Anonymous Referee #1

Received and published: 19 September 2017

This paper presents an evaluation of a low-cost sensor (OPC-N2) for monitoring ambient particulate matter. Three inter comparison field campaigns have allowed for determination of precision, comparison with reference instruments and suitability for long-term monitoring. This study gives new insights on the ability of these low-cost instruments to measure ambient particulate matter and notably, the identification and correction of bias related to high relative humidity conditions. The manuscript is clear, well-written and is suitable for publication after considering minor changes.

1. Figure 3 shows that calculated average coefficients of variance (CV) (line 17 page 9) are influenced by a few high values and are below 0.1 most of the time. This effect of a few high values on average CV should be considered in the discussion. Would it be possible to quantify the bias due to relative humidity?

Response:

The mean CV for the times when the RH was less than 85%, when we typically observed little influence from ambient RH on the measured particle mass concentration by the OPC-N2, was 0.3 ± 0.25 , 0.23 ± 0.14 and 0.2 ± 0.18 for PM₁, PM_{2.5} and PM₁₀ mass concentrations, respectively. This was only slightly lower than the overall average (at all RH: 0.32 ± 0.16 , 0.25 ± 0.14 and 0.22 ± 0.13 for PM₁, PM_{2.5} and PM₁₀ mass concentrations, respectively). However, while we observed higher CV when the RH was above 85% (0.34 ± 0.30 , 0.27 ± 0.14 and 0.23 ± 0.21 for PM₁, PM_{2.5} and PM₁₀ mass concentrations, respectively). However, while we observed higher CV when the RH was above 85% (0.34 ± 0.30 , 0.27 ± 0.14 and 0.23 ± 0.21 for PM₁, PM_{2.5} and PM₁₀ mass concentrations, respectively) and suggests that the individual OPC-N2 responded slightly differently to the effect of RH these were within the variability. Thus it suggests that ambient RH did not affect the precision of the OPC-N2 significantly.

We now state in the paper that the following on p17 L20 "Whilst the accuracy of the instrument was significantly worse at high RH the precision remains the same within error. The CV analysis conducted in section 3.1.2 is repeated for the same dataset but put into low (RH<85%) and high RH (RH>85%) subsets. For high RH conditions the CV for PM₁, PM_{2.5} and PM₁₀, was 0.34±0.30, 0.27±0.14 and 0.23±0.21, respectively. For low RH conditions the CV for PM₁, PM_{2.5} and PM₁₀, was 0.30±0.25, 0.23±0.14 and 0.20±0.18, respectively."

2. Determination of K value: please detail the calculation of K and its uncertainties. Humidograms on Figure 8 show that fitted models may possibly be not suitable. When fitted curves are used for prediction or for quantification, quality and suitability of fitted models need to be examined through an analysis of residues. Here I expect that the model is not suitable (overestimation at low RH and conversely). In this case this would support the assumption page 19 of the necessity to use two models for low and high relative humidities - that would improve the correction independently of aerosol composition.

Response:

We have repeated the analysis using two models as suggested by the reviewer, a linear correction for times when the ambient low RH was low (<85%) and for times at higher RH (>85%) a fitting based upon κ -Kohler theory (Eqn 6). We then compared the results of using this binary two model approach, to that originally applied, using Eqn 6 for all ambient RH. The results are shown below as scatterplots of the corrected OPC-N2 against the TEOM concentrations for PM_{2.5}. As can be seen Figure 1, there was little improvement in the slope or r^2 with the two model correction (Cv2) compared to the using correction with Eqn 6 for all RH (C). What was noticeable was that the intercept for the two model approach (Cv2) moved closer to zero, suggesting that at the lower mass concentrations the correction was improved. Similar trends were also observed for PM₁₀.



Figure 1: Scatterplots of corrected OPC-N2 against the TEOM for PM_{2.5} mass concentrations. The two model approach (Cv2) is in red and the one model approach in blue.

Therefore we have added the following text at page 20, line 15:

"There were also times when the OPC-N2 were clearly over-corrected (e.g. from 20th 10 February onwards), generally when the ambient RH was low (Fig 6). This suggests that when the RH was below a threshold, Eqn 6 overcorrects the data and this can be observed in the humidograms shown in Figure 8. Typically, at RH <85% the hygroscopic growth of real atmospheric aerosols is small and it may be more appropriate to apply a linear regression correction factor for data recorded under these RH conditions. Therefore we applied a binary two model approach to correct the OPC-N2 mass concentrations, where a linear correction (using the TEOM as reference concentration) for when RH <85%, and above this threshold in RH Eqn 6 was used. As can be seen Figure S9 (Supporting Information), there was little change in the slope or r^2 value with the two model correction compared to the using correction with Eqn 6 for all RH. What was noticeable was that the intercept for the two model approach moved closer to zero, suggesting that at the lower mass concentrations the correction was improved. Similar trends were also observed for PM₁₀."

3. Many figures are small and difficult to read and assess (1;5;6;7;9;S2;S4;S5). In particular for readability Figure 1 could be reduced to the second period (21st to 24th of September) and Figures 5 and S5 need to be re-scaled since most concentrations are flattened by a few very high values.

Response

We have fixed figures 1, 5, 7, 9, S2 and S4 as suggested. We have not rescaled Fig 6 and S5 as the point of this figure is to show that there are times when the OPC-N2 over-estimated the PM mass concentration over a very large scale, and rescaling the y-axis would lose this information.

4. Tables 1, 2 and 3: are they results of linear regressions (slopes) or ratios? In the first case indicate if intercepts are strained to zero or are non-significant.

Response:

All presented relationships are linear regressions (slopes). The intercepts were not constrained to zero and they vary from instrument to instrument. In Table 1, similar intercepts were observed for relationships between the OPC-N2 and TSI and GRIMM, and were around -1, -12 and -10 for PM₁, PM_{2.5} and PM₁₀, respectively. For Table 2, the intercepts against the TEOM were also significant, at -12 and -15 for PM_{2.5} and PM₁₀, respectively. The intercepts were found to be notably improved with the application of the RH correction (Table 3), and were around zero for the GRIMM (both size fractions) and about -3 for the TEOM.

We chose not to constrain the regression to zero to not bias the analysis, and the significant negative intercepts likely reflect the influence of a few high measurements by the OPC-N2 in Tables 1 and 2.

We have included this information in the headings for Tables 1-3, with the new heading for Table 1 shown as an example:

"Table 1: Slopes (linear regression) of measured PM mass concentrations of the reference instruments against the median and inter-quartiles for OPC-N2. The intercepts were not constrained to zero. Correlation co-efficient, r2 is given in parenthesis.

5. The statement lines 16-17 page 20 (while two of the OPC-N2 had a similar distribution to the GRIMM (OPC13 and 14), the other two OPC-N2 appeared to show evidence for instrument drift as the mode has shifted relative to the GRIMM) is not obvious from Figure 9.

Response:

We have adjusted this sentence to now read:

"The remaining four OPC-N2 were compared to GRIMM and in January after running for 4 months (Fig 8A), and while three of the OPC-N2 had a similar distribution to the GRIMM (OPC12, 13 and 14), OPC9 appeared to show evidence for instrument drift as the mode has shifted relative to the GRIMM."

We have also added the following sentence to the conclusions to highlight this apparent instrument drift

"One out of four OPC-N2 tested for long-term monitoring appeared to show evidence for instrument drift relative to reference instruments."

Anonymous Referee #2

Received and published: 4 October 2017

This manuscript describes the evaluation of a low cost optical particle sensor with respect to ambient PM monitoring. The advent of such low cost sensors is an important development in the PM monitoring field which will be important for future spatial distribution measurements and hence epidemiological health studies. The topic is well within the scope of AMT, and could be useful to community in understanding the advantages and limitations of such technology. However, the manuscript is not entirely well written, suffering at times from lack of clarity, and incomplete information. The issues are described further below. If these issues can be addressed then I believe this manuscript could be publishable in AMT and provide useful information.

1. Overall, the manuscript is too qualitative with respect to understanding how accurate and precise these sensors may be. On too many occasions the authors use the terminology "reasonable" to describe the agreement or precision etc.. Such terminology is far too subjective. What is considered "reasonable"? The authors should strive to be more quantitative in this respect, as many people will want to use such sensors and their recommendation may carry some weight within the community.

Response:

The term reasonable was used to make the paper more readable. We did throughout the paper apply quantitative analysis of the accuracy and precision of the OPC-N2 such as CV (Fig 3) and comparison to reference instrument (slopes by linear regression, e.g. Tables 1-3) and therefore feel we have provided this information. Low-cost sensors are by their nature a compromise between cost and quality and therefore we don't believe that the same criteria for research or monitoring grade instruments should necessarily apply when considering their performance.

The reviewer does make a valid point that we should define what we mean by reasonable and as a result we have made a number of changes the text in the following locations to address this:

Abstract, page 1 line 30:

"Inter-unit precision for the 14 OPC-N2 sensors of 22±13% for PM10 mass concentrations was observed"

page 3, line 10:

"Laboratory assessments of the performance of a number of low-cost miniature OPC's have shown promising results, with adequate precision observed when compared to reference instrumentation (Manikonda et al., 2016)."

Page 2, line 2:

"The level of precision demonstrated between multiple OPC-N2 suggests that they could be suitable device for applications where the spatial variability in particle concentration was to be determined, but need characterisation."

Page 6 line 24:

"which is not strictly true for airborne particles in an urban atmosphere but is considered a standard approximation."

Page 13, line 9:

"The time series of the median OPC-N2 $PM_{2.5}$ concentrations along with the two reference instruments are shown in Figure 5, and for a large portion of the inter-comparison all instruments appear to be in agreement."

Page 13, line 22:

"In Fig 6, the agreement between the OPC-N2 and the TSI instrument appears to vary as a function of ambient RH, with better agreement observed between the two instruments during periods of relatively low ambient RH."

Page 23, line 22

"Comparison of the OPC-N2 to the reference optical instruments demonstrated reasonable agreement for the measured mass concentrations of PM_1 , $PM_{2.5}$ and PM_{10} as evidenced by the stated accuracy and precision."

2. In my opinion, such technology has a long way to go before it can be a useful in determining the spatial distribution of PM and hence be used in health studies. One could argue that the accuracy is less important than the inter-instrument variability in this regard. However, a CV between sensors varying from 0.2 to 0.8 does not inspire confidence (ie. fig 3). The authors seem to think that such a CV is adequate, however if that is the case they must justify why they think that to be "reasonable". On pg 9, line 20 the author's state that the CV is "perhaps not unreasonable". This is entirely speculative, and depends upon the application. For most applications I doubt this is reasonable. The authors overall seem to be saying that this is a good sensor for deployment for spatial/health studies, when in reality the data they show indicate that is not really the case. I suggest this technology remains quite far from easily being used in such studies, especially because of the variability between instruments, the need for corrections on individual instruments, and the poor accuracy. These limitations need to be front and center in this manuscript to avoid confusion.

Response:

We note that Reviewer 1 agreed with our interpretation of Fig 3.

