Response to interactive comments from Referee #3

Thank you for the time you put into reviewing our manuscript and the helpful feedback. Please find in the following our responses and proposed changes to the original manuscript, which improve the manuscript. Below the comments from Referee #3 are given in black. Our responses to the comments are shown in blue. Text added or changed in the manuscript is marked in italics.

The paper presents the application of a heated inlet to mitigate against the influence of hygroscopic growth of particles on the imputed particle size distribution and mass loading by an OPC, in this case the 'lower-cost' OPC-R1. The paper is well written and the procedures are mostly clear, though considering that this seems to be an 'open-sourced' project, it would be helpful to have more specific information on the construction of the heater – more information on electronics is included in the linked Github page, but not sufficient to recreate the design without significant leaps. However, my main concern is that it the interpretation of results is such that it is difficult to know whether such a device will actually improve measurements by low-cost sensors, or introduce other, possibly more difficult to correct errors. Additional experiments that seek to separate 'drying' from 'particle evaporation' (by varying particle volatility or treatment mechanisms) could do this, but are a substantial extension beyond what is included here.

We have updated the document for Zenodo/Github with more information about the dryer construction and the electronics.

Regarding the interpretation of the results, we have now included results of field campaigns where the PM concentrations of the low-cost dryer are compared to gravimetric analysis and "reference-equivalent" monitors during "real" conditions of hygroscopic growth and fog.

In general, I share similar concerns to the previous two reviewers, and offer some specific points that should be addressed below. I am in particular concerned about the influence of and lack of 'control' on the temperature of the / sample and the fact that it may be as high as 70 degrees C. We don't really know what the temperature it is, but it appears it may be high as this is when the most consistent behavior with the reference instrument is observed (when the Palas was operated at a fixed temperature). The paper (rightly) includes multiple allusions to the potential for loss of semivolatile material, and this is indeed a major concern for application of this system for ambient aerosol, especially in urban areas where most lowcost sensors are deployed. Heated tubes of this type are used as 'thermodenuders' to remove semivolatile organics, and previous deployments in urban areas find that nearly 50% of organic aerosol will be removed by heating to 70 C (Paciga et al. 2016), though with a longer residence time (I calculate around 8 seconds for your heater geometry vs. 50 seconds for the Paciga et al system). If organic aerosol or ammonium nitrate are substantial components of the sampled aerosol, this heater will remove much of this material and bias any measurements low. The removal of water and semivolatile removal components could have been isolated by running experiments with diffusion or Nafion drivers in parallel with the heated tube. However, in the absence of such data and especially in the absence of information about the actual temperature of heating, it is (and will be) hard to interpret what the heater is actually doing to the aerosol. This is especially tricky because much of fine aerosol number and mass will be in the submicron range, and evaporation will push the entire size distribution out of the range of diameters the OPC(s) can detect.

We have added a new section 3.1.3 "Study on the drying temperature" where we include more information about the air temperature in the dryer. The drying process in our prototype is complex, due to the discontinuity of the dryer but, it is actually as the reviewer assumed, that temperatures higher than 40 °C are reached when the dryer is on and that therefore part of the semi-volatiles is lost. How much time the dryer is on depends on the ambient temperature which makes difficult to control the temperature. In order to address this problem the following text has been included:

"To get more information about the temperature profile inside the dryer, experiments were performed in the laboratory where the temperature of the air flowing inside the dryer was measured. The experiments showed that the maximum wall temperature is reached at 40 cm (Fig. S6). In the last centimeters the air is cooled down before the sensor inlet due to the lack of heated wire (the last 2.5 cm were left wire-free for ease of handling). It was observed that at 40 cm the air is heated up to approx. 65.9 ± 0.5 °C. This is in agreement with the experiments which show that the sensor with low-cost dryer behaves similar to reference instrument if the IADS is heated at 70 °C. As the thermocouple influences the air flow, the measured temperature may have some bias, but it is clear that it is higher than 40 °C, which is the maximum temperature recommended by the WMO/GAW guidelines for ambient air monitoring. Moreover, it was observed that the T_{OPC} *is usually 10 – 13 °C higher than*

