Response to interactive comments from Referee #1

Thank you for the time you put into reviewing our manuscript and the helpful feedback. Please see our following responses and proposed changes to the original manuscript, which we believe, help to improve this paper and increase its impact. Below the comments from Referee #1 are given in black. Our responses to the comments are shown in blue. Text added or changed in the manuscript is marked in italics.

This work evaluates the performance of a low-cost thermal dryer for the OPC-R1, in comparison to an OPC-R1 without a dryer and the Palas Fidas 200, which has an automated drying system. Two separate scenarios which can lead to erroneously high PM readings at elevated relative humidity (RH) were investigated in a laboratory setting. (1) Fog conditions, where water droplets suspended in the air are incorrectly detected as particles, and (2) hygroscopic growth of particles, leading to elevated size and mass measurement relative to a measurement at lower RH. To evaluate the performance of the R1 with dryer against the other two instruments, both the time series and “drying efficiency” are presented. The authors conclude that the low-cost dryer was able to mitigate positive artifacts due to the two effects outlined above, but raise concerns over the comparability of their “fog” conditions to the real-world scenario. They state the need for further work to characterise the temperature profile and optimise the power requirements of their drying system.

This work is a useful addition to the literature on low-cost particulate matter sensors. The literature review is thorough and highlights that drying systems are both under-studied and potentially simpler to implement than postcalibration of measurements for RH. The two scenarios being investigated are clearly distinguished and explained.

I would like to highlight two main aspects of the paper which I think should be clarified. I think this is good work; my concerns relate to how the results are presented. I think the results could be more clearly placed within the context of standard reference measurements of PM, and certain aspects of the method are unclear.

We thank the reviewer for the detailed and insightful review and for the positive statement about our work. In the revised version, we have included a new section comparing the OPC-R1 sensor with low-cost dryer with gravimetric measurements and a “reference-equivalent” instrument during field measurements. We believe this new section places our results within the context of standard reference measurements of PM.

It is not clear to me what the authors are trying to achieve through use of their low-cost dryer. Multiple comparisons are made which all suggest a different target. Are they trying to (1) completely remove all water, (2) produce “reference-equivalent” PM readings to supplement regulatory monitors, or (3) simply reduce the positive artifact due to the presence of water? I think this needs to be stated from the outset as it affects which evaluation method is most appropriate.

In order to clarify the aim of the investigation, we have added the following text:

“The aim of this study was to evaluate a prototype of a low-cost dryer built for a low-cost OPC under two different scenarios, namely fog events and hygroscopic growth, to reduce the influence of RH on the PM readings, i.e. to obtain “reference-equivalent” PM readings. For that purpose, experiments simulating both scenarios were performed under laboratory conditions and we quantified the effect of the dryer compared to a “reference-equivalent” monitor and to a low-cost OPC without a dryer. Additionally, two field campaigns were carried out with the aim of testing the prototype under real atmospheric conditions. In phase I, measurements with the gravimetric reference method, a continuous “reference-equivalent” monitor, and an OPC with dryer were performed in an urban background with daily averages of RH between 70 – 90 %. In phase II, measurements during a fog event with hourly averages of 100 % RH were carried out and the results of the OPC with dryer were compared to a continuous “reference-equivalent” monitor and to a sensor without dryer. Moreover, it was also evaluated whether the use of the low-cost dryer would allow a sensor calibration using exclusively a univariate linear regression (ULR) against gravimetric measurements, without the need of extra variables like RH.”

Comparing the R1 + dryer to the R1 without a dryer proves only aim (3), that water has been removed by heating. The reading is closer to dry particles, but we don’t know how close. Comparing the R1 to the Palas may prove (1) or (2). My uncertainty here is due to the lack of information on how the Palas PM measurements were processed. The Palas can output (1) raw particle number size distribution, (2) PM “classic” and (3) PM “ambient”. The authors could have obtained PM mass from (1) by applying their own density and penetration factors, (2) is calculated by the Palas using a default assumed density ρ = 1.3 g/cm^{-3}, whilst (3) has an additional empirical correction factor applied to make the readings "reference-equivalent" (Di Antonio 2020).