We don't agree that the OPC-N2 is far away from being useful in mapping spatial distribution of PM, but rather that the results presented in this paper show that this sensor can provide an accurate measure of PM concentration provided they are calibrated against reference instrument and the corrected for the RH artefact. All instruments need calibration if they are to provide useful data, and the OPC-N2 is no exception and we believe that the correction factors presented in this work enable the OPC-N2 to provide more accurate measurements. This is most explicitly evidenced in Figs S8-9, where we observed notable improvement in agreement, not just with reference instrument (in this case the TEOM) from 250-400% to 33%, but also between the four OPC-N2. We believe the proposed correction factor using k-theory is a significant advance in the use of these low cost OPCs, as we stated in Section 3.4, page 22, line 28:

"The use of κ -Kohler theory to derive a correction factor based on ambient RH improved the agreement between the OPC-N2 and reference instruments; however a limitation of this approach is that the bulk aerosol hygroscopicity is related to particle composition, typically the inorganic fraction (e.g. (Gysel et al., 2007)). Variation in ambient particle composition could account for the large spread observed in the ratio of OPC-N2/TEOM at high RH (Fig 7)"

The precision of these instruments was stated within the text (Section 3.1.2) where on average, the 14 OPC-N2 were found to have CV of 22±13% for PM10 mass concentrations without any corrections applied, with only the occasional spike in CV evident in Fig 3. In Fig 3, these are uncorrected results and the cause of the spikes in CV appears to be a result of the aforementioned RH

artefact (Please see our response to Reviewer 1, comment #1), as each OPC-N2 was found in Section 3.2 and 3.3 to respond differently to RH artefact.

On page 10, line 11 we used the phrase that this CV between the 14 OPC-N2 was "perhaps not unreasonable for low-cost sensor" as low-cost sensors are by nature a compromise between cost and quality. Much of the literature to date has focused on the accuracy of low-cost sensors and consequently there is little literature quantifying their precision. Sousan et al. (2016) reported for their laboratory measurements, the OPC_N2 had a CV of between 4.2-16%, which while lower than the current work, this would be expected for a controlled environment.

Recent work by Lewis et al., (2016) showed that 20 unspecified PM sensors sampling ambient air had an inter-quartile range of around 20 ug m-3, significantly higher than the current work. Wang et al. (2015) reported for three low-cost light scattering particle sensors, standard deviations of 15-90 μ g m⁻³ for repeated laboratory measurements of concentrations up 1000 μ g m⁻³. With limited comparable studies to assess the precision of the OPC-N2, we can only present our findings but suggest that the precision of the OPC-N2 is significantly improved compared to Lewis et al. for ambient measurements.

In addition, the precision and accuracy of the OPC-N2 was also found to improve with the application Eqn 6, as shown by Table 3, Figures 8, S8-9, compared to uncorrected concentrations.

The reviewer does make a valid point that we should avoid subjective terms (such as reasonable) and as such we have made several changes to the text to avoid this, please see our response to the previous comment.

Furthermore, we did state within in the text (page 10, line 13) "precision of the OPC-N2 would need to be considered when comparing multiple units", so we did highlight this issue in the text. However, we agree that we should make this point clear and so we have altered Section 4.0 (page 24, line 2) to highlight this issue:

"For PM_{10} mass concentrations, a CV of 22+13% between the 14 OPC-N2 employed in this study was observed, with some of the variability likely due to use of separate but identical inlets, and therefore could be considered reasonable for a low-cost sensor but this level of precision needs to be considered when using multiple units."

3. The comparison of the sensor with a TEOM needs to be justified more concretely. It is not clear how they can be comparing "apples-to-apples" with a TEOM which by their own admission uses a nafion dryer to dry particles first (while the OPC does not). The authors should explain exactly what the TEOM they are using is providing and how it an be compared to the OPC sensor. Are they truly comparing the same quantity? At first glance it does not seem like they are, but not enough information is provided to determine this. For that matter, why are they comparing with a TEOM at all, if they have just finished assessing the accuracy with a TSI/GRIMM. By doing so, they are adding another uncertain variable into the assessment which may not be needed.

Response:

We compared against a TEOM instrument as this an accepted regulatory standard instrument for particle mass measurements, and in particular was being run as part of the UK monitoring network . As we stated in the text (page 5 line 27), this is not a 'apples-to-apples' comparison, we were aware that the OPC-N2 and TEOM are fundamentally two different techniques and so there would be disagreement. But they are two approaches to the same measurement and we were interested to see how the calculated particle mass concentration by the OPC-N2 compared to a more direct measurement of particle mass concentrations by the TEOM. We note that many previous studies have compared optical particle counters to TEOM to see how the particle mass

measurements from optical particle counter instruments compare with the assumptions made to convert particle number concentrations to mass (see e.g. Wang et al 2016).

Therefore, we feel we are justified in comparing the measured particle mass by the OPC-N2 to a TEOM.

4. The description of the OPC sensor that is being investigated is highly lacking information. The authors need to improve their description of the sensor significantly. Although it may have been described in other work (which they have not even cited), it should be in part described here as well. Reading this short paragraph description I am left wondering: How does it sample? With a pump? Passively? How does the data collection work? What data is collected exactly? Does it only provide a mass concentration value? Does it provide number concentrations as well? What is the time resolution? What does the manufacturer say it should do? All these things and likely more need to be described in the methods section.

Response;

The OPC-N2 samples via a small fan, and can sample at min time resolution of 10s. The OPC-N2 is described in more detail in Sousan et al. (2016), and we have added reference to this paper. As we described in the second paragraph of Section 2.1.1, the OPC-N2 has been designed to log via Alphasense software on a laptop, and is also where we describes the custom built logging system we built.

The OPC-N2 collects number concentration and converts this to mass concentration via on-board factory calibration, as we describe in detail in Section 2.3.

Number concentration per size bin is available by size bin but we chose to focus on the mass concentration as this is the output that majority of users of an OPC-N2 are likely to use.

To include this additional information, the first paragraph of Section 2.1.1 (page 4, line 8) now reads:

"The Optical Particle Sensor (OPC) under evaluation in the current work is the OPC-N2 manufactured commercially by Alphasense (<u>www.alphasense.com</u>) and is described in detail in Sousan et al. (2016). The OPC-N2 can be considered as a miniaturized OPC as it measures 75x60x65 mm and weighs under 105 g, and as such is significantly cheaper (approx. £200) than the comparable reference instruments (see next section). The OPC-N2 samples via small fan aspirator and measures particle number concentration over a reported size range of 0.38 to 17 µm across 16 size bins, and maximum particle count of 10,000 per second. The minimum time resolution is 10s. The measured particle number concentration is converted via on-board factory calibration to particle mass concentrations for PM₁, PM_{2.5} and PM₁₀ size fraction according to European Standard EN481 (OPC-N2 manual). According the OPC-N2 manual, the standard definition for PM₁₀ in EN 481 extends beyond the particle size measured by the OPC-N2, and may consequently underestimate PM₁₀ value by up to 10%. Further discussion on calculations for conversion from particle number to mass concentrations is given in Section 2.3. All OPC-N2 in this study were firmware version 18."

5. If the GRIMM instrument is noted to always be 20% higher than the TSI, then which one is the standard? I am assuming that the TSI is the so-called "gold standard", as it is calibrated with a known stream of particles at some point or another. Is that the case? The authors make it sound as if they realize that the GRIMM is consistently incorrect. If so, then why are they using the GRIMM as a comparison at all? If they are trying to assess the accuracy of the OPC then they should determine which standard is truly accurate, and only compare to one of them. It does not make sense to me to be assessing accuracy with an instrument which is not providing the correct values. It seems the true measure of accuracy is using the TSI, so why not simply use that?

Response:

Both the GRIMM and TSI 3330 are accepted and widely used instruments for measuring particle number size distribution, and we are not claiming that the TSI is the gold standard. The GRIMM is an instrument that is has been designated a federal equivalent method (FEM) for measuring particle mass concentrations by the US EPA, and as such we do not consider it inferior to the TSI 3330.

That they did not agree is not entirely unexpected, as while there are usually excellent correlation, the slopes are rarely unity between different optical particle counters (See e.g. (Castellini et al., 2014; Dinoi et al., 2017).

Therefore as both TSI and GRIMM are widely used and airborne particle measurements are inherently instrument dependent, we chose to compare to both instruments in this study to see if there were any differences.

6. Since the reference instruments and the OPC are essentially coarse particle instruments, the inlet fabrication and geometry are critical in transmitting the largest particles into any of these instruments. Any slight bends and differing bends between instruments will highly impact the large particles that enter the instruments. How is this mitigated? Are they the same between standards and the OPSs? If not, then I don't see how any real analysis of accuracy can be made, since some large particles being lost preferentially can severely affect the PM10 mass. The authors could potentially calculate the losses as a function of size and inlet bends etc, using online calculators at the very least, to be sure they are at least consistent between instruments. This is less of a concern for the precision determination.

Response:

At EROS for the intensive inter-comparison all 14 OPC-N2 were fitted with a 12cm long stainless steel tubing that sampled horizontally at the same height (1.5m). The TSI 3330 and GRIMM also sampled at the same height. The GRIMM has a horizontal inlet that connects to black conductive tubing, which was of a similar length. The TSI has meanwhile has a vertical inlet and due to inlet constraints in a bend in the conductive tubing was necessary. Due to size of the inlets on the instruments, they were different diameters, 3/8" for OPC-N2 and ¼" for TSI and GRIMM. As a result of the above, we could not use the same length tubing or orientation for each instrument and while aware of this potential for different particle sampling efficiencies but were restricted by practicalities of the sampling location.

As suggested by the reviewer, we calculated the expected particle loss in a sample lines (using an on-line calculator, (Von der Weiden et al., 2009) for the TSI as it was the only one with bend in the inlet. With the sampling set up we used, we calculated a sampling efficiency of 92% for 10 μ m particles.

We have added this additional information to the text at Section 2.2.1 (page 5, line 23):

"Minimal lengths (12cm) of stainless steel tubing (OPC-N2) and conductive black tubing (TSI 3330 and GRIMM) were used to sample outside air, with each OPC having its own inlet at a height of 1.5 m. The vertical inlet for the TSI 3330 necessitated a bend in the tubing, however the calculated sampling efficiency (using von der Weiden et al., 2009) was 92% for particles with a diameter of 10 μ m. Therefore, while the inlet arrangement of the TSI 3330 may have affected the intercomparison, particularly when considering the accuracy of the OPC-N2, we were limited to what was practical."

The TEOM by design has a vertical inlet, and so we placed the OPC-N2 for this comparison as close to the TEOM inlet as possible on the roof, using the same length inlet as the intensive inter-comparison in September and so we believe should not overly affected the inter-comparison.

6. While I do not doubt that the OPC has an artefact associated with RH, I also notice in many of the figures that the inaccuracy seems to be worse at higher PM loading. Is it possible that the high RH may also be correlating with high mass? In that case which one is more important? Is it truly the RH or is it the mass that is causing the artefact? By their own admission, the authors note that there are other factors at play. Can these factors be determined? It would seem that rather than a correction based only on Kholer theory, additional corrections are needed. It might be possible to make a multivariate empirical correlation between the OPC/TSI ratio and the RH, mass, and/or others. Can this be done? A multivariate analysis may help to determine what factors are truly responsible for the discrepancy and to what degree.

Response:

We think that it is the RH that is causing the artefact not the particle mass and we feel that this was best evidenced by Figure 6. For a given range of RH, we did not observe a curve as would be expected if there was mass loading effect, rather a straight line. This strongly suggests that RH was the cause. The artefact at high RH was due to particle hygroscopicity, and so will also be affected by the particle composition. This was likely why there were times at high RH when the OPC are in better agreement with the reference instruments (See e.g. Fig 7).

While it is likely possible to make a correction factor based upon the RH and particle composition, as we discussed in Section 3.4 for this study we did not have access to on-line measurements of particle composition, so we cannot formulate this correction factor. This will be the focus of future work.