the ambient temperature, which means that the dryer may not start heating when the ambient temperature is higher than 22 – 25 °C, as the T_{OPC} could be already higher than the temperature limit set for the dryer (35 °C). This problem could be solved by changing the upper limit temperature loop in the Arduino code. However, this change also increases the maximum air temperature in the dryer, which is already too high for producing "reference-equivalent" PM readings. Therefore, we recommend that new versions of the low-cost dryer should focus on the control of the RH in the sample flow, as the T_{OPC} value is highly dependent on the ambient air temperature."

There is a strong basis for the limitation of inlet temperature to 40 C in 'reference' instruments (as noted by WMO/GAW guidelines) due to the influences discussed above. For example, early studies with the 'TEOM' found a strong and variable bias due to loss of semivolatiles due to heating (typically to 50 C) (Allen et al. 1997; Charron et al. 2004) and later versions did away with this heating.

As mentioned in the previous comment, we have addressed this problem in a new section (see comment above).

We have added a new Discussion section in which this topic is addressed in the following sentence:

"Additionally, a temperature limit of 40 °C should be introduced, as recommended by the WMO/GAW guidelines."

The 'fog' measurements, don't appear to really be fog, but are likely residual contaminants from the humidifier. This is evident by the small size (as noted by the authors) relative to actual fog droplet. There's a wide literature on this, going back decades (e.g. (Rodes et al. 1990)). Therefore, while the application of this heater systems for aerosol measurements in foggy environments may be a goal, it doesn't appear to be one tested here. Rather, these are similar experiments to the others shown, but with aerosols of unknown composition.

We were aware that the mineral composition in the water could have an effect in the experiments. Consequently, we selected the humidifier U350 that integrates a filter unit (250 AQUA PRO). Moreover, test experiments were performed using deionized water without a significant difference in the results. We have also double-checked with recent literature and the particle distribution we obtained corresponds to what other researchers have obtained for deionized water using ultrasonic humidifiers (Sain et al. 2018). We have added the following in the manuscript:

"This model of humidifier integrates a filter unit (250 AQUA PRO) that allows the generation of pure water droplets."

The results in Figure 6 may be helpful to separate 'drying' from 'evaporating' because they are with a non-volatile aerosol at known RH. However, they are still difficult to interpret. I expect this is because there are interactions between different drying conditions (the low-cost dryer is probably 'over drying' relative to reference conditions) and the size cutoff of the two OPCs (the R1 may be 'missing' a substantial amount of material between 0.18 and 0.3 microns).

The reviewer is right, the OPC with dryer is over drying relative to the Fidas® 200. We believe the new section "Study on the drying temperature" help to interpret the drying process in the low-cost sensor. We have also added the following sentence:

"It should be also highlighted that approx. 80 % of the particles seen by the Fidas® 200 have a mean diameter from 0.17 to 0.35 µm, which means that the OPCs are not detecting a substantial amount of material."

The paper alludes to testing various inorganic aerosols (Line 111) but these data aren't included. In particular, tests with ammonium nitrate would likely highlight the influence of heating on semivolatile material. This additional data should be included/discussed.

We have included one experiment with ammonium nitrate as well as an experiment with a mixture of the aerosols in the supplemental material.

Specific points

L15 – Not clear that comparing average PM2.5 concentrations are appropriate comparisons. Other distribution parameters and comparative statistics are helpful.