We have added more information about the method used to obtain the mass concentration from the reference instrument:

"*The mass concentrations were directly obtained from the instrument using the "PM-Ambient" algorithms provided by the manufacturer*."

Firstly, I would be unable to reproduce these findings without knowing which of the three methods the authors have used.

We have also added the following text in the conclusions for further clarification:

"The investigated dryer has been proved to be very effective on reducing the water content of hygroscopic particles or fog. The results also indicate that our prototype dries the particles more than a reference-grade instrument, which suggest that this design of low-cost dryer is over-dimensioned in terms of heating power to have "reference-equivalent" PM readings."

Secondly, if the PM "ambient" readings have been used, I think it is worth pointing out that (under default IADS settings) the Palas is not trying to fully dry the particles because it is aiming to be reference-equivalent. Hence this is relevant to aim (2). The EU reference standard specifies conditioning for 24-hr at 20°C, 50% RH (see CEN standard 12341), ie not completely dry. Given this, the Palas can be expected to potentially measure PM which still contains bound water and not remove all fog droplets as it may not sufficiently lower the RH.

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)."

Thirdly, when the authors manually set the IADS to 70°C, they were overriding these reference-equivalent settings and were likely removing nearly all the water. This comparison is most appropriate to aim (1).

The following sentence has been added:

"In order to determine an "apparent temperature" of the low-cost dryer, experiments in expert mode varying the temperature of the IADS were performed and the closest result is presented in Fig. 4b, in which the IADS was set using the expert mode at 70 °C. This "apparent temperature" is not the real temperature of the dryer as it was designed to keep a constant heat flux (through electric heating) and therefore the dryer has a temperature profile that varies through the length. More information about the air temperature inside the dryer has been summarized in section 3.1.3."

A short explanation of what each of the authors' comparisons (R1 without dryer, Palas automated settings, Palas at 70°C) actually means in the context of how much of the water is being removed and the relation to regulatory PM monitoring would make this paper more impactful.

The following sentence has been added:

"The comparison with the reference instrument running in automatic mode shows how close the OPC with dryer is at getting "reference-equivalent" PM readings whereas comparing it with an OPC without dryer helps to quantify the amount of water that the dryer can actually remove."

A more precise description of how the Palas PM values were obtained is essential to make this work replicable by other authors. There should also be a more detailed description of how the R1 PM values were derived- were they the firmware-output values and what density and refractive index have been used? I would also point out that some discrepancy between R1 and Palas PM readings is to be expected because the R1 has a larger minimum detectable diameter.

Apart from the new text which describes how the mass concentration readings are calculated by the Fidas® 200 (see above), a new table collecting all the necessary technical specifications of the Fidas® 200 and the OPC-R1 (including density and refractive index as stated by the reviewer) has been added.

Moreover, the following text has been added to give more information about the derivation of the PM values for OPC-R1:

"The mass concentrations were directly obtained from the PM outputs of the sensor."

My second point of feedback is that the meaning of the T_{OPC} quantity should be clarified. In particular, the authors should specify what this temperature corresponds to- is the sensor situated (1) on the OPC circuit board, or (2) within the sample air flow? The authors should then be clear about what they are preventing from overheating by including an upper temperature limit.

The following text has been added to specify where the temperature sensor is:

"*The temperature sensor is located in the OPC circuit board."*

In order to clarify what we mean with "to avoid overheating" we have added a new sentence,

"Previous experiments showed that the OPC-R1 switches automatically off if T_{OPC} reaches 44 °C so we selected an upper limit for T_{OPC} of 35 °C."

and rewritten the following sentence:

"If T_{OPC} is equal to or more than 35 °C the dryer switches off and starts cooling down to avoid overheating the sensor."

