We thank the reviewers for their comments on our manuscript. Please find our detailed responses to the comments from both reviewers below in red.

Response to Reviewer 1

General Comments: This manuscript suggests a correction, (previously determined and published elsewhere), may be applied to commercial low-cost optical particle counter measurements that enable a direct comparison to PM2.5 measurements at high relative humilities in the field.

It is true that both Crilley et al. (2018) and Di Antonio et al. (2018) have previously provided correction methodologies, but these were for specific locations with specific meteorology. It is important to note that a universal correction is not possible for individual particles due to differences in composition (hence hygroscopicity), size and shape. Instead, if it is possible to correct for an ensemble of aerosols, using average hygroscopicity, this would make it significantly easier to correct low-cost optical particle counters without having to employ detailed calibrations on the particle size distribution. Therefore, the main aim of this paper is to investigate whether a universal correction method can be reasonably applied to reported particle mass concentrations by low-cost OPC across a wide variety of locations. By achieving this aim (with associated errors), we believe this paper of significant use to the AMT readership. To clarify the aims, we have added the following sentence to the end of the introduction:

"The aim of this paper was to investigate whether a universal correction method can be reasonably applied to reported particle mass concentrations by low-cost OPC across a wide variety of locations."

Data is presented from an alphasense OPC-N2 and a TEOM-FDMS at five field locations. Whereas the OPC-N2 measures ambient (wet) aerosol size and number, which assumes a uniform particle density and reports the wet aerosol mass concentration, the TEOM-FDMS measures dry aerosol mass concentration

The authors argue that hydroscopicity of the aerosol can be calculated from the difference in wet and dry particle mass using the OPC-N2 measurement for wet particle mass and the TEOM-FDMS measurement for dry particle mass measurement. It is difficult to accept the authors' conclusions without a clearer outline of several key assumptions.

First, the argument hinges on the idea that the OPC-N2 and the TEOM-FDMS instruments report the same aerosol size distribution in a controlled setting with low relative humidity; however this assumption is neither explicitly stated nor validated.

Previous work has shown that a calibration for the reported OPC-N2 particle mass concentrations (as well as other low-cost OPC) is often needed with respect to reference instrumentation (e.g. TEOM-

FDMS). This has been investigated in depth both in the field (see e.g. Crilley et al, Di Antonio et al, Pope et al. and in the lab (e.g. Sousan et al. 2016).

As mentioned by the reviewer, there are several assumptions made in relating the OPC and TEOM measurements to each other. These assumptions include particle sphericity, density and size distribution. Firstly, we are using the reported particle mass measurements by the OPC-N2 throughout. In the conversion from particle number distribution, it is assumed that the particles are all spherical with a uniform density across the particle size distribution measured by the OPC-N2. In addition, it is also assumed that the particle mass below the OPC-N2 particle size cut-off (300 nm) is inconsequential. The key assumption made is that both the OPC-N2 and TEOM are responding to the same dry aerosol mass for particles below the cut-off for the TEOM-FDMS (PM2.5). The work by Di Antonio et al (2018) demonstrated that with correction, the OPC-N2 can report the same dry aerosol size distribution as reference instrument.

To clarify these assumptions, the following text has been added to section 2.3:

"To calculate the particle mass concentration from measured particle number size distribution, spherical particles of a uniform density and shape are assumed by the OPC, which is not strictly true for airborne particles in an urban atmosphere but is considered a standard approximation. For full details see Crilley et al. (2018). The OPC-N2 assumes the ambient particle density to be 1.65 g cm-3 across all size bins to derive the particle mass concentrations from the measured particle number concentrations (Crilley et al., 2018), therefore we have used this density for the dry particles (ρp) in Eq. (2). We assume that the particle density is uniform across the particle size distribution measured by the OPC-N2. Furthermore, we assume that both the OPC-N2 and reference instrument are responding to same dry aerosol mass for the all particles below the size cut-off on the reference instrument. We also note that we assume both the OPC-N2 and reference instrument responses are linear over the range of measured concentrations at each site."

Although the authors argue that the use of a dryer in front of the OPC-N2 measurement would render the low-cost OPC more expensive, such a lab-based comparison would substantially bolster their argument.

It is true that lab-based study to exploring the response of the OPC-N2 fitted with a dryer before the inlet would be useful, but this is outside the scope of the current work. We aimed to explore the universal applicability of the correction factor by using a number of field datasets. We do note that previous work by Sousan et al (2016) investigated the response of the OPC-N2 under low RH in a controlled laboratory setting and found good agreement in reported mass concentrations with GRIMM PAS-1.108 (r2 > 0.97) but the slopes varied depending on the aerosol composition. This would suggest that drying the aerosol before measurement by the OPC-N2 could improve the accuracy, but it would not negate the need for in-situ calibration.

It is disconcerting that the OPC-N2 and reference measurements are correlated but do not agree (1:1) at low humidity (e.g. <60%; Fig. 3). The reason for this disagreement is not clearly articulated.

This is likely due to the previously stated differences in density, composition, size and shape of individual particles. Some particle-phase chemical species are more likely to be found in high concentrations at different times of the day due to changes in sources and sinks. This is demonstrated by recent high temporal aerosol composition measurements using an aerosol mass spectrometer (AMS) in urban locations where a distinct diurnal trend in bulk aerosol composition (e.g. ammonium, sulphates, nitrates, organics etc) was observed, see e.g. Young et al 2014.

In addition, the authors do not discuss the fact that field measurements from different times of day and days likely have different compositions, which leads to large scatter in observed PM2.5 vs. relative humidity (e.g. Fig. 1 and Fig. 2) based on wind velocity and source.