7. It remains unclear why RH should cause an artefact. I do not dispute that one exists, but the authors should attempt to explain why fundamentally the RH should make any difference to the OPC. In principle the OPC is determining if a particle scatters or not. If it does, then it is counted. So even if RH affects scattering (which it will), then I do not see how it will stop the scattering all together such that a particle is not counted. The authors need to provide a plausible hypothesis at least to explain this issue. What does the manufacturer say the specifications should be for the OPC sensor? It seems like no attempt was made to contact the manufacturer to get an idea of how the mass is calculated. Given they are assessing their instrument; one would think they would be agreeable to helping them out. How do these results compare with what the manufacturer says it should do in terms of accuracy and precision?

Response:

The effect of RH and particle hygroscopicity upon particle refractivity and size is well known. κ -Kohler theory allows the effect to be modelled. Hygroscopic particles take up water as a function of RH, with more water taken up at higher RH. Typically, this effect is particularly important for inorganic aerosols. We explain this at the start of Section 3.3 (Page 20, line 3):

"Clearly there were times when there was a significant instrument artefact for the OPC-N2 (Figs 4 and S4) and the highest over-estimations occurred at high RH at both EROS and Tyburn Rd (e.g. Fig 5 and 6). The size of hygroscopic particles is known to be dependent on RH, as the particle refractive index and size are both a function of RH. Inorganic aerosols (e.g. sodium chloride, nitrate and sulphate), make up a large portion of the PM10 observed at EROS (Yin et al., 2010), and are known to demonstrate an exponential increase in hygroscopic growth at high RH (e.g. (Hu et al., 2010; Pope et al., 2010)."

We also note that Section 3.4 (Page 25, line 16) is a discussion on the cause of the OPC-N2 interference, and in this section we directly attribute this artefact to particle water content, as we stated in at page 21, starting at line 4:

"In the previous sections, the significant positive artefact observed by the OPC-N2 relative to the reference instruments were at times when the ambient RH was high, pointing to particle water content as the cause. This result is perhaps not surprising, as many studies in the literature have shown that particle water content can be a major reason for discrepancies between techniques that measure ambient particle mass (See e.g. (Charron et al., 2004)). The use of κ -Kohler theory to derive a correction factor based on ambient RH improved the agreement between the OPC-N2 and reference instruments"

Therefore, this artefact due to RH is not whether or not a particle is counted, rather the size bin that the particle is assigned to. Thus, as the OPC-N2 on-board calculation applies a single particle density for all size bins to convert the particle number concentration to particle mass, assigning a particle to wrong size bin will result in an over-estimation of the particle mass concentration.

We did contact Alphasense for more information on how the particle mass was calculated but they were unwilling to share that information with us, which was also the experience of Sousan et al. (2016). The manual of the OPC-N2 does not give any information with regards to accuracy and precision of the calculated particle mass concentrations, only for the number size distributions. This was part of the reason for focusing on particle mass concentrations.

8. There are many studies where mobile measurements of PM were made in urban and suburban areas. By looking at the spatial variation of the PM in those studies, one can get an idea of what kind of inter-instrument variability is required for this to be a useful instrument. Some attempt at this should be done, at least qualitatively.

Response:

The spatial variability of PM_{10} mass concentrations in urban areas is hugely variable, ranging from limited (e.g. 20-24 µg m⁻³ (Harrison et al., 1999), to more substantial such as 24-40 µg m⁻³ (Boogaard et al., 2010), 67-142 µg m⁻³(Chan et al., 2001), and likely reflects the spatial heterogeneity of the major sources (e.g. traffic). Similar trends are also found for $PM_{2.5}$ with one study finding the concentration ranged from 6.7-48.3 µg m⁻³ across a city (Martuzevicius et al., 2004).

Considering the CV reported for PM_{10} mass concentrations by 14 OPC-N2 (22±13%), then we would expect these instruments to be suitable precision for the many urban areas where there is notable spatial variation.

Minor issues:

Pg 2, line 2: the term "reasonable" is used here and not justified.

Please see our response to comment #1.

Pg 2, line 30: this line is awkwardly written. Remove the "are" and use "companies" or "manufacturers" but not both.

Response:

Changed to:

"There are a wide range of low-cost particle sensors available commercially from manufacturers including Dylos, TSI, Airsense and Alphasense."

Pg 3 , line 20: define "PUWP" and "dylos" Pg 3, line 19: add "the" before "dylos" (if I am reading this correctly)

Response:

Dylos is the name of the instrument, so does not need defining. The PUWP is an acronym and so the definition has now been included. The sentence now reads:

"Previous field testing of low-cost particle sensors has found that the Dylos (Steinle et al., 2015) and (Gao et al., 2015) performed well for ambient sampling of particle mass concentration in both an urban and rural environments when compared to reference instruments, however they were assessed were over a short period (4-5 days)."

Pg 3, line 21: remove the "s" from "environments" Pg 3, line 22: add "they" after "however" Pg 3, line 29: "sites" to "site" Pg 4, line 11: replace "were" with "used"

Response:

All of the above have been fixed

Pg 4, line 15-17: awkwardly written. Please improve. And remove "s" from "systems"

Response: Changed to:

"Therefore, we developed a custom built system for logging the OPC-N2 during the intercomparison, utilizing either a Raspberry Pi 3 or Arduino system."

Pg 5, line 17-18: it is not clear what this is supposed to be used for in this paper.

Response:

We collected RH data from the nearby met station. This has been added to the text:

"In addition, RH measurements from the nearby Elms Road Meteorological station were also obtained, which is located approximately 100 m away from EROS."

Pg 5, line 29: briefly describe what the point of the "filter dynamic system" is.

Response:

The following text has been added to explain the use of the FDMS

"the TEOM monitor was fitted with a Filter Dynamic Measurement System (FDMS) (Grover et al., 2006), to correct semi-volatile particle loss."

Pg 6, line 6: add an "s" to "OPC" Pg 8, line 15: awkwardly written. Please improve.

Response: Changed to:

"This demonstrates that the highest and lowest reporting OPC was not consistently reporting the highest and lowest PM2.5 concentration, respectively over the whole 3 day period."

Pg 9, line 20: far too speculative without backing it up.

Response:

Please see our response to comment #1

Pg 10, line 5: define what "consistent" means to you. Fig 3 indicates it is not at all consistent:

Response:

We have changed the text to include the mean and standard deviation as below:

"Throughout the measurement period, the CV was fairly consistent (mean of 0.22±0.13), with spikes in CV values evident during periods of high PM2.5 concentrations, in agreement with trends observed in Fig 1."

Pg 10, line 7: again, "reasonable" is too subjective. Pg 10, line 23: again, the use of "reasonable": : :..what does this mean?

Response:

Please see our response to comment #1

Pg 11, line 5: it should not agree with the GRIMM as you have already stated it is 20% off to begin with.

Response:

The reviewer makes a valid point and we have changed the text to read:

"While the TSI and GRIMM have the same particle size cut-off (0.3 μ m), these instruments have been shown to disagree (Fig S1) possibly due to different particle collection efficiencies."

Pg 15: how is the volatile fraction determined? (briefly). What does "gravimetrically corrected" mean in this context?

Response:

The volatile fraction is determined by the FDMS system on the TEOM, and represents the mass of semi-volatile particles. We have added an explanation to the caption on Fig 7:

"Figure 7: Time series for hourly measured PM mass concentrations by the TEOM, four OPC-N2 and GRIMM at Tyburn Rd urban background AURN station. The volatile particle mass concentration as measured by the TEOM-FDMS and relative humidity measured at Tyburn Rd also shown."

The term gravimetrically corrected means that the optical instruments have been corrected by comparison to gravimetric determination of particle mass.

Table 2: units of slope? Or unitless?

Response:

The slopes are unit less as we have plotted measurements of the same units.

Pg 17, line 1: is this the median of all OPCs or all them individually?

Response:

Each OPC-N2 at Tyburn Rd was plotted as function of RH and showed the same trends.

Pg 21, lines 7-8: this has no bearing on the current study.

Response:

We disagree, this statement is entirely relevant to the current work as we have found that RH was a major artefact on the measured particle mass concentrations by the OPC-N2. This statement shows that this artefact due to particle water content is not just specific to the OPC-N2 but generally an issue across instruments that measure particle mass concentrations.

Pg 22, line 15: what is "knock on"??

Response: We have removed this term.

Pg 22, line 20: remove "while"

Fixed

Pg 22, line 23: "suitable" is not what the reader gets from this paper. See my comments above.

Response:

We disagree, as we have stated in our response to previous comments (#2) and will keep this sentence the same

Figure 1: difficult to see as there are too many lines. Perhaps shorten the time scale and zoom in. Perhaps a log scale would help too.

Figure 5: too small to see anything other than the peak. Perhaps use a log scale to better see what is going on.

Figure 6: Too small to see anything. I suggest you split the y-axis and zoom in to where the majority of data is.

Response:

Figures 1 and 5 have been fixed as suggested.

Figure 6: We have not split the y axis as suggested as we want to show all the data, the point of this figure is to show times when the OPC-N2 over-estimated the PM concentration, and splitting the y-axis would lose this information.

Response to interactive comment from W.R. Stanley

1. Albeit briefly, European Standard EN481 is mentioned in the OPC-N2 user manual when describing how PM is calculated from the particle number concentration data.

Response:

We have added that particle mass concentrations are calculated by OPC-N2 according to EN481 to the Section 2.1.1, please see response to Reviewer 2, comment #4.

2. The author could be more specific about the inlet arrangements with their use of the OPC-N2. In addition to comments made in this subject by referee RC2, with its small fan aspirator, the air-flow through the device may easily affected by changes to its default inlet or the nature of the ambient air e.g. breeze across the inlet. Possible differences in response between these and the reference instruments due to such factors should be discussed.

Response:

Please see our response to Reviewer 2, comment #6 on this issue. We have added discussion that the inlet arrangement may have affected the inter-comparison.

References:

Boogaard, H., Montagne, D.R., Brandenburg, A.P., Meliefste, K., Hoek, G., 2010. Comparison of short-term exposure to particle number, PM10 and soot concentrations on three (sub) urban locations. Science of The Total Environment 408, 4403-4411.

Castellini, S., Moroni, B., Cappelletti, D., 2014. PMetro: Measurement of urban aerosols on a mobile platform. Measurement 49, 99-106.

Chan, L.Y., Kwok, W.S., Lee, S.C., Chan, C.Y., 2001. Spatial variation of mass concentration of roadside suspended particulate matter in metropolitan Hong Kong. Atmospheric Environment 35, 3167-3176. Charron, A., Harrison, R.M., Moorcroft, S., Booker, J., 2004. Quantitative interpretation of divergence between PM10 and PM2.5 mass measurement by TEOM and gravimetric (Partisol) instruments. Atmospheric Environment 38, 415-423.

Dinoi, A., Donateo, A., Belosi, F., Conte, M., Contini, D., 2017. Comparison of atmospheric particle concentration measurements using different optical detectors: Potentiality and limits for air quality applications. Measurement 106, 274-282.

Gysel, M., Crosier, J., Topping, D.O., Whitehead, J.D., Bower, K.N., Cubison, M.J., Williams, P.I., Flynn, M.J., McFiggans, G.B., Coe, H., 2007. Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2. Atmos. Chem. Phys. 7, 6131-6144.

Harrison, R.M., Jones, M., Collins, G., 1999. Measurements of the physical properties of particles in the urban atmosphere. Atmospheric Environment 33, 309-321.

