#### Dear Editor,

We thank you for the time and care you have put into this paper. We also agree that this paper is an important one and so have carefully addressed all of your comments, and modified the text accordingly. Please see our responses below.

**1.** While I appreciate the work done for improving the manuscript based on reviewers suggestions/comments, I still feel some additional corrections are needed before publication.

Essentially, based on the evidence provided, I am not fully convinced that "the OPC-N2 instrument demonstrated reasonable agreement with reference instrument for the measured mass concentration", which seems like a very positive statement considering the obvious limitations for its applicability. I believe you have to be much more cautious in your conclusions.

#### Response:

We think that for a low-cost sensor that the OPC-N2 does show reasonable agreement to the reference instruments used in this paper, especially considering that the OPC-N2 is at least 1/100<sup>th</sup> the cost of a TEOM-FDMS system. We are not trying to say that the agreement was excellent or great but rather can be considered as reasonable for low-cost sensor as evidenced by the stated precision and accuracy throughout the paper. The precision of the OPC-N2 was found to be unaffected by RH, with a CV of 22+13% between the 14 OPC-N2 employed in this study for PM10. The accuracy of the OPC-N2 relative to the optical reference instruments, as shown by the similar distributions in measured particle mass concentrations (Fig 4), suggests that there was similar measurement. A large positive artefact was observed at high RH (>85%), hence the need for a correction factor, which resulted in OPC-N2 being within 33% of a TEOM-FDMS. Therefore, we believe we are justified in stating, for a low-cost sensor, the agreement between reference and OPC-N2 measurements was reasonable.

*Therefore to clarify this point we have modified this sentence found in the abstract (line 23) and Section 4.0 (line 22) to now read:* 

# "Comparison of the OPC-N2 to the reference optical instruments demonstrated reasonable agreement, for a low-cost sensor, to the measured mass concentrations of PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10.</sub>"

**2.** First-of-all, there is a clear need for RH correction and this is far from being obvious. Humidograms in Fig. 8 are biased with respect to usual hygroscopic growth curves showing an increase from RH85% instead of RH70% for natural aerosol – your statement that « at RH <85% the hygroscopic growth of real aerosol is small » is actually false and should be RH70%-. I believe this is because you are considering ambient RH while the RH inside the instrument after travelling through the inlet is already lower. This can be fixed for the specific case of this study but without internal RH measurement, the correction may be quite different. This means, the instrument cannot be used without a clear RH controlling process. Then, you apply a correction assuming a  $\kappa$ -value. In practice, given the short duration of your experiment, a constant  $\kappa$  is acceptable, but it is no longer the case when instrument is used for longer periods. I believe you actually can see different aerosol populations in fig8 at high RH, leading to huge differences in OPC/TEOM. Applicability of a constant RH/  $\kappa$  correction scheme is to be questioned for longer measurement periods. This has to be made more clear.

Response: In the paper, we state that "The ratio of measured mass concentrations by the OPC-N2 relative to the reference instruments was plotted as a function of RH, and appeared to show an exponential increase above ~85% RH, similar to hygroscopic particle growth curves."

#### And

"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"

Thus we were not saying that there would be no hygroscopic growth under 85% RH, rather that we observed a near exponential increase above 85%, and that below this threshold a linear regression may be more appropriate.

The correction factor developed requires the input of ambient RH because we believe the measured PM mass is due to the hygroscopic growth of the particles. We agree that a calibration that uses internal RH would be better but this is impossible because the OPC-N2 cannot be opened. We did consider using a further calibration parameter that accounted for the change from ambient to internal RH. However, we felt that incorporating multiple factors in the calibration led to less confidence in the calibration because multiple parameters always lead to a better fit whether or not they are physically realistic. We also note that since the OPC-N2 inlet does not have a sheath flow gas then the temperature difference, and hence RH difference, between ambient and inside the instrument is likely small. To clarify this potential issue with the correction factor, we have included the following text at line 33, page 18:

"Ideally, a measure of RH internal to the instrument could be made to allow for calculation of particle hygroscopicity within the instrument. However, the OPC-N2 design does not allow for this, so we assume that ambient and instrument RH are identical. In reality, the instrument is likely to be slightly warmer than ambient and hence the RH within the instrument will be slightly lower than ambient. This difference will result in a lower apparent hygroscopicity."

