."

"Once the RH reached 65 %, the low-cost dryer of the OPC-R1 started heating and a pronounced decrease in the PM2.5 concentration is observed, probably due to not only the evaporation of the water but also the evaporation of semi-volatile species."

As I noted in my first review, you are not measuring fog droplets in the lab experiments as claimed on lines 240 and 347 and elsewhere in Sec. 3.1.1, but measuring mineral residue with some associated water. Even deionized water has some residuals, and these are not 'filtered' by the atomizer. You are atomizing water that has some dissolved solids (not 'pure water'), and even very low concentrations of this dissolved materials makes particles. See for example Fig. 2 and Fig. 3 in the paper you cited in your response (Sain et al), which includes data for a humidifier using nanopure water – a far more effective purification than whatever 'filter' is included in the humidifier might accomplish. The humidifier may be emitting fog droplets, but these appear to/should quickly dry to increase room RH unless the air is saturated, thus are no longer 'fog droplets', and you are left with residual particles, not droplets.

As explained in the previous answer, we now agree with the reviewer. It should be mentioned that the content of impurities in the water during the experiments does not constitute a problem as the natural fog is also not pure water. We have added the following sentence for clarification:

"This model of humidifier integrates a filter unit (250 AQUA PRO) that allows the generation of water droplets with a lower concentration of impurities than compared without a filter. The impurities in tap water consist mainly of calcium, magnesium and sodium, which are responsible for the characteristic "white dust" generated by ultrasonic humidifiers (Sain et al., 2018). Moreover, fluoride, nitrate, phosphate, sulphate, aluminium, copper, and iron, among other species, can also be found in different quantities depending on the water quality (Yao et al., 2020; Lau et al., 2021). These impurities act as condensation nuclei retaining part of the water in the liquid phase, just as fine, suspended particles do during the fog formation in ambient air."

Minor points

Fig S6 – no temperature on x-axis

It has been added.

Fig 8 and others seems to show smoothed data, as noted not appropriate for discretely measured data.

Fig. 8, Fig. 4, Fig. 6, Fig. S3 and Fig. S4 are now unsmoothed.

References:

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