Lewis, A.C., Lee, J.D., Edwards, P.M., Shaw, M.D., Evans, M.J., Moller, S.J., Smith, K.R., Buckley, J.W., Ellis, M., Gillot, S.R., White, A., 2016. Evaluating the performance of low cost chemical sensors for air pollution research. Faraday Discussions 189, 85-103.

Manikonda, A., Zíková, N., Hopke, P.K., Ferro, A.R., 2016. Laboratory assessment of low-cost PM monitors. Journal of Aerosol Science 102, 29-40.

Martuzevicius, D., Grinshpun, S.A., Reponen, T., Górny, R.L., Shukla, R., Lockey, J., Hu, S., McDonald, R., Biswas, P., Kliucininkas, L., LeMasters, G., 2004. Spatial and temporal variations of PM2.5 concentration and composition throughout an urban area with high freeway density—the Greater Cincinnati study. Atmospheric Environment 38, 1091-1105.

Sousan, S., Koehler, K., Hallett, L., Peters, T.M., 2016. Evaluation of the Alphasense optical particle counter (OPC-N2) and the Grimm portable aerosol spectrometer (PAS-1.108). Aerosol Science and Technology 50, 1352-1365.

Von der Weiden, S., Drewnick, F., Borrmann, S., 2009. Particle Loss Calculator–a new software tool for the assessment of the performance of aerosol inlet systems. Atmos. Meas. Tech 2, 479-494. Wang, Y., Li, J., Jing, H., Zhang, Q., Jiang, J., Biswas, P., 2015. Laboratory Evaluation and Calibration of Three Low-Cost Particle Sensors for Particulate Matter Measurement. Aerosol Science and Technology 49, 1063-1077.

Wang, Z., Calderón, L., Patton, A.P., Sorensen Allacci, M., Senick, J., Wener, R., Andrews, C.J. and Mainelis, G., 2016. Comparison of real-time instruments and gravimetric method when measuring particulate matter in a residential building. *Journal of the Air & Waste Management Association*, *66*, pp.1109-1120.

Evaluation of a low-cost optical particle counter (Alphasense OPC-N2) for ambient air monitoring

- 3
- Leigh R. Crilley¹, Marvin Shaw², Ryan Pound², Louisa J. Kramer¹, Robin Price³, Stuart
 Young², Alastair C Lewis², Francis D. Pope^{1*}
- 6

¹School of Geography, Earth and Environmental Sciences, University of Birmingham,
8 Birmingham, United Kingdom, B15 2TT.

- 9 ²National Centre for Atmospheric Science, Wolfson Atmospheric Chemistry Laboratories,
- 10 University of York, York, United Kingdom, YO10 5DD.
- ³Birmingham Open Media (BOM), 1 Dudley Street, Birmingham, B5 4EG.
- 12 *Corresponding author <u>f.pope@bham.ac.uk</u>

13 Abstract

14 A fast growing area of research is the development of low-cost sensors for measuring air 15 pollutants. The affordability and size of low-cost particle sensors makes them an attractive 16 option for use in experiments requiring a number of instruments such as high density spatial 17 mapping. However, for these low-cost sensors to be useful for these types of studies their 18 accuracy and precision needs to be quantified. We evaluated the Alphasense OPC-N2, a 19 promising low-cost miniature optical particle counter, for monitoring ambient airborne 20 particles at typical urban background sites in the UK. The precision of the OPC-N2 was assessed by co-locating 14 instruments at a site to investigate the variation in measured 21 22 concentrations. Comparison to two different reference optical particle counters as well as a 23 TEOM-FDMS enabled the accuracy of the OPC-N2 to be evaluated. Comparison of the OPC-24 N2 to the reference optical instruments demonstrated reasonable agreement for the measured 25 mass concentrations of PM1, PM2.5 and PM10. However, the OPC-N2 demonstrated a 26 significant positive artefact in measured particle mass during times of high ambient RH 27 (>85%) and a calibration factor was developed based upon κ -Kohler theory, using average 28 bulk particle aerosol hygroscopicity. Application of this RH correction factor resulted in the 29 OPC-N2 measurements being within 33% of the TEOM-FDMS, comparable to the agreement 30 between a reference optical particle counter and the TEOM-FDMS (20%). Inter-unit 31 precision for the 14 OPC-N2 sensors of $22\pm13\%$ for PM₁₀ mass concentrations was observed.

Overall, the OPC-N2 was found to accurately measure ambient airborne particle mass concentration provided they are i) correctly calibrated and ii) corrected for ambient RH. The level of precision demonstrated between multiple OPC-N2 suggests that they would be suitable device for applications where the spatial variability in particle concentration was to be determined.

6

7 1.0 Introduction

8 Airborne particles are of global concern due to their detrimental health effects, particularly in 9 the fine fraction ($PM_{2.5}$, particles with an aerodynamic diameter less than 2.5 µm) and as a 10 result are a regulated pollutant in the EU, USA and other states. Monitoring ambient particle 11 mass concentrations is typically performed using a small number of fixed instruments with 12 gaps in the spatial coverage usually estimated via modeling or interpolation. This is often 13 unsatisfactory as there can be micro-environments in urban areas that result in large spatial 14 and temporal inhomogeneity in airborne particle concentrations, which in turn makes 15 assessment of human exposure to airborne particles difficult (de Nazelle et al., 2017).

16 Into this gap a fast growing area is the development of low-cost sensors for measuring the 17 concentrations of a wide range of species in the atmosphere including gases and particles (Lewis et al., 2016; Rai et al., 2017; Snyder et al., 2013). However the question remains as to 18 19 whether the uncertain quality of data from these low cost sensors can be of value when 20 attempting to determine pollutant concentrations at high spatial resolution (Kumar et al., 21 2015). Sensors for both gases and particles can suffer from drift and a number of interference 22 artefacts such as relative humidity (RH), temperature and other gas phase species (Lewis et 23 al., 2016; Mueller et al., 2017; Popoola et al., 2016). Despite these challenges, recent work 24 has shown that low-cost gas sensors can be deployed in large scale networks provided 25 appropriate corrections for known artefacts are applied (Borrego et al., 2016; Mead et al., 26 2013; Mueller et al., 2017), with clustering of multiple gas sensors into one unit shown to be 27 an effective methodology (Lewis et al., 2016; Mueller et al., 2017; Smith et al., 2017).

For low-cost particle sensors, their reported performance across the literature is somewhat mixed (Borrego et al., 2016; Castellini et al., 2014; Sousan et al., 2016; Viana et al., 2015) and can depend on the type of particle sensor employed. There are a wide range of low-cost particle sensors are available commercially from manufacturers including Dylos, TSI, 1 Airsense and Alphasense. The more widely used and available low-cost particle sensors can 2 be considered as miniaturized versions of optical particle counters (OPC) and employ a light 3 scattering technique to measure ambient particle concentrations (See e.g. (Gao et al., 2015; 4 Sousan et al., 2016). While these miniature OPC are not meant to compete with more 5 established instrumentation in terms of their accuracy and precision, their affordability and 6 size makes them attractive for use in experiments requiring a number of such instruments, 7 such as personal monitoring (See e.g. (de Nazelle et al., 2017; Steinle et al., 2015)). However 8 to be useful in these types of studies, the precision and accuracy of these instruments needs to 9 be evaluated.

10 Laboratory assessments of the performance of a number of low-cost miniature OPC's have 11 shown promising results, with adequate precision observed compared to reference 12 instrumentation (Manikonda et al., 2016). Sousan et al., (2016) evaluated the Alphasense 13 OPC-N2 in a laboratory study using reference aerosols (Arizona road dust, NaCl and welding 14 fumes) and found reasonable agreement for size distributions and particle mass between the 15 OPC-N2 and a GRIMM Portable Aerosol Spectrophotometer, provided appropriate and 16 specific calibrations were applied. While these results are encouraging (Manikonda et al., 17 2016; Sousan et al., 2016), laboratory-based studies using reference aerosols may not be 18 representative of their performance when measuring ambient particles, owing in part to the 19 complex mixture and variable relative humidity and temperature encountered in the real-20 world. Previous field testing of low-cost particle sensors has found that the Dylos (Steinle et 21 al., 2015), Portable University of Washington Particle (PUWP) monitors (Gao et al., 2015) 22 performed well for ambient sampling of particle mass concentration in both an urban and 23 rural environment when compared to reference instruments however they were assessed over 24 a short period (4-5 days). In contrast, at a roadside location poor agreement between two 25 different OPC sensors compared to reference instruments was observed by Borrego et al. 26 (2016). Clearly, the results are mixed and longer-term assessment of the stability and 27 longevity of these instruments are needed, as these are critical parameters when considering 28 their worth for use in large-scale networks.

We evaluate here the Alphasense OPC-N2, a promising low-cost miniature optical particle counter (Sousan et al., 2016), for monitoring ambient airborne particles at typical urban background sites in the UK. We assessed the inter-unit precision of the OPC-N2 by colocating 14 instruments at a single site to investigate the variation in measured particle mass 1 concentration in the PM_{10} , $PM_{2.5}$ and PM_1 size fractions between OPC-N2. In order to 2 determine the accuracy of the OPC-N2, we compared it to two well-established commercial 3 optical particle counters that employ a similar light scattering technique as well as a TEOM-4 FDMS, a regulatory standard instrument for particle mass concentration measurements.

5 **2.0 Method**

6 2.1 Instrumentation

7 2.1.1 Alphasense Optical particle sensor (OPC-N2)

8 The Optical Particle Sensor (OPC) under evaluation in the current work is the OPC-N2 9 manufactured commercially by Alphasense (www.alphasense.com) and is described in detail 10 in Sousan et al. (2016). The OPC-N2 can be considered as a miniaturized OPC as it measures 75x60x65 mm and weighs under 105 g, and as such is significantly cheaper (approx. £200) 11 12 than the comparable reference instruments (see next section). The OPC-N2 samples via small 13 fan aspirator and measures particle number concentration over a reported size range of 0.38 to 17 µm across 16 size bins, and maximum particle count of 10,000 per second. The 14 15 minimum time resolution is 10s. The measured particle number concentration is converted 16 via on-board factory calibration to particle mass concentrations for PM_1 , $PM_{2.5}$ and PM_{10} size 17 fraction according to European Standard EN481 (OPC-N2 manual). According the OPC-N2 18 manual, the standard definition for PM_{10} in EN 481 extends beyond the particle size 19 measured by the OPC-N2, and may consequently underestimate PM10 value by up to 10%. 20 Further discussion on calculations for conversion from particle number to mass 21 concentrations is given in Section 2.3. All OPC-N2 in this study used firmware version 18.

The OPC-N2 is designed to log data via a laptop using software supplied by Alphasense, however this may not be practical when using multiple OPC-N2 at once or for personal monitoring. Therefore, we developed a custom built system for logging the OPC-N2 during the inter-comparison, utilizing either a Raspberry Pi 3 or Arduino system. The Python code to log the outputs from OPC-N2 on a Raspberry Pi 3 is made available in the Supplementary Material. The Python code makes use of the py-opc python library for operating the OPC-N2 written by Hagan (2017).

1 2.1.2 Reference Instruments

2 The first reference instrument was a TSI 3330 optical particle spectrophotometer (OPS), 3 which measures particles number concentrations between 0.3 - 10 µm across 16 size bins. 4 with a maximum particle count of 3000 particles cm⁻³. A GRIMM Portable Aerosol 5 Spectrometer (PAS-1.108, forthwith referred to as the GRIMM) was also utilized, which records particle number concentrations in 15 bins from $0.3 - 20 \ \mu\text{m}$. The TSI 3330 and 6 7 GRIMM were both recently calibrated and serviced. All measurements of airborne particle 8 concentrations are inherently operationally defined and as a result the TSI 3330 and the 9 GRIMM were chosen as reference instruments as they measure particle size in similar size 10 bins by a similar photometric technique to the Alphasense OPC-N2.