We do not agree with the statement the OPC cannot be used without clear RH controlling process, as we show that below an RH of 85% the correction factor was near linear. This suggests that what is needed is calibration, an issue common to all instruments.

We note that we do discuss the issue of the large spread at high RH in Fig 8 in Section 3.4, where we discuss that this is likely due to changes in aerosol composition and that this would affect the  $\kappa$ -value, we already state the k is composition dependent. Therefore we have added the following in Section 3.4 to discuss the implications on long-term monitoring:

"Variation in ambient particle composition could account for the large spread observed in the ratio of OPC-N2/TEOM at high RH (Fig 8), as an average hygroscopicity correction will overestimate when PM with higher hygroscopicity is measured and vice versa for lower hygroscopicity particles. This would have potentially significant implications when using the OPC-N2 for longer-term monitoring, as the  $\kappa$  value may not be constant over the monitoring period. Therefore, this suggests the need for regular calibrations to account for changes in bulk aerosol composition and as a result  $\kappa$  values."

To emphasize this important point that  $\kappa$ -value is composition dependent to the reader; we have added text to the conclusions, see our response to comment 4.

**3.** I am also questioning Table 3 and the huge difference for TEOM in PM2.5 and PM10, which does not seem to be discussed. There is a loss mechanism somewhere that is not accounted for it seems.

#### Response:

We have added the following text as a discussion on the difference observed for PM2.5 and PM10 relative to the TEOM, at page 20, line 14:

"However, it was also evident from Table 3 that the slope was different for  $PM_{10}$  and  $PM_{2.5}$  mass fractions for all OPC-N2 when compared to the TEOM, and suggests that differing responses for the OPC-N2 to two size fractions. This may be related to the observed variation in  $\kappa$  between the size fractions relative to the TEOM or an unaccounted loss mechanism; the exact cause will be investigated further in future work."

**4.** Your paper is an important one and this is why you have to be very careful with the final statements. Essentially, what needs to be known is whether the instrument responds to PM quality objectives for air quality studies, and, if not, which kind of corrections are needed for its applicability. For me, your study shows that the sensor does not respond to DOQ as formulated for PM2.5 or PM10 standard application but, applying specific correction procedures, you can reach a reasonable agreement. Not vice versa. In the case of PM2.5 where the standard is a 1-yr average, I also tend to believe the correction procedures will be meaningless, given the intrinsic variability in RH/  $\kappa$ . This has to be made very clear in the conclusions, otherwise, the risk is high that your paper will be simply referenced as blank check for applying this sensor to any kind of PM studies, without the proper limitations in mind.

#### Response:

We have added the following text to Section 4.0 to highlight these limitations:

"All low cost PM sensors will likely require calibration factors to obtain the dry particle weight unless they actively dry the PM containing air stream before it enters the device. The use of heated inlets could be used to reduce the RH in the air stream but would have consequences on the power requirements of the sensor, potentially making them less attractive for battery led operation. Thus it highlights that the OPC-N2 does not respond the same as reference instruments to ambient particle mass, but provided appropriate correction factors are applied, reasonable agreement with OPC-N2 to reference instruments can be achieved. Furthermore, the dependence of the OPC-N2 on a correction for RH and  $\kappa$  may limit its application for longer-term monitoring as the  $\kappa$  value may change over time, and this will be the focus of future work. This is especially salient when considering using the OPC-N2 to compare to air quality standards that are one-year averages of PM<sub>2.5</sub> and PM<sub>10</sub>."

In conclusion, I expect that you modify the paper in this direction before proceeding with publication.