I suspect case (1) applies, from my own work with the OPC-N3, a similar model of OPC. This makes TOPC the temperature of the OPC box/components rather than the sample. Therefore the upper temperature limit of 35 °C would be preventing the OPC from overheating and is not an appropriate control measure to prevent excessive sample heating.

The reviewer is right. In order to state this problem in the manuscript we have added the following text:

"During the design phase, adding a temperature and a RH sensor at the end of the dryer was considered, but it was discarded as it would have affected the particle flow. The main advantage of using T_{OPC} is the fact that it forms part of the OPC-R1, i.e. the T_{OPC} data is part of the output of the sensor. However, the disadvantage is that using T_{OPC} does not prevent sample overheating."

As the authors state in the penultimate line, it is a shame that no information has been provided on the temperature (and RH) that the dryer conditions the sample to. This would be an important parameter in assessing which of the above three aims the dryer is best suited to. From the fact that the IADS had to be set to 70 °C to achieve comparable PM readings, I suspect that the sample is being heated much higher than the 35 °C limit on TOPC. This information would also be useful to assess potential loss of semi-volatile components.

We have added a new section 3.1.3 "Study on the drying temperature" containing the following text:

"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."

I have a few remaining minor points, which I shall outline more briefly below.

Figure 1 presents the averaged particle number and mass distributions during a fog event. I assume these have been averaged over some time period corresponding to the fog event. The authors should indicate (1) whether the data have been averaged and (2) over what time period.

In order to make it clearer, we have changed the x axis in Fig. 1a so that it only covers the range of time that has been used to calculate the average of the size distributions in Fig. 1b and 1c.

We have also added some clarifications in the text as follows:

"An example can be seen in Fig. 1a, where the one-minute average PM concentration…"

"The number of particles and the normalized mass concentration are averages over the time from 9:00 to 9:11h."

Some improvements could be made to the Methods section. Near the end of the paper, line 220 gives typical flow rate and power requirements for the self-developed and Palas drying systems. I think these would be better placed earlier, in the Methods section (say around line 125). Please also give the dimensions of the Palas' heated inlet for comparison. The authors should specify the orientation of the R1's inlet/dryer- is it pointed upwards as pictured in Figure 3A (similarly to the Palas inlet)? Within Methods, the authors say they used "sodium chloride, potassium chloride, ammonium sulphate, and ammonium nitrate" (line 111) with the aerosol generator, then go on to simply say how they used ammonium sulfate on lines 192/193. The information on lines 192/193 would be best moved to Methods, and I am not sure why these other salts have been included with no further discussion.

As stated above, we have included a new table with the technical specifications of the Fidas® 200 and the OPC-R1 in the Methods section. The dimensions of the Palas inlet has been included in the text:

"It is 1.2 m long and has an inner and an outer diameter of 12.7 and 48 mm, respectively."

We have now pointed out the orientation of the low-cost dryer:

"As it is shown in Fig. 2, the dryer is placed in a vertical position to minimize particle losses."

The information in lines 192/193 has been moved to the Methods section.

Regarding the other salts, it was not included because the same conclusion is reached with all of them, the low-cost dryer always dries more than the IADS in automatic mode. We have now added two more interesting experiments in the supplemental material: one with pure ammonium nitrate and one with a mixture of the salts, and it has been indicated in the manuscript with the following text:

"*Experiments were carried out with different aerosols ($(NH_4)_2SO_4$, NH_4NO_3, KCl and NaCl) and different IADS settings (automatic mode, IADS off (min. 20 °C), 35 °C, 50 °C, and 65 °C)."*

"For this experiment, the Fidas® 200 ran in automatic mode. Experiments with NH_4NO_3 and the mixture of the salts can be seen in Fig. S3 and S4 of the supplemental material, respectively. The DRH and ERH of $(NH_4)_2SO_4$ as well as the other tested salts are indicated in Table S2 in the supplemental material."

There are a few remaining points to clarify within the results sections. On line 158, the authors state the humidifier was turned off after reaching 300 µg m^{-3}. Given the differing RH sensitivity of the three instruments, the authors should state which of the three instruments was used to determine when this threshold had been reached.