For the sake of this inter-comparison, we have taken the TSI 3330 and GRIMM data as an accurate measure of particle mass concentrations. The reference instrument used for the factory calibration of the OPC-N2 by Alphasense is the TSI 3330 (Sousan et al., 2016) and hence included for comparison.

15 **2.2 Inter-comparison locations**

16 2.2.1 Elms Rd Observatory Station

17 The instruments were housed within the Elms Road Observatory Station (EROS) located on the University of Birmingham campus. The site is classed as urban background, with 18 19 emissions from nearby road and a construction site the major sources of particles. Fourteen 20 OPC-N2 were deployed at EROS, enabling the precision of the OPC-N2 to be assessed along 21 with the accuracy relative to the reference instruments, the TSI 3330 and GRIMM. An intensive inter-comparison ran for just over 5 weeks, from 26th August till 3rd October 2016, 22 during which all 14 OPC-N2, TSI 3330 and GRIMM sampled ambient air. Minimal lengths 23 24 (12cm) of stainless steel tubing (OPC-N2) and conductive black tubing (TSI 3330 and GRIMM) were used to sample outside air, with each OPC having its own inlet at a height of 25 1.5 m. The vertical inlet for the TSI 3330 necessitated a bend in the tubing, however the 26 27 calculated sampling efficiency (using von der Weiden et al., 2009) was 92% for particles with a diameter of 10 µm. Therefore, while the inlet arrangement of the TSI 3330 may have 28 29 affected the inter-comparison, particularly when considering the accuracy of the OPC-N2, 30 we were limited to what was practical. Sampling intervals for the OPC-N2, TSI 3330 and GRIMM were 10, 60 and 6 seconds, respectively. In addition, RH measurements from the
 nearby Elms Road Meteorological station were also obtained which is located approximately
 100 m away from EROS.

At the conclusion of the intensive inter-comparison, a subset of the OPC-N2 (5) continued to
sample at EROS along with the GRIMM, to test the robustness and suitability of the OPC-N2
for longer-term monitoring. The long-term monitoring concluded on 1 February 2017,
meaning that these OPC-N2s sampled ambient air for up to 5 months.

8 **2.2.2 Tyburn Rd**

9 For regulatory purposes, an accepted method for measuring particle mass concentrations is a 10 Tapered Element Oscillating Microbalance (TEOM) and therefore we also compared the 11 OPC-N2 to this technique despite the difference in particle measurement approaches. An 12 urban background air monitoring station part of the UK Automatic and Rural Urban Network 13 (AURN) nearby EROS (Tyburn Rd) was chosen for this inter-comparison. At the Tyburn Rd 14 AURN station, the TEOM monitor was fitted with a Filter Dynamic Measurement System 15 (FDMS) (Grover et al., 2006), to correct for semi-volatile particle loss. A subset of OPC-N2 16 (4) and the GRIMM PAS 1.108 that were deployed at EROS sampled at Tyburn Rd station 17 for 2 weeks during February 2017. The OPC-N2 was housed individually within waterproof boxes on the roof of the cabin near to the TEOM inlet in order to keep the inlet length the 18 19 same as used at EROS. The GRIMM sampled from a nearby separate inlet.

20 2.3 Data Analysis

21 All OPC employed in this study count the number of particles and determine the size based 22 upon particle light scattering of a laser, and to convert to particle mass concentration must 23 apply a number of assumptions. To calculate the particle mass concentration, spherical 24 particles of a uniform density and shape are assumed, which is not strictly true for airborne 25 particles in an urban atmosphere but is considered a standard approximation. Therefore to 26 ensure a fair comparison between the different OPC, the same calculations and assumptions 27 must be applied to all three OPC measurements. The TSI 3330 data was processed using the TSI AIM software to convert the particle count concentration to particle mass measurements. 28 29 The particle counts from the GRIMM data was converted to particle mass (via particle 30 volume) using the same calculations, as outlined in the TSI AIM software manual according 31 to Equations 1 to 3:

$$1 \qquad D_{pv} = LB \left[\frac{1}{4} \left(1 + \left(\frac{UB}{LB} \right)^2 \right) \left(1 + \left(\frac{UB}{LB} \right) \right) \right]^{\frac{1}{3}} \tag{1}$$

$$2 \qquad v = \frac{\pi D_{pv}^3 n}{6} \tag{2}$$

$$3 \quad m = \rho v \tag{3}$$

4 where D_{pv} is the volume weighted diameter, LB the channel lower boundary, UB the channel 5 upper boundary, v is the particle volume for a channel, n is number weighted concentration 6 per channel, m is the particle mass per channel and ρ is the particle density.

The OPC-N2 converts, on board via a factory determined calibration, particle counts to particle mass concentration in PM_{1} , $PM_{2.5}$ and PM_{10} mass concentrations. There is no further information provided by Alphasense on how this calculation is performed apart from the applied particle density across all size bins was 1.65 g cm⁻³. Therefore, we assumed calculations are similar to Eqns 1 and 2 as applied to the TSI and GRIMM data and used the same particle density (1.65) across all size bins to calculate particle mass for all OPC.

All instrument time series were corrected for drift against a reference time. As the sampling intervals varied slightly between the different OPC, a 5 min average of particle concentrations was used for inter-comparison between instruments.

16 **3.0 Results and Discussion**

17 **3.1 EROS inter-comparison**

18 **3.1.1** Comparison of reference optical light scattering instruments

The two light scattering optical particle counters used as reference instruments in this study were found to be well correlated ($r^2 > 0.9$), with the GRIMM recording between 20-30% higher concentrations for all three particle mass fractions (Fig S1, Supporting Information). The GRIMM is known to overestimate number concentration (Sousan et al., 2016 and references therein) and this difference may reflect differing efficiencies in particle detection between the two instruments.

1 **3.1.2 Performance of the OPC-N2**

2 The performance of the custom built logging systems varied between 44-94% successful data 3 capture, with the Arduino and Raspberry Pi systems giving 44-65% and >92%, respectively. 4 The Raspberry Pi data logger system was used for the long-term measurements and for the inter-comparison with the AURN site due to its better performance. The data losses were due 5 6 to hardware issues and not related to performance of the OPC-N2. Due to the missing data, 7 only a subset of measured PM_{2.5} concentrations when all 14 OPC-N2 were logging are shown 8 in Fig 1, along with measured concentrations by the reference instruments. From Fig 1, while 9 there are times when there appears to be excellent agreement between the OPC-N2 and the 10 reference instruments, there are times when the OPC-N2 record a significant positive artefact, 11 and during these times the spread in measured concentrations increases. For example, on the 12 morning of the 18th September, the range of measured concentrations by the individual OPC-N2 was from approximately 30-150 μ g m⁻³, whereas the reference instruments reported ~10 13 μ g m⁻³. The cause of the positive artefact is investigated in later sections, but it points to the 14 individual OPC-N2 responding differently to this artefact. Similar trends were also observed 15 16 for PM_1 and PM_{10} , see Figure S2 in the Supporting Information.

17





19

Figure 1: Time series of $PM_{2.5}$ concentrations measured by all OPC-N2 and the reference instruments, TSI 3330 and GRIMM for selected period with high OPC-N2 data coverage.

22

As there is a considerable spread in response for the OPC-N2 relative to the reference instruments, we then quantified whether it was always the same OPC-N2 reading low and

1 high. Due to the aforementioned data capture issues, this analysis was only applied to days when all 14 OPC-N2 were running, 21st-24th September (Fig 1). The results are shown as a 2 3 rank order plot, where the OPC-N2 observations are ordered from the highest reported value 4 to the lowest over this period, normalised to the median concentration at the start of the 5 analysis (t=0), shown for PM_{2.5} mass concentration in Figure 2. The ranking of the OPC-N2's 6 showed some variability over time within periods of 1-6 hours, which was particularly 7 noticeable during periods when the OPC-N2 signals underwent large changes in 8 concentrations. This demonstrates that the highest and lowest reporting OPC was not 9 consistently reporting the highest and lowest the lowest PM_{2.5} concentrations, respectively over the whole 3 day period. The same trend was also observed for PM1 and PM10 mass 10 concentrations, as shown in Figure S3 (Supporting Information). 11

12

For the 3 day time period (21st-24th of September) we applied the rank order analysis, two 13 14 subsets of concentrations measured by the OPC-N2 were evident in the time series (Fig 1); 15 one a period of highly variable mass concentrations (0:00 21/9/16 to 12:00 22/9/16) of 16 September) followed by more stable mass concentrations (12:00 22/9/16 onward). This was reflected in the corresponding rank order plots where relatively consistent OPC rank orders 17 18 were observed throughout the variable and comparatively stable PM concentrations periods. 19 However, there is a noticeable transition between the two periods in the rank order plot, observed at approximately 12:00 on the 22nd). This transition in rank orders would reflect the 20 21 difference in OPC PM sensitivities, random noise and offset values between each OPC. Over 22 the 3 day period the OPCs appeared to hold their response characteristics and hence rank 23 orders well, suggesting that over this timescale quantitative concentrations could be directly 24 compared. Due to the changing response and the incomplete data coverage, for the rest of the 25 analysis in this paper, when comparing to the reference instruments the median and inter-26 quartiles concentrations of all 14 OPC-N2 were used.

27





Figure 2: Sensor ranking analysis for measured PM_{2.5} mass concentrations for the 14 OPCN2 over a 3 day period (21st-24th of September) with high OPC-N2 data coverage.

5

6 One measure of the precision of a group of instruments is the coefficient of variance (CV) 7 and this was calculated for the measured ambient mass concentrations of all 14 OPC-N2 to 8 assess the variability between 14 instruments. The average CV was 0.32±0.16, 0.25±0.14 and 9 0.22 ± 0.13 for PM₁, PM_{2.5} and PM₁₀ mass concentrations, respectively. This is higher than the 10 value of 0.1 considered acceptable for duplicate instruments by the US EPA (see Sousan et 11 al., 2016 and references therein) but perhaps not unreasonable for low-cost sensors. This may 12 in part be due the OPC-N2 all sampling from separate but identical inlets but suggests the 13 precision of the OPC-N2 would need to be considered when comparing multiple units. To 14 analyse whether the CV for the OPC-N2 varied over the month, the median concentration 15 was plotted along with the CV (shown for PM_{2.5} in Fig 3). Throughout the measurement 16 period, the CV was fairly consistent (mean of 0.22±0.13), with spikes in CV values evident 17 during periods of high PM_{2.5} concentrations, in agreement with trends observed in Fig 1. We 18 observed a similar trend of consistent CV values for both PM1 and PM10 concentrations 19 suggesting reasonably stable agreement between all OPC-N2 over a 5 week period. 20



Figure 3: Time series of the hourly average median OPC and CV during the September
intensive inter-comparison at EROS for PM_{2.5} mass concentration.