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

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### 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 a low-cost 25 sensor, to the measured mass concentrations of  $PM_1$ ,  $PM_{2.5}$  and  $PM_{10}$ . However, the OPC-N2 26 demonstrated a significant positive artefact in measured particle mass during times of high 27 ambient RH (>85%) and a calibration factor was developed based upon  $\kappa$ -Kohler theory, 28 using average bulk particle aerosol hygroscopicity. Application of this RH correction factor 29 resulted in the OPC-N2 measurements being within 33% of the TEOM-FDMS, comparable 30 to the agreement between a reference optical particle counter and the TEOM-FDMS (20%). 31 Inter-unit precision for the 14 OPC-N2 sensors of 22±13% for PM<sub>10</sub> 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 11 75x60x65 mm and weighs under 105 g, and as such is significantly cheaper (approx. £200) 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 14 to 17 µm across 16 size bins, and maximum particle count of 10,000 per second. The 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<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> size 17 fraction according to European Standard EN481 (OPC-N2 manual). According the OPC-N2 18 manual, the standard definition for PM<sub>10</sub> in EN 481 extends beyond the particle size measured by the OPC-N2, and may consequently underestimate PM10 value by up to 10%. 19 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<sup>-3</sup>. 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 OPC-N2 were deployed at EROS, enabling the precision of the OPC-N2 to be assessed along 20 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 26<sup>th</sup> August till 3<sup>rd</sup> October 2016, 22 during which all 14 OPC-N2, TSI 3330 and GRIMM sampled ambient air. Minimal lengths 23 (12cm) of stainless steel tubing (OPC-N2) and conductive black tubing (TSI 3330 and 24 25 GRIMM) were used to sample outside air, with each OPC having its own inlet at a height of 26 1.5 m. The vertical inlet for the TSI 3330 necessitated a bend in the tubing, however the 27 calculated sampling efficiency (using von der Weiden et al., 2009) was 92% for particles with 28 a diameter of 10 µm. Therefore, while the inlet arrangement of the TSI 3330 may have 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 28 TSI AIM software to convert the particle count concentration to particle mass measurements. 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  $\rho$  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<sup>-3</sup>. 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**

#### **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<sub>2.5</sub> 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  $\mu$ g m<sup>-3</sup>, whereas the reference instruments reported ~10 13  $\mu$ g m<sup>-3</sup>. The cause of the positive artefact is investigated in later sections, but it points to the 14 15 individual OPC-N2 responding differently to this artefact. Similar trends were also observed 16 for  $PM_1$  and  $PM_{10}$ , see Figure S2 in the Supporting Information.

17





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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.

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, 21<sup>st</sup>-24<sup>th</sup> 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<sub>2.5</sub> 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<sub>2.5</sub> 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 (21<sup>st</sup>-24<sup>th</sup> 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 22<sup>nd</sup>). 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.





Figure 2: Sensor ranking analysis for measured PM<sub>2.5</sub> 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  $0.22\pm0.13$  for PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> and PM<sub>10</sub> 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 PM<sub>1</sub> fraction, with the OPC-N2 and GRIMM in agreement but appearing to over-16 estimating the PM<sub>1</sub> mass concentrations with respect to the TSI 3330. While the OPC-N2 has 17 a higher particle size cut-off (0.38  $\mu$ m) compared to the TSI (0.3 $\mu$ m) and may explain the 18 observed difference in frequency distribution for PM<sub>1</sub> (Fig 1). While the TSI and GRIMM 19 have the same particle size cut-off (0.3  $\mu$ m), these instruments have been shown to disagree 20 (Fig S1) possibly due to different particle collection efficiencies.



Figure 4: Histogram of measured PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations by the TSI 3330, 4 5 GRIMM and median and inter-quartile values for the 14 OPC-N2. Note the different x and y 6 axis scales.

7 When the median and inter-quartile OPC-N2 concentrations were plotted against the TSI and 8 GRIMM concentrations, the slope was greater than unity for all three size fractions (Table 1) 9 indicating that the OPC-N2 were over-estimating the ambient particle mass concentrations

(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, r<sup>2</sup> is given in parenthesis.

	$PM_1$		PM <sub>2.5</sub>		$PM_{10}$	
OPC-N2	TSI	GRIMM	TSI	GRIMM	TSI	GRIMM
25 <sup>th</sup>	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 <sup>th</sup>	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<sub>2.5</sub> 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. 15<sup>th</sup> 12 September), pointing to a significant instrument artefact. On the 15<sup>th</sup> 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 PM<sub>1</sub> and PM<sub>10</sub> mass fractions (Fig S4, Supporting Information) with the OPC-N2 over-estimating the PM<sub>10</sub> concentration by several 16 orders of magnitude on 15<sup>th</sup> 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 function of ambient RH, with better agreement observed between the two instruments during 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

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- 13
- 14



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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.