We have included more statistics for the field measurements and updated the full abstract to the following:

"The use of low-cost sensors for air quality measurements has become very popular in the last decades. Due to the detrimental effects of particulate matter (PM) on human health, PM sensors like photometers and optical particle counters (OPC) are widespread and have been widely investigated. The negative effects of high relative humidity (RH) and fog events in the mass concentration readings of these types of sensors are well documented. In the literature, different solutions to these problems - like correction models based on the Köhler theory or machine learning algorithms - have been applied. In this work, an air pre-conditioning method based on a low-cost, thermal dryer for a low-cost OPC is presented. This study was done in two parts. The first part of the study was conducted in laboratory to test the low-cost dryer under two different scenarios. In one scenario, the drying efficiency of the low-cost dryer was investigated in the presence of fog. In the second scenario, experiments with hygroscopic aerosols were done to determine to which extent the low-cost dryer reverts the growth of hygroscopic particles. In the second part of the study, the PM10 and PM2.5 mass concentrations of an OPC with dryer were compared to gravimetric measurements and a continuous Federal Equivalent Method (FEM) instrument in the field. The feasibility of using univariate linear regression (ULR) to correct the PM data of an OPC with dryer during field measurement was also evaluated. Finally, comparison measurements between an OPC with dryer, an OPC without dryer and a FEM instrument during a real fog event are also presented. The laboratory results show that the sensor with the low-cost dryer at its inlet measured an average of 64 % and 59 % less PM2.5 concentration compared to a sensor without the low-cost dryer during the experiments with fog and with hygroscopic particles, respectively. The outcomes of the PM2.5 concentrations of the low-cost sensor with dryer in laboratory conditions reveals, however, an excess of heating compared to the FEM instrument. This excess of heating is also demonstrated in a more in-depth study on the temperature profile inside the dryer. The correction of the PM10 concentrations of the sensor with dryer during field measurements by using ULR showed a reduction of the maximum absolute error (MAE) from 4.3 µg m^{-3} (raw data) to 2.4 µg m^{-3} (after correction). The results for PM2.5 make evident an increase in the MAE after correction: from 1.9 µg m^{-3} in the raw data to 3.2 µg m^{-3}. In light of these results, a low-cost, thermal dryer could be a cost-effective add-on that could revert the effect of the hygroscopic growth and the fog in the PM readings. However, special care is needed when designing a low-cost dryer for a PM sensor to produce FEM similar PM readings, as high temperatures may irreversibly change the sampled air by evaporating the most volatile particulate species and thus deliver underestimated PM readings. New versions of a low-cost dryer aiming at FEM measurements should focus on maintaining the RH at the sensor inlet at 50 %, and avoid reaching temperatures higher than 40 °C in the drying system. Finally, we believe that low-cost dryers have a very promising future for the application of sensors in citizen science, in sensor networks for supplemental monitoring, and for epidemiological studies."

L18 – Here and elsewhere there is discussion of 'accuracy' of sensors, but nowhere is data from this arrangement compared to a 'true' reference measurements, and so this seems a bit of a bold claim.

We agree that the results are difficult to interpret in the context of standard reference measurements without comparing with the "true" reference. Therefore, we have added new results that includes a comparison of the OPC-R1 with gravimetric measurements during field deployment.

L31 – Accuracy is one limiting factor, but there are other key concerns: power consumption, durability, … . What are 'certain applications'?

We have changed it to the following text:

"The accuracy needed for certain applications e.g. regulatory air quality monitoring or environmental epidemiology is at this moment one of the limiting factors for the use of low-cost sensors."

Figure 1- These data traces appear to be smoothed. This is not appropriate for discrete data points.

Plotted data in Fig. 1 are now unsmoothed.

L76 – It's not clear what 'quality' means here. A key considering is that the variable space covered by training and deployment data sets coincide.

We have changed it to the following sentence:

“However, they also have limitations such as the high dependency on the quality (accuracy of all input variables, outlier detection) and length of the training data and the extensive computational resources required.”

L138 – As noted above and by another referee, a lack of specificity about what exactly this temperature refers to is a key question. The key temperature for evaporation of semivolatiles will the air temperature within the heater, as semivolatile materials evaporated will be a function of this temperature.

As stated in a previous answer to a comment, we have included a new section about the air temperature inside the dryer and added some more information in the supplemental material.