4 **3.2 Comparison of Alpha sense OPC to reference instruments**

5 **3.2.1 Particle mass concentration measurement at EROS**

6

1

7 The median and inter-quartiles of the measured PM concentrations from the 14 OPC-N2 were 8 used to compare the measured particle mass concentrations to the reference instruments 9 (Figure 4). From Fig 4, the notably similar distributions across all three particle size fractions 10 for the first and third quartiles indicate good agreement between the 14 OPC-N2, further 11 highlighting the reasonable degree of precision between the OPC-N2 as shown in the 12 previous section. At typical ambient PM_{2.5} and PM₁₀ mass concentrations for the UK, similar 13 distributions were observed for the OPC-N2 and reference instruments (Fig 1), suggesting 14 reasonable agreement between the devices. In contrast, different distributions were observed 15 for the PM₁ fraction, with the OPC-N2 and GRIMM in agreement but appearing to over-16 estimating the PM₁ mass concentrations with respect to the TSI 3330. While the OPC-N2 has 17 a higher particle size cut-off (0.38 μ m) compared to the TSI (0.3 μ m) and may explain the 18 observed difference in frequency distribution for PM₁ (Fig 1). While the TSI and GRIMM 19 have the same particle size cut-off (0.3 μ m), these instruments have been shown to disagree 20 (Fig S1) possibly due to different particle collection efficiencies.

21



Figure 4: Histogram of measured PM₁, PM_{2.5} and PM₁₀ mass concentrations by the TSI 3330,
GRIMM and median and inter-quartile values for the 14 OPC-N2. Note the different x and y
axis scales.

When the median and inter-quartile OPC-N2 concentrations were plotted against the TSI and
GRIMM concentrations, the slope was greater than unity for all three size fractions (Table 1)
indicating that the OPC-N2 were over-estimating the ambient particle mass concentrations

12

(approx. 2 to 5 times, Table 1). Overall, the OPC-N2 and GRIMM were in better agreement
compared to the TSI for all size fractions (Table 1). The GRIMM was found to record PM
concentrations 20-30% higher compared to the TSI (Figure S1), and this could in part
account for the observed lower slopes between the GRIMM and the OPC-N2.

Table 1: Slopes of measured PM mass concentrations of the reference instruments against the
median and inter-quartiles for OPC-N2. The intercepts were not constrained to zero.
Correlation co-efficient, r² is given in parenthesis.

	PM ₁		PM _{2.5}		PM_{10}	
OPC-N2	TSI	GRIMM	TSI	GRIMM	TSI	GRIMM
25 th	2.93+0.01	2.34+0.1	3.16+0.03	2.62 + 0.02	2.05 + 0.02	1.85 + 0.02
	(0.9)	(0.92)	(0.66)	(0.77)	(0.64)	(0.6)
Median	3.19+0.02	2.63+0.01	3.53+0.04	3.02+0.03	2.29+0.03	2.06+0.02
	(0.86)	(0.91)	(0.63)	(0.76)	(0.57)	(0.67)
75 th	3.90+0.02	3.24+0.02	4.77+0.06	4.21+0.04	2.73+0.04	2.47+0.35
	(0.87)	(0.89)	(0.59)	(0.71)	(0.53)	(0.57)

8

9 The time series of the median OPC-N2 PM_{2.5} concentrations along with the two reference 10 instruments are shown in Figure 5, and for a large portion of the inter-comparison all 11 instruments appear to be in agreement. However, there were a number of times when the OPC-N2 readings were up to an order of magnitude higher relative to the reference (e.g. 15th 12 September), pointing to a significant instrument artefact. On the 15th September, the GRIMM 13 14 and TSI also move out of agreement and may point to the same artefact affecting the 15 GRIMM. Similar trends were also observed for the PM₁ and PM₁₀ mass fractions (Fig S4, Supporting Information) with the OPC-N2 over-estimating the PM₁₀ concentration by several 16 orders of magnitude on 15th September (peak mass concentrations in the order of 15,000 µg 17 m^{-3}). Note that as EROS is an urban background site, it was unlikely to be affected by plumes 18 19 from sources such as vehicles and as a result these high concentrations spikes may not be 20 real.

21

The factors contributing to this apparent artefact shown by the OPC-N2 were investigated. In Fig 6, the agreement between the OPC-N2 and the TSI instrument appears to vary as a

24 function of ambient RH, with better agreement observed between the two instruments during

25 periods of relatively low ambient RH. However, during times when the RH was high (>90%),

1 the OPC-N2 recorded concentrations markedly higher than that measured by the TSI 3330 2 (Fig 6). Similar trends were also observed for PM_1 and PM_{10} mass concentrations (Figure S5, 3 Supporting Information). Thus, it points to ambient RH as a significant contributing factor 4 affecting the particle mass concentrations measured by the OPC-N2, and this is tested further 5 in later sections. There are distinct differences in design in OPC-N2 compared to the 6 reference instruments (GRIMM and TSI 3330) as both the TSI 3330 and GRIMM utilise a 7 sheath flow unlike the OPC-N2. The sheath flow in both devices will be warmed to 8 temperatures higher than the ambient air due to proximity to the instrument pumps and 9 electronics. This would mean that they measure at a lower RH than ambient and could 10 explain why no RH dependence was observed on measured particle concentrations by the GRIMM and TSI 3330. 11

- 12
- 13
- 14



15

Figure 5: Time series of the measured $PM_{2.5}$ mass concentrations by the TSI, GRIMM and median concentration measured by the 14 OPC-N2 at EROS.

18







Figure 6: Measured concentrations by the TSI 3330 compared to the median concentration
measured by the 14 OPC-N2, coloured by the ambient relative humidity. Also shown are the
1:1 (solid) and 0.5:1 and 2:1 (dashed) lines.

5

6 **3.2.3 Comparison to TEOM-FDMS at AURN monitoring station**

7

8 We deployed a subset of the OPC-N2 devices (4) and the GRIMM at an urban background 9 AURN station, to enable comparison of the measured ambient particle mass concentrations to a TEOM-FDMS. The time series of the measured concentrations of PM₁₀ and PM_{2.5} for all 10 11 instruments is shown in Fig 7. The two reference instruments were found to be well correlated ($r^2 > 0.91$, Figure S6, Supporting Information) but with the GRIMM reading was 12 about 20% lower than the TEOM, in agreement with previous work (Grover et al., 2006). 13 14 From Fig 6, periods of agreement between the four OPC-N2 and the reference instruments 15 (GRIMM and TEOM) were apparent, along with times when the four OPC-N2 measured 16 concentrations that were notably higher than the reference instruments. Overall, when compared to the TEOM, the OPC-N2 measurements were 2.5-3.9 times higher for both the 17 PM_{10} and $PM_{2.5}$, with considerable scatter observed (Table 2). 18

19

20



Figure 7: Time series for hourly measured PM mass concentrations by the TEOM, four OPCN2 and GRIMM at Tyburn Rd urban background AURN station. The volatile particle mass
concentration as measured by the TEOM-FDMS and relative humidity measured at Tyburn
Rd also shown."

8

9 Closer inspection of Fig 7 indicated that the times when the four OPC-N2 over-estimated the particle mass concentrations were during times of high RH (e.g. 12-14th Feb), as observed in 10 the previous section. However, there were periods of high RH when the four OPC-N2 and 11 TEOM were in better agreement (e.g. 20th Feb onwards), indicating that the large positive 12 13 artefact observed in the OPC-N2 was not just related to RH. Rather, it appears that positive 14 artefact was observed during times when the volatile fraction measured by the TEOM was relatively high, as well as higher RH, as was observed on 12-14th Feb (Fig 7). Thus, it 15 suggests that the ambient aerosol composition also contributed to the significant positive 16 17 artefact in the OPC-N2. A recent laboratory study found that the particle mass concentrations measured by OPC-N2 for all three size fractions were highly linear with respect to 18 19 gravimetrically corrected reference instruments but that the slope was dependent on the 20 aerosol type (Sousan et al., 2016). Sousan et al. (2016) observed in the PM_{10} fraction slopes greater than unity for Arizona road dust but less than unity for salt and therefore suggest that 21 22 changes in aerosol composition may also account for the differences observed between the 23 reference instruments and OPC-N2 (Figs 7). This result highlights a limitation when 24 comparing optical methods to gravimetric - as differences may be due to changes in particle

mass, size distribution or composition: as all can affect the ability of a particle to scatter light
(Holstius et al., 2014).

3

From Fig 6, the times when there was a large positive artefact in the OPC-N2 occurred when the RH was above 85%. If we exclude these times when the RH was over this threshold, better agreement between the four OPC-N2 and the TEOM was observed, with slopes between 1.1-1.7 for both size fractions (Table 2). One of the OPC-N2 recorded notably higher mass concentrations compared to the reference instruments (OPC11), compared to the other three OPC-N2 (Table 2), and this highlights the need to calibrate each OPC individually before use in field measurements.

11

12 Table 2: Slopes of measured PM mass concentrations of the reference instruments (TEOM

13 and GRIMM) against the OPC-N2. The correlation co-efficient, r^2 is given in parenthesis.

14 The intercepts were not constrained to zero.

15

		PM ₁₀			PM _{2.5}				
		OPC6	OPC8	OPC10	OPC11	OPC6	OPC8	OPC10	OPC11
ALL	TEOM	2.6	2.8	2.5	3.5	3.3	3.1	2.9	3.9
		(0.64)	(0.68)	(0.64)	(0.67)	(0.7)	(0.74)	(0.7)	(0.72)
	GRIMM	3.7	3.6	3.2	4.4	3.8	3.7	3.4	4.6
		(0.66)	(0.69)	(0.66)	(0.68)	(0.71)	(0.74)	(0.71)	(0.72)
	TEOM	1.4	1.4	1.2	1.7	1.3	1.4	1.1	1.6
<85%		(0.82)	(0.83)	(0.83)	(0.83)	(0.79)	(0.8)	(0.79)	(0.79)
RH	GRIMM	1.8	1.9	1.6	2.2	2.0	2.1	1.7	2.4
		(0.83)	(0.84)	(0.84)	(0.84)	(0.89)	(0.89)	(0.9)	(0.88)

16

17 **3.3 Development of correction factor for ambient RH**

Clearly there were times when there was a significant instrument artefact for the OPC-N2 (Figs 4 and S4) and the highest over-estimations occurred at high RH at both EROS and Tyburn Rd (e.g. Fig 5 and 6). Whilst the accuracy of the instrument was significantly worse at high RH the precision remains the same within error. The CV analysis conducted in section 3.1.2 is repeated for the same dataset but put into low (RH<85%) and high RH (RH>85%) subsets. For high RH conditions the CV for PM₁, PM_{2.5} and PM₁₀,

- was 0.34±0.30, 0.27±0.14 and 0.23±0.21, respectively. For low RH conditions the CV for
 PM₁, PM_{2.5} and PM₁₀, was 0.30±0.25, 0.23±0.14 and 0.20±0.18, respectively.
- 3

The size of hygroscopic particles is known to be dependent on RH, as the particle refractive index and size are both a function of RH. Inorganic aerosols (e.g. sodium chloride, nitrate and sulphate), make up a large portion of the PM_{10} observed at EROS (Yin et al., 2010), and are known to demonstrate an exponential increase in hygroscopic growth at high RH (e.g. (Hu et al., 2010; Pope et al., 2010).