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 PM<sub>10</sub> and PM<sub>2.5</sub> 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



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. 12-14<sup>th</sup> 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. 20<sup>th</sup> 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 12-14<sup>th</sup> 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 21 greater than unity for Arizona road dust but less than unity for salt and therefore suggest that 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 <sub>10</sub>			PM <sub>2.5</sub>				
		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**

18 Clearly there were times when there was a significant instrument artefact for the OPC-N2 19 (Figs 4 and S4) and the highest over-estimations occurred at high RH at both EROS and 20 Tyburn Rd (e.g. Fig 5 and 6). Whilst the accuracy of the instrument was significantly 21 worse at high RH the precision remains the same within error. The CV analysis 22 conducted in section 3.1.2 is repeated for the same dataset but put into low (RH<85%) 23 and high RH (RH>85%) subsets. For high RH conditions the CV for PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub>,

- was 0.34±0.30, 0.27±0.14 and 0.23±0.21, respectively. For low RH conditions the CV for
   PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub>, 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  $\kappa$ -Kohler theory (Petters and Kreidenweis, 2007), which describes the relationship 14 between particle hygroscopicity and volume by a single hygroscopicity parameter,  $\kappa$ . The  $\kappa$ -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  $\rho_w$  and 21  $\rho_{p}$ , respectively. The density of water is 1 g cm<sup>-3</sup>, and the bulk dry particle density is assumed 22 to be 1.65 g cm<sup>-3</sup>. The value for  $\kappa$  can be found by a non-linear curve fitting of a humidogram 23 24 (m/m<sub>o</sub> vs a<sub>w</sub>), 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 TEOM and OPC-N2 at RH less than 85%, the scaling factors shown in Table 2 are used 27 28 calibrate the dry mass of the OPC-N2 to that observed in the TEOM, both in the PM<sub>2.5</sub> and 29 PM<sub>10</sub> fractions.

30

31 Figure 8 shows the humidogram plots, for both the  $PM_{2.5}$  and  $PM_{10}$  fractions, obtained by

32 plotting the ratio of OPC-N2 to the reference instrument (TEOM and GRIMM) outputs

33 versus ambient RH. Ideally, a measure of RH internal to the instrument could be made to



10









Figure 8: Measured and fitted humidograms ( $m/m_o$  vs RH) recorded at the Tyburn Road AURN site for  $PM_{10}$  and  $PM_{2.5}$  size fractions and reference instruments (TEOM and

GRIMM). The dry mass (m0) is given by the TEOM or GRIMM and the humidified mass is
 given by the OPC-N2. Measured data is given by the black circles, the fitted data is given by
 the blue (TEOM-FDMS) and red (GRIMM) line.

4

5 We then applied this fitting constant to model the expected OPC/Reference instrument ratio 6 for a given RH as a result of particle hygroscopic growth, by re-arranging Equation 5:

7

$$8 \qquad \frac{m}{m_o} = 1 + \frac{\frac{\rho_W}{\rho_p}}{\frac{1}{-1 + \frac{1}{a_W}}} \tag{6}$$

9

Where the  $m/m_0$  is the ratio of the OPC-N2 to the reference instruments. Using Equation 6, 10 11 the mass concentrations measured by the OPC-N2 were corrected and significantly better 12 agreement between the corrected OPC-N2 and reference instruments was observed for 13 measurements across the whole range of ambient RH (Tables 2 and 3). Overall, the corrected 14 OPC-N2 mass concentrations using Eqn 6 were notably better, within 33% and 52% of the 15 TEOM and GRIMM, respectively (Table 3) compared to 250-400% without the correction 16 factor (Table 2). The time series for the corrected data is shown in Figures S7 and S8 17 (Supporting Information) and there are periods were there is good agreement between TEOM 18 and the corrected OPC-N2. However, it was also evident from Table 3 that the slope was 19 different for PM<sub>10</sub> and PM<sub>2.5</sub> mass fractions for all OPC-N2 when compared to the TEOM, 20 and suggests that differing responses for the OPC-N2 to two size fractions. This may be 21 related to the observed variation in  $\kappa$  between the size fractions relative to the TEOM or an 22 unaccounted loss mechanism; the exact cause will be investigated further in future work.