Eq. 1 – This approach doesn’t seem helpful, as it combines differences in instrument response (e.g. due to different size cuts and calibrations) with ‘drying’ (which is actually drying + evaporation of semivolatiles)

Thank you for the note. We have made a modification in Equation 1 that we believe helps to understand the meaning of the drying efficiency: instead of calculating the term “$1 - \frac{\mathrm{PM2.5}_{d,i}}{\mathrm{PM2.5}_{r,i}}$”, only $\frac{\mathrm{PM2.5}_{d,i}}{\mathrm{PM2.5}_{r,i}}$ is now calculated. We have added also the following explanation:

“Each drying efficiency provides different information. The η_r gives an idea about how close the average PM2.5 readings are between the reference instrument and the sensor with low-cost dryer. In other words, the higher the η_r the closer the PM2.5 to “reference-equivalent” PM2.5 readings. The η_s, in contrast, helps to estimate the actual drying capacity of the low-cost dryer. In the experiments with the air humidifier it is possible to estimate with η_s the ability of the low-cost dryer of removing water from the sample flow. In the case of the experiments with hygroscopic salts, η_s estimates the ability of the low-cost dryer to avoid hygroscopic growth.”

L149 – R2 isn’t ‘coefficient of correlation’ – this also seems to be a minimal/insufficient way to compare these data sets under the ‘best possible’ conditions. What about slope/offset/bias?

We have changed it to coefficient of determination and added more statistics in Table 3, 4 and 5.

L159 – Given the size distribution measured, it is likely not sedimentation, but rather diffusive loss to chamber surfaces.

We have removed “the sedimentation curve started”.

L161 – The different size cut of the two OPCs could be a factor here. A minor growth could push particles into the detection window of the Palas. So this is likely not more particles, but growth of the same ones.

That is exactly what we meant by “increase of the water droplets which were too small to be detected at lower RH”. For better explanation, we have changed the sentence to the following:

“…possibly due to the growth of water droplets which were below the detection limit of the instruments at lower RH.”

L164 – As noted by other reviewer, this is likely because the dryer is going well beyond ‘reference’ conditions, and probably evaporating some of the salts/organics in these particles.

We have added the following text:

“This result was expected, as the Fidas® 200 under default settings does not aim to completely dry the sampled air but seeks to meet the requirements for FEM instruments as set in the EU directive 2008/50/EC. These requirements are met when the PM readings of the FEM instrument correspond to the values of the measured PM filters of the standard gravimetric analysis after being pre-conditioned at 19 to 21 °C and 45 to 50 % RH for at least 48 h (EN 12341).”

L168 – As noted above, these are not fog droplets, but probably particles of residuals from humidifier.

As we have explained before, the experiments were done with fog droplets as the possible residuals were filtered by the humidifier.

L189 – Previous work in this area has determined that there is not an ‘optimum’ and recommends the 40 C upper limit. This may be an important need, but as indicated by other reviewer, it needs to be made clear that this is clearly beneficial relative to computational approaches.

We have included the following sentence:

"One possible solution is introducing an adaptive heating to the dryer control to keep the RH of the air at the sensor inlet constant at 50 %. In such a case the temperature needed to maintain the RH of the air at 50 % could be adjusted so that higher temperatures than 40 °C would only be reached in during fog events, where the RH is close to 100% in order to be able to counter-react the effect of the fog in the PM readings. As can be seen in Fig. S8 in the supplemental material, the IADS also achieved temperatures higher than 40 °C during the real fog event."

L203 – As noted above, sedimentation is likely not an important loss process for particles in this size range (all sub-micron)

We have changed it to the following:

"…the decrease could have other causes, for instance, the sedimentation of the heavier particles or particle deposition onto the wall."

L235 – Not clear whether there would be any benefit to drying flow to an electrochemical (or other gas) sensor, as they typically respond to both temperature and RH, and possibly to absolute humidity. Therefore, a 'dryer' that doesn't actually remove water won't necessarily help (and may make things more complicated when it comes to signal interpretation)

A dryer for electrochemical sensors has already been tested in our laboratory and the results can be read in Samad et al. 2020. We have added the following text:

"Moreover, the design of the dryer can be easily adapted to other models or types of sensors, including, for instance, electrochemical sensors for gases as it has been tested in Samad et al. (2020)"

References

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