9

10 The ratio of measured mass concentrations by the OPC-N2 relative to the reference 11 instruments was plotted as a function of RH, and appeared to show an exponential increase 12 above ~85% RH, similar to hygroscopic particle growth curves (Pöschl, 2005). As a result, 13 we applied κ -Kohler theory (Petters and Kreidenweis, 2007), which describes the relationship 14 between particle hygroscopicity and volume by a single hygroscopicity parameter, κ . The κ -15 Kohler theory can be adapted to relate particle mass to hygroscopicity at a given RH by 16 equation 5 (Pope, 2010):

17

18
$$a_w = \frac{(m/m_o - 1)}{(m/m_o - 1) + (\frac{\rho_w}{\rho_p}\kappa)}$$
 (5)

19

20 Where a_w is the water activity (a_w = ambient RH/100), m and m_o are the wet and dry (RH = 0%) aerosol mass, respectively. The density of the dry particles and water is given by ρ_w and 21 ρ_{p} , respectively. The density of water is 1 g cm⁻³, and the bulk dry particle density is assumed 22 to be 1.65 g cm⁻³. The value for κ can be found by a non-linear curve fitting of a humidogram 23 24 (m/m_o vs a_w), and was calculated using the TEOM measurements at Tyburn Rd in the first 25 instance as the TEOM system employs a Nafion dryer and so measures dry particle mass 26 (Grover et al., 2006). To account for the differences in mass concentration measured by the 27 TEOM and OPC-N2 at RH less than 85%, the scaling factors shown in Table 2 are used 28 calibrate the dry mass of the OPC-N2 to that observed in the TEOM, both in the PM_{2.5} and 29 PM₁₀ fractions.

30

Figure 8 shows the humidogram plots, for both the $PM_{2.5}$ and PM_{10} fractions, obtained by plotting the ratio of OPC-N2 to the reference instrument (TEOM and GRIMM) outputs versus RH. When using the TEOM for m_o, similar κ constants were calculated for all OPC- 1 N2, ranging from 0.38-0.41 and 0.48-0.51 for $PM_{2.5}$ and PM_{10} , respectively, which is within 2 the expected range for Europe (0.36 ± 0.16, (Pringle et al., 2010). Similar κ values were 3 observed when using the GRIMM mass concentrations as the dry particle mass (m_o), ranging 4 from 0.41-0.44 and 0.38-0.41 for PM_{2.5} and PM₁₀, respectively.

- 5
- 6





7

9 Figure 8: Measured and fitted humidograms (m/m_o vs RH) recorded at the Tyburn Road 10 AURN site for PM_{10} and $PM_{2.5}$ size fractions and reference instruments (TEOM and 11 GRIMM). The dry mass (m0) is given by the TEOM or GRIMM and the humidified mass is 12 given by the OPC-N2. Measured data is given by the black circles, the fitted data is given by 13 the blue (TEOM-FDMS) and red (GRIMM) line.

14

We then applied this fitting constant to model the expected OPC/Reference instrument ratio
 for a given RH as a result of particle hygroscopic growth, by re-arranging Equation 5:

3

$$4 \qquad \frac{m}{m_o} = 1 + \frac{\frac{\rho_w}{\rho_p}\kappa}{-1 + \frac{1}{a_w}} \tag{6}$$

5

6 Where the m/m_0 is the ratio of the OPC-N2 to the reference instruments. Using Equation 6, 7 the mass concentrations measured by the OPC-N2 were corrected and significantly better 8 agreement between the corrected OPC-N2 and reference instruments was observed for 9 measurements across the whole range of ambient RH (Tables 2 and 3). Overall, the corrected 10 OPC-N2 mass concentrations using Eqn 6 were notably better, within 33% and 52% of the 11 TEOM and GRIMM, respectively. (Table 3) compared to 250-400% without the correction 12 factor (Table 2). The time series for the corrected data is shown in Figures S7 and S8 13 (Supporting Information) and there are periods were there is good agreement between TEOM 14 and the corrected OPC-N2.

There were also times when the OPC-N2 were clearly over-corrected (e.g. from 20th February 15 16 onwards), generally when the ambient RH was low (Fig 6). This suggests that when the RH 17 was below a threshold, Eqn 6 overcorrects the data and this can be observed in the 18 humidograms shown in Figure 8. Typically, at RH <85% the hygroscopic growth of real 19 atmospheric aerosols is small and it may be more appropriate to apply a linear regression 20 correction factor for data recorded under these RH conditions. Therefore we applied a 21 binary two model approach to correct the OPC-N2 mass concentrations, where a linear 22 correction (using the TEOM as reference concentration) for when RH <85%, and above 23 this threshold in RH Eqn 6 was used. As can be seen Figure S9 (Supporting Information), there was little change in the slope or r² value with the two model correction compared 24 25 to the using correction with Eqn 6 for all RH. What was noticeable was that the intercept 26 for the two model approach moved closer to zero, suggesting that at the lower mass concentrations the correction was improved. Similar trends were also observed for 27 **PM₁₀**. Also during the period from the 20^{th} February, the volatile particle fraction was also 28 29 lower (Fig 6) and this indicates a significantly different aerosol composition. Since κ is 30 composition dependent, a single global fit to κ will result in poor fitting when the true κ is 31 significantly different to the average κ . The preceding discussion suggests that further refinement to the correction factors applied to the OPC-N2 is possible, depending on the ambient RH and better knowledge of aerosol composition. RH measurement is relatively trivial and can be achieved with small sensors but aerosol composition determination still requires significant analytical equipment and expertise.

5

7

6 Table 3: Summary of the comparison between the corrected OPC-N2 (via Eqn 6) against the

OPC-N2	TEOM		GRIMM	
	<i>PM</i> _{2.5}	PM_{10}	<i>PM</i> _{2.5}	<i>PM</i> ₁₀
OPC6	1.08±0.03	0.87±0.02	1.26±0.03	1.27±0.03
OPC8	1.11±0.03	0.89±0.02	1.29±0.03	1.23±0.03
OPC10	0.98±0.03	0.80±0.02	1.16±0.03	1.17±0.03
OPC11	1.33±0.04	1.06±0.03	1.53±0.04	1.51±0.04

reference instruments. Intercepts were not constrained to zero.

8

9 **3.3.1 Longer-term monitoring with OPC-N2 at EROS**



Figure 9: Histogram of measured $PM_{2.5}$ concentrations by the GRIMM PAS 1.108 and the 4 OPC-N2s for January. The uncorrected OPC-N2 concentrations are shown in the left plot

17 (A), while the right plot (B) shows the RH corrected OPC-N2 concentrations.

1

2 After the conclusion of the intensive measurements at EROS (Section 3.1), five of the OPC-3 N2 continued monitoring for a further 4 months to examine if there was any evidence of 4 instrument drift over time, along with the GRIMM as reference. One of the OPC-N2 failed in 5 December, and so was excluded from this analysis. The remaining four OPC-N2 were compared to GRIMM and in January after running for 4 months (Fig 9A), and while three of 6 7 the OPC-N2 had a similar distribution to the GRIMM (OPC12, 13 and 14), OPC9 appeared 8 to show evidence for instrument drift as the mode has shifted relative to the GRIMM. 9 However, the increased frequency of higher mass concentrations not observed by the 10 GRIMM but by all four OPC-N2 (Fig 9A) suggests that ambient RH is also a factor, as the 11 average RH in January (91%) was higher than September (84%). Therefore, we calculated 12 the correction for RH as described in the previous section (Eqn 6), as changes in aerosol 13 composition would affect the particle hygroscopicity. In addition, the κ was only fitted for the 14 data with RH < 95% since the hygroscopicity of aerosol is highly sensitive to any error in the 15 RH measurement above this value. Application of the RH correction factor resulted in better 16 agreement between each of the OPC-N2, with similar corrected distributions observed (Fig 17 9B). Furthermore, the corrected OPC-N2 concentrations also had better agreement with the 18 GRIMM during January (Fig 9B) compared to uncorrected concentrations (Fig 9A), 19 suggesting that changes in the particle water content were the cause. Thus, at least over a four 20 month measurement period, there appears to be no evidence for instrument drift in the OPC-21 N2, once appropriate correction factors were applied.

3.4 Discussion on the OPC-N2 interferences

23 In the previous sections, the significant positive artefact observed by the OPC-N2 relative to 24 the reference instruments were at times when the ambient RH was high, pointing to particle 25 water content as the cause. This result is perhaps not surprising, as many studies in the 26 literature have shown that particle water content can be a major reason for discrepancies 27 between techniques that measure ambient particle mass (See e.g. (Charron et al., 2004)). The 28 use of ĸ-Kohler theory to derive a correction factor based on ambient RH improved the 29 agreement between the OPC-N2 and reference instruments; however a limitation of this 30 approach is that the bulk aerosol hygroscopicity is related to particle composition, typically 31 the inorganic fraction (e.g. (Gysel et al., 2007)). Variation in ambient particle composition 32 could account for the large spread observed in the ratio of OPC-N2/TEOM at high RH (Fig 33 7), as an average hygroscopicity correction will overestimate when PM with higher

1 hygroscopicity is measured and vice versa. Furthermore, Eqn 6 may not be required for 2 locations where the ambient RH is lower than 85%, as typically atmospheric particle growth 3 due to water below this threshold is limited and a simple linear regression may be sufficient. 4 Thus, in-situ and seasonally specific calibrations for the OPC-N2 are required to account for 5 possible differences in ambient aerosol properties. However as κ values for continental 6 regions tend to fall within a narrow range globally (0.3±0.1, (Andreae and Rosenfeld, 2008), 7 with some systematic deviations for certain regions (Pringle et al., 2010), this average κ value 8 could be used in lieu of calibration with reference instrument (e.g. a TEOM) to determine the 9 correction factor (C) according to Eqn 7:

10

11
$$C = 1 + \frac{0.3/1.65}{-1 + \frac{1}{a_W}}$$
 (7)

12

However, it should be noted that while *in situ* calibration of an OPC-N2 with suitable reference instrumentation is preferable, for many locations around the world, and especially low and middle income countries (LMICs), this may not be possible and so using an appropriate κ value from the literature in Eqn 7 may be a reasonable approximation.

18 **4.0** Applicability of OPC-N2 for ambient monitoring

19 The Alphasense OPC-N2 was evaluated for use in ambient monitoring of airborne particle 20 mass concentration, with TEOM-FDMS and two commercial optical light scattering 21 instruments; GRIMM PAS 1.108 and TSI 3330 employed as reference instruments. 22 Comparison of the OPC-N2 to the reference optical instruments demonstrated reasonable 23 agreement for the measured mass concentrations of PM₁, PM_{2.5} and PM₁₀ as evidenced by the 24 stated accuracy and precision. However, the OPC-N2 demonstrated a significant large 25 positive artefact in measured particle mass during times of high ambient RH, and a 26 calibration factor was developed based on bulk particle aerosol hygroscopicity. Application 27 of the RH correction factor, based upon κ-Kohler theory, resulted in notable improvement 28 with the corrected OPC-N2 measurements within 33% of a TEOM-FDMS. While higher 29 than the slope of 1±0.1 allowed by the US EPA, it is comparable to the agreement of a 30 GRIMM to the TEOM (20%). All low cost PM sensors will likely require calibration factors 31 to obtain the dry particle weight unless they actively dry the PM containing air stream before 32 it enters the device. The use of heated inlets could be used to reduce the RH in the air stream

1 but would have consequences on the power requirements of the sensor, potentially making

- 2 them less attractive for battery led operation. For PM_{10} mass concentrations, a CV of
- 3 22+13% between the 14 OPC-N2 employed in this study was observed, with some of the
- 4 variability likely due to use of separate but identical inlets, and therefore could be
- 5 considered reasonable for a low-cost sensor but this level of precision needs to be
- 6 considered when using multiple units. One out of four OPC-N2 tested for long-term
- 7 monitoring appeared to show evidence for instrument drift relative to reference instruments.
- 8 Overall, the OPC-N2 have been shown to accurately measure ambient airborne particle mass
- 9 concentration provided they are correctly calibrated and corrected for RH. The reasonable
- 10 level of precision demonstrated between multiple OPC-N2 suggests that they would be
- 11 suitable for applications where a number of instruments are required such as spatial mapping
- 12 and personal exposure studies.