The following clarification has been added:

"After the reference instrument reached a PM2.5 mass concentration of 300 µg m^{-3},…."

Wet towels were added before switching the humidifier off in experiment 1 (fig 4A) but after in experiment 2 (fig 4B). It is not clear to me why this difference is present. Additionally, the analysis from line 157 would read more clearly if the authors actually name (before further discussion) which of the two experiments is being described (IADS set to automatic or 70°C). The authors simply use "Fig. 4a", "Fig. 4b" to indicate which experiment they are referring to, but I think this could be clearer.

During the evaluation of the experiment with Fidas® 200 in automatic mode we observed that the RH was decreasing after switching off the humidifier. In order to avoid that decrease, we decided to introduce the wet towels before.

We have rewritten the following sentences to clarify which of the experiments we are referring to:

"…, *in Fig. 4a, the IADS of the reference instrument was kept in automatic mode, i.e. the default settings under which the instrument works during field measurements, whereas in Fig. 4b, it was set at 70 °C using the expert mode."*

"As shown in Fig. 4a during the experiment with the IADS in automatic mode,…"

Please give errors for the values of η_r and η_s. Additionally, for the experiment with IADS at 70°C, only η_s is given, please also specify η_r. The Palas settings have changed so it would be informative to compare the η_r values from each experiment.

We have added the errors of η_r and η_s calculated with the standard deviation. The second part of this comment is really good because it made us re-evaluate the meaning of η_r. As it was defined in Equation 1, it was difficult to understand the meaning (this is why it was not included in that experiment). We have now made a simple change, that is, instead of calculating the term "$1 - \frac{PM2.5_{d,i}}{PM2.5_{r,i}}$", we use only $\frac{PM2.5_{d,i}}{PM2.5_{r,i}}$, so the meaning is more comprehensible as it is explained now in the text as follows:

"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."

Do the authors know the typical temperature of the IADS during the automated setting experiment? It would be informative to compare this to the 70°C set temperature in the latter experiment.

Yes, we have this information and we have added three figures (one for each experiment) with the IADS temperatures and heating power in a new supplement (Fig. S1, S2, S5 and S9).

In line 196, the authors discuss the lack of a "sudden decrease" in the Palas PM2.5. I think it would be worth recognising that there is still a decrease, just slower than the R1 for the reasons discussed in the paper. The fact that the Palas PM2.5 is less than the R1 without any dryer shows that some sample drying must be occurring, as the pre-experiment calibration should have largely removed other sources of disagreement.

The reviewer is right. We have changed the sentence to:

This decrease is also observed with the reference instrument, but at lower pace.

In line 214, the author suggest the drops and jumps in PM2.5 measured by the OPC R1 with dryer may be due to loss of semi-volatile components during heating. However the sample is ammonium sulfate (non-volatile), so where have the semi-volatile components come from? If unfiltered air has been used with the aerosol generator, this should be specified, particularly as this could also limit the extent of water uptake by the ammonium sulfate.

Actually, the loss of semi-volatile components due to the excess of heating was meant for field measurements. Consequently, it is not related to the experiment. As the reviewer rightly stated, the ammonium sulfate particles used are non-volatile. We have moved the text into a new Discussion section so it is clear that it is not related to the experiments and the general thoughts and discussion start.

Finally, a few suggestions regarding the wording/readability in certain places.

• Line 98: "made from greenhouse glass"

We have changed this.

• Line 106: "consisting of a"

We have changed this.

• Line 124: “brass tube 50 cm in length”

We have changed this.

• Line 200: “much more slowly”

We have changed this.

• Line 203: “the decrease could have other causes”

We have changed this.

Overall, good work! I found it very interesting to read.

Thank you so much!

References

Di Antonio, A. (2020). Development of novel methodologies for utilising low-cost sensors for ambient Particulate Matter measurement (Doctoral thesis).

Thank you for this reference. We were not aware that Di Antonio also tested a thermal dryer for OPC-N2. We have included this work in the references.