23

There were also times when the OPC-N2 were clearly over-corrected (e.g. from 20<sup>th</sup> February 24 25 onwards), generally when the ambient RH was low (Fig 6). This suggests that when the RH 26 was below a threshold, Eqn 6 overcorrects the data and this can be observed in the 27 humidograms shown in Figure 8. Typically, at RH <85% the hygroscopic growth of real 28 atmospheric aerosols is small and it may be more appropriate to apply a linear regression 29 correction factor for data recorded under these RH conditions. Therefore we applied a binary 30 two model approach to correct the OPC-N2 mass concentrations, where a linear correction 31 (using the TEOM as reference concentration) for when RH <85%, and above this threshold in

1 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 2 3 correction with Eqn 6 for all RH. What was noticeable was that the intercept for the two 4 model approach moved closer to zero, suggesting that at the lower mass concentrations the correction was improved. Similar trends were also observed for PM<sub>10</sub>. Also during the period 5 from the 20<sup>th</sup> February, the volatile particle fraction was also lower (Fig 6) and this indicates 6 a significantly different aerosol composition. Since  $\kappa$  is composition dependent, a single 7 8 global fit to  $\kappa$  will result in poor fitting when the true  $\kappa$  is significantly different to the 9 average  $\kappa$ . The preceding discussion suggests that further refinement to the correction factors 10 applied to the OPC-N2 is possible, depending on the ambient RH and better knowledge of 11 aerosol composition. RH measurement is relatively trivial and can be achieved with small 12 sensors but aerosol composition determination still requires significant analytical equipment 13 and expertise.

14

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

OPC-N2	TEOM		GRIMM		
	<i>PM</i> <sub>2.5</sub>	<i>PM</i> <sub>10</sub>	<i>PM</i> <sub>2.5</sub>	$PM_{10}$	
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	

16 reference instruments. Intercepts were not constrained to zero.

17

18 **3.3.1 Longer-term monitoring with OPC-N2 at EROS** 

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- 21





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Figure 9: Histogram of measured PM<sub>2.5</sub> 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
(A), while the right plot (B) shows the RH corrected OPC-N2 concentrations.

5

After the conclusion of the intensive measurements at EROS (Section 3.1), five of the OPC-6 7 N2 continued monitoring for a further 4 months to examine if there was any evidence of instrument drift over time, along with the GRIMM as reference. One of the OPC-N2 failed in 8 9 December, and so was excluded from this analysis. The remaining four OPC-N2 were 10 compared to GRIMM and in January after running for 4 months (Fig 9A), and while three of 11 the OPC-N2 had a similar distribution to the GRIMM (OPC12, 13 and 14), OPC9 appeared 12 to show evidence for instrument drift as the mode has shifted relative to the GRIMM. 13 However, the increased frequency of higher mass concentrations not observed by the 14 GRIMM but by all four OPC-N2 (Fig 9A) suggests that ambient RH is also a factor, as the 15 average RH in January (91%) was higher than September (84%). Therefore, we calculated the correction for RH as described in the previous section (Eqn 6), as changes in aerosol 16 17 composition would affect the particle hygroscopicity. In addition, the  $\kappa$  was only fitted for the 18 data with RH < 95% since the hygroscopicity of aerosol is highly sensitive to any error in the 19 RH measurement above this value. Application of the RH correction factor resulted in better 20 agreement between each of the OPC-N2, with similar corrected distributions observed (Fig 21 9B). Furthermore, the corrected OPC-N2 concentrations also had better agreement with the 22 GRIMM during January (Fig 9B) compared to uncorrected concentrations (Fig 9A), 23 suggesting that changes in the particle water content were the cause. Thus, at least over a four 24 month measurement period, there appears to be no evidence for instrument drift in the OPC-25 N2, once appropriate correction factors were applied.