13 Acknowledgements

- 14 The authors wish to thank Peter Porter and Birmingham City Council for help in collocating
- 15 the sensors next to the Tyburn Road AURN site. Funding is acknowledged from EPSRC
- 16 (Global Challenges Research Fund IS2016). AL and MS acknowledge funding from the
- 17 NERC National Capability programme ACREW and NE/N007115/1
- 18

19

1 References

- 2 Andreae, M.O., Rosenfeld, D., 2008. Aerosol-cloud-precipitation interactions. Part 1. The
- 3 nature and sources of cloud-active aerosols. Earth-Science Reviews 89, 13-41.
- 4 Borrego, C., Costa, A.M., Ginja, J., Amorim, M., Coutinho, M., Karatzas, K., Sioumis, T.,
- 5 Katsifarakis, N., Konstantinidis, K., De Vito, S., Esposito, E., Smith, P., André, N., Gérard,
- 6 P., Francis, L.A., Castell, N., Schneider, P., Viana, M., Minguillón, M.C., Reimringer, W.,
- 7 Otjes, R.P., von Sicard, O., Pohle, R., Elen, B., Suriano, D., Pfister, V., Prato, M., Dipinto, S.,
- 8 Penza, M., 2016. Assessment of air quality microsensors versus reference methods: The
- 9 EuNetAir joint exercise. Atmospheric Environment 147, 246-263.
- 10 Castellini, S., Moroni, B., Cappelletti, D., 2014. PMetro: Measurement of urban aerosols on a
- 11 mobile platform. Measurement 49, 99-106.
- 12 Charron, A., Harrison, R.M., Moorcroft, S., Booker, J., 2004. Quantitative interpretation of
- 13 divergence between PM10 and PM2.5 mass measurement by TEOM and gravimetric
- 14 (Partisol) instruments. Atmospheric Environment 38, 415-423.
- de Nazelle, A., Bode, O., Orjuela, J.P., 2017. Comparison of air pollution exposures in active
- 16 vs. passive travel modes in European cities: A quantitative review. Environment International
- 17 99, 151-160.
- 18 Gao, M., Cao, J., Seto, E., 2015. A distributed network of low-cost continuous reading
- sensors to measure spatiotemporal variations of PM2.5 in Xi'an, China. EnvironmentalPollution 199, 56-65.
- 21 Grover, B.D., Eatough, N.L., Eatough, D.J., Chow, J.C., Watson, J.G., Ambs, J.L., Meyer,
- 22 M.B., Hopke, P.K., Al-Horr, R., Later, D.W., Wilson, W.E., 2006. Measurement of Both
- 23 Nonvolatile and Semi-Volatile Fractions of Fine Particulate Matter in Fresno, CA. Aerosol
- 24 Science and Technology 40, 811-826.
- 25 Gysel, M., Crosier, J., Topping, D.O., Whitehead, J.D., Bower, K.N., Cubison, M.J.,
- 26 Williams, P.I., Flynn, M.J., McFiggans, G.B., Coe, H., 2007. Closure study between chemical
- 27 composition and hygroscopic growth of aerosol particles during TORCH2. Atmos. Chem.
- 28 Phys. 7, 6131-6144.
- Hagan, D., 2017. py-opc, <u>https://github.com/dhhagan/py-opc</u>.
- 30 Holstius, D.M., Pillarisetti, A., Smith, K.R., Seto, E., 2014. Field calibrations of a low-cost
- aerosol sensor at a regulatory monitoring site in California. Atmos. Meas. Tech. 7, 1121 1131.
- Hu, D., Qiao, L., Chen, J., Ye, X., Yang, X., Cheng, T., Fang, W., 2010. Hygroscopicity of
- 34 inorganic aerosols: size and relative humidity effects on the growth factor. Aerosol and Air 25 Ouglity Research 10, 255, 264
- 35 Quality Research 10, 255-264.
- 36 Kumar, P., Morawska, L., Martani, C., Biskos, G., Neophytou, M., Di Sabatino, S., Bell, M.,
- Norford, L., Britter, R., 2015. The rise of low-cost sensing for managing air pollution in
 cities. Environment International 75, 199-205.
- 39 Lewis, A.C., Lee, J.D., Edwards, P.M., Shaw, M.D., Evans, M.J., Moller, S.J., Smith, K.R.,
- 40 Buckley, J.W., Ellis, M., Gillot, S.R., White, A., 2016. Evaluating the performance of low
- 41 cost chemical sensors for air pollution research. Faraday Discussions 189, 85-103.
- 42 Manikonda, A., Zíková, N., Hopke, P.K., Ferro, A.R., 2016. Laboratory assessment of low-
- 43 cost PM monitors. Journal of Aerosol Science 102, 29-40.
- 44 Mead, M.I., Popoola, O.A.M., Stewart, G.B., Landshoff, P., Calleja, M., Hayes, M., Baldovi,
- 45 J.J., McLeod, M.W., Hodgson, T.F., Dicks, J., Lewis, A., Cohen, J., Baron, R., Saffell, J.R.,
- 46 Jones, R.L., 2013. The use of electrochemical sensors for monitoring urban air quality in low-
- 47 cost, high-density networks. Atmospheric Environment 70, 186-203.

- 1 Mueller, M., Meyer, J., Hueglin, C., 2017. Design of an ozone and nitrogen dioxide sensor
- unit and its long-term operation within a sensor network in the city of Zurich. Atmos. Meas.
 Tech. Discuss. 2017, 1-29.
- 4 Petters, M., Kreidenweis, S., 2007. A single parameter representation of hygroscopic growth
- 5 and cloud condensation nucleus activity. Atmospheric Chemistry and Physics 7, 1961-1971.
- 6 Pope, F.D., 2010. Pollen grains are efficient cloud condensation nuclei. Environmental
- 7 Research Letters 5, 044015.
- 8 Pope, F.D., Dennis-Smither, B.J., Griffiths, P.T., Clegg, S.L., Cox, R.A., 2010. Studies of
- 9 single aerosol particles containing malonic acid, glutaric acid, and their mixtures with sodium
- 10 chloride. I. Hygroscopic growth. The Journal of Physical Chemistry A 114, 5335-5341.
- 11 Popoola, O.A.M., Stewart, G.B., Mead, M.I., Jones, R.L., 2016. Development of a baseline-
- temperature correction methodology for electrochemical sensors and its implications forlong-term stability. Atmospheric Environment 147, 330-343.
- 14 Pöschl. U., 2005. Atmospheric aerosols: composition, transformation, climate and health
- 15 effects. Angewandte Chemie International Edition 44, 7520-7540.
- 16 Pringle, K.J., Tost, H., Pozzer, A., Pöschl, U., Lelieveld, J., 2010. Global distribution of the
- 17 effective aerosol hygroscopicity parameter for CCN activation. Atmos. Chem. Phys. 10,
- 18 5241-5255.
- 19 Rai, A., Kumar, P., Pilla, F., Skouloudis, A., Di Sabatino, S., Ratti, C., Yasar, A., Rickerby,
- 20 D., 2017. End-user Perspective of Low-cost Sensors for Outdoor Air Pollution Monitoring.
- 21 Science of The Total Environment.
- 22 Smith, K.R., Edwards, P., Evans, M.J., Lee, J.D., Shaw, M.D., Squires, F.A., Lewis, A.,
- 23 2017. Clustering approaches to improve the performance of low cost air pollution sensors.
 24 Faraday Discussions, 1-15.
- 25 Snyder, E.G., Watkins, T.H., Solomon, P.A., Thoma, E.D., Williams, R.W., Hagler, G.S.W.,
- 26 Shelow, D., Hindin, D.A., Kilaru, V.J., Preuss, P.W., 2013. The Changing Paradigm of Air
- 27 Pollution Monitoring. Environmental Science & Technology 47, 11369-11377.
- 28 Sousan, S., Koehler, K., Hallett, L., Peters, T.M., 2016. Evaluation of the Alphasense optical
- 29 particle counter (OPC-N2) and the Grimm portable aerosol spectrometer (PAS-1.108).
- 30 Aerosol Science and Technology 50, 1352-1365.
- 31 Steinle, S., Reis, S., Sabel, C.E., Semple, S., Twigg, M.M., Braban, C.F., Leeson, S.R., Heal,
- 32 M.R., Harrison, D., Lin, C., Wu, H., 2015. Personal exposure monitoring of PM2.5 in indoor
- and outdoor microenvironments. Science of The Total Environment 508, 383-394.
- 34 Viana, M., Rivas, I., Reche, C., Fonseca, A.S., Pérez, N., Querol, X., Alastuey, A., Álvarez-
- 35 Pedrerol, M., Sunyer, J., 2015. Field comparison of portable and stationary instruments for
- 36 outdoor urban air exposure assessments. Atmospheric Environment 123, 220-228.
- 37 Yin, J., Harrison, R.M., Chen, Q., Rutter, A., Schauer, J.J., 2010. Source apportionment of
- 38 fine particles at urban background and rural sites in the UK atmosphere. Atmospheric
- 39 Environment 44, 841-851.
- 40

Supporting Information for:

Evaluation of a low-cost optical particle counter (Alphasense OPC-N2) for ambient air monitoring

Leigh R. Crilley¹, Marvin Shaw², Ryan Pound², Louisa J. Kramer¹, Robin M. Price³, Stuart Young², Alastair C Lewis², Francis D. Pope^{1*}

 ¹School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham, United Kingdom, B15 2TT.
 ²National Centre for Atmospheric Science, Wolfson Atmospheric Chemistry Laboratories, , University of York, York, United Kingdom, YO10 5DD.
 ³Birmingham Open Media (BOM), 1 Dudley Street, Birmingham, B5 4EG.

*Corresponding author - <u>f.pope@bham.ac.uk</u>



Fig S1: Comparison of the TSI 3330 and GRIMM PAS 1.108 at EROS during the September intensive measurement period for PM_1 (top), $PM_{2.5}$ (middle) and PM_{10} (bottom) mass concentrations.



Figure S2: PM_1 and PM_{10} concentrations measure by all OPC-N2 and the reference instruments, TSI and GRIMM for selected period with high data coverage.



Davs Figure S3: Rank order plot of OPC observations (1 min averaged at $10\mu m$) over a 3 day period (21st – 24th of September), y-axis according to OPC ranking in the final hour of time series.



Fig S4: Time series of the measured PM1 and PM10 mass concentrations by TSI and GRIMM and the median mass concentration measured by the OPC-N2





Figure S5: Measured concentrations by the TSI 3330 OPC compared to the median concentration measured by the 14 Alphasense OPCs for PM_1 and PM_{10} mass concentrations, coloured by the ambient relative humidity. Also shown are the 1:1 (solid) and 0.5:1 and 2:1 (dashed) lines.





Figure S6: Comparison of the GRIMM and TEOM (y axis) during the measurements at Tyburn Rd



Figure S7: Time series of uncorrected OPC-N2 (top panel) and corrected OPC-N2 $PM_{2.5}$ mass concentrations at Tyburn Rd. The TEOM and GRIMM concentrations shown for reference.



concentrations at Tyburn Rd. The TEOM and GRIMM concentrations shown for reference.



Figure S9: Scatterplots of corrected OPC-N2 against the TEOM for $PM_{2.5}$ mass concentrations. The two model approach (Cv2) is in red and the one model approach in blue.