#### **3.4 Discussion on the OPC-N2 interferences**

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

1 water content as the cause. This result is perhaps not surprising, as many studies in the 2 literature have shown that particle water content can be a major reason for discrepancies 3 between techniques that measure ambient particle mass (See e.g. (Charron et al., 2004)). The 4 use of  $\kappa$ -Kohler theory to derive a correction factor based on ambient RH improved the 5 agreement between the OPC-N2 and reference instruments; however a limitation of this 6 approach is that the bulk aerosol hygroscopicity is related to particle composition, typically 7 the inorganic fraction (e.g. (Gysel et al., 2007)). Variation in ambient particle composition 8 could account for the large spread observed in the ratio of OPC-N2/TEOM at high RH (Fig 9 8), as an average hygroscopicity correction will overestimate when PM with higher hygroscopicity is measured and vice versa. This would have potentially significant 10 11 implications when using the OPC-N2 for longer-term monitoring, as the  $\kappa$  value may not be 12 constant over the monitoring period. Therefore, this would suggest the need for regular 13 calibrations to account for changes in bulk aerosol composition and as a result  $\kappa$  values. 14 Furthermore, Eqn 6 may not be required for locations where the ambient RH is lower than 15 85%, as typically atmospheric particle growth due to water below this threshold is limited 16 and a simple linear regression may be sufficient. Thus, in-situ and seasonally specific 17 calibrations for the OPC-N2 are required to account for possible differences in ambient 18 aerosol properties. However as  $\kappa$  values for continental regions tend to fall within a narrow 19 range globally  $(0.3\pm0.1)$ , (Andreae and Rosenfeld, 2008), with some systematic deviations for 20 certain regions (Pringle et al., 2010), this average  $\kappa$  value could be used in lieu of calibration 21 with reference instrument (e.g. a TEOM) to determine the correction factor (C) according to 22 Eqn 7:

23

24 
$$C = 1 + \frac{0.3/1.65}{-1 + \frac{1}{a_W}}$$
 (7)

25

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  $\kappa$  value from the literature in Eqn 7 may be a reasonable approximation.

#### 1 4.0 Applicability of OPC-N2 for ambient monitoring

2 The Alphasense OPC-N2 was evaluated for use in ambient monitoring of airborne particle 3 mass concentration, with TEOM-FDMS and two commercial optical light scattering 4 instruments; GRIMM PAS 1.108 and TSI 3330 employed as reference instruments. 5 Comparison of the OPC-N2 to the reference optical instruments demonstrated reasonable 6 agreement for a low-cost sensor to the measured mass concentrations of PM<sub>1</sub>, PM<sub>2.5</sub> and 7 PM<sub>10</sub> as evidenced by the stated accuracy and precision However, the OPC-N2 demonstrated 8 a significant large positive artefact in measured particle mass during times of high ambient 9 RH, and a calibration factor was developed based on bulk particle aerosol hygroscopicity. 10 Application of the RH correction factor, based upon K-Kohler theory, resulted in notable 11 improvement with the corrected OPC-N2 measurements within 33% of a TEOM-FDMS. 12 While higher than the slope of 1±0.1 allowed by the US EPA, it is comparable to the 13 agreement of a GRIMM to the TEOM (20%). All low cost PM sensors will likely require 14 calibration factors to obtain the dry particle weight unless they actively dry the PM 15 containing air stream before it enters the device. The use of heated inlets could be used to 16 reduce the RH in the air stream but would have consequences on the power requirements of 17 the sensor, potentially making them less attractive for battery led operation. Thus it highlights 18 that the OPC-N2 does not respond the same as reference instruments to ambient particle 19 mass, but provided appropriate correction factors are applied, reasonable agreement with 20 OPC-N2 to reference instruments can be achieved. Furthermore, the dependence of the OPC-21 N2 on a correction for RH and  $\kappa$  may limit its application for longer-term monitoring as the  $\kappa$ value may change over time, and will be the focus of future work. This is especially salient 22 23 when considering using the OPC-N2 to compare to air quality standards that are one-year 24 averages of  $PM_{2.5}$  and  $PM_{10}$ .

25

For PM<sub>10</sub> 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. One out of four OPC-N2 tested for long-term monitoring appeared to show evidence for instrument drift relative to reference instruments.

32 Overall, the OPC-N2 have been shown to accurately measure ambient airborne particle mass 33 concentration provided they are correctly calibrated and corrected for RH. The reasonable

- 1 level of precision demonstrated between multiple OPC-N2 suggests that they would be
- 2 suitable for applications where a number of instruments are required such as spatial mapping
- 3 and personal exposure studies.

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- 9

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