The reviewer makes a good point. Therefore, we have added the following sentence to the end of section 3.1 to clarify this.

"The scatter in OPC/TEOM observed in Fig 2 as function of RH was likely due to temporal variability in aerosol composition due to changing sources and sinks (both local and regional)."

Finally, assumptions of spherical particles and refractive index required to calculate particle size from OPC light scattering measurements should also be discussed and are currently not mentioned.

We have included text outlining these assumptions in Section 2.3, please see our earlier response.

I think that this paper requires major revisions.

Specific Comments:

Table 1) Show range and mean +/- SD

We have made these changes to Table 1, as shown below

Table 1: Summary of measurement datasets. Reported OPC-N2 $PM_{2.5}$ mass concentrations are uncorrected. For the Nicaragua measurements there was no co-located reference instrumentation.

Only one 24 hr average gravimetric PM$_{2.5}$ concentration was available for Nairobi, presented with stated measurement uncertainty.

Site	**Date**		**RH (%)**	**OPC-N2 PM$_{2.5}$ (µg m^{-3})**	**Reference PM$_{2.5}$ (µg m^{-3})**
Birmingham	Oct 2016-Feb 2017	Average ± SD	89±10	38 ± 54	11 ± 8
		Range	44 - 99	0.3 - 566	0.5 - 63
Beijing	Dec 2016	Average ± SD	36±15	74±82	75±66
		Range	13 - 81	3 - 274	2.7 - 208
Delhi	Jan-Feb 2018	Average ± SD	59±25	218±228	168±76
		Range	9 - 100	12 - 1113	50 - 478
Nairobi	Feb-Mar 2017	Average ± SD	51±18	32±16	27.6±6.8
		Range	16 - 89	4 - 135	
Nicaragua	Feb-Dec 2017	Average ± SD	77±11	21±55	NA
		Range	39 - 91	0.5 - 742	

Table 2) should have the same format as Table 1, with the addition of ratios to PM2.5

We do not understand what the reviewer is asking for here, as Table 2 reports slopes and consequently have not made any changes to Table 2.

Figure 1 and Figure 2. Reasons for high variability in measured PM2.5 with RH should be mentioned.

Please see our response to earlier comment.

Figure 3. Show 1-1 line and make the aspect ratio of figures 1:1.

These changes have been made.

Figure 4. Should this data be presented considering suspected decline in OPC-N2 performance? If so, how much of this data should be presented and how will that be determined?

We have re-evaluated the data and we now do not feel there is enough evidence to suggest a decline in performance. As pointed out by reviewer 2, just because there appears to be change in amplitude in the reported PM2.5 mass concentration in the last 2 days (Fig 4), this does not mean that there is a decline in performance. The time series in Fig 4 is not long enough to ascertain this, and there we have removed this discussion on suspected declining performance from the text and will show the whole dataset.

The paragraph now reads:

"During the measurements in Delhi, the OPC-N2 typically over-reported the PM2.5 mass concentrations relative to the reference (Fig. 6A). The OPC-N2 assumes a uniform particle density of 1.65 g cm-3 in the particle counts to mass conversion, and this density may be inappropriate for Delhi aerosol during winter. Previous measurements of aerosol density during winter in Delhi at midday were on average 1.28±0.12 g cm-3 (Sarangi et al., 2016), lower than applied by the OPC-N2. Generally, the OPC/TEOM ratio was below 1 (Fig 2A) and so this would not explain this discrepancy. From Fig. 6A, higher PM2.5 mass concentrations relative to the reference were reported by the OPC-N2 towards the start of the measurement period (Fig. 6A), generally during the times when the RH was high (e.g. 25-29th Jan, Fig. 6B). We also note that the reported concentrations by the OPC-N2 towards the end of the measurements in Delhi (11th Feb onwards, Fig. 6A), were in better agreement with reference. The cause of this change in performance is unclear but could reflect lower RH or changes in aerosol composition"

Response to Reviewer 2 (Don Collins)

This manuscript describes the use of data collected with a low cost optical particle counter together with reference grade PM instruments to assess overall accuracy and, especially, the sensitivity to relative humidity. The manuscript is reasonably well written and understandable, though some minor editing would be required prior to publication. There is currently considerable interest in the use and performance of low cost air quality sensors and also considerable need for establishment of best practices for operation and data analysis. As noted in the manuscript, though only an Alphasense

OPC-N2 was used for this analysis, the findings can at least qualitatively be extended to the array of similar low cost PM sensors that rely on particle light scattering for measurement. Perhaps simply because of the advantage of hindsight, there are changes in techniques and instrumentation that could have provided a more easily interpreted dataset. The use of a mixture of reference instruments and the rather narrow range in humidity encountered at each of the study sites somewhat limits confidence and extension of the results. But despite the limitations of the dataset, the results would still be valuable to others using these or similar low-cost sensors and the manuscript should be publishable after the concerns identified below are addressed.

We thank the reviewer for their positive comments.

We agree that examining different low-cost OPC would have been useful but were limited by available instrumentation during the respective field campaigns. We would like to note that we primarily used the TEOM-FDMS as reference instrument at the sites of primary interest in the manuscript (Beijing, Delhi and Birmingham Tyburn).

Section 2.2: There needs to be some discussion of the relationship between the reported ambient relative humidity and that in the sensor. Were the OPC-N2's inside some sort of enclosure? And if so, how was its temperature related to that outside? Would solar heating impact the temperature during the daytime? And if the sensor and outdoor temperatures are not always the same, could that help explain the large spread in derived PM2.5 at high RH mentioned on page 7, line 3?

Yes, the OPC-N2 were housed inside a small enclosure that was placed outside the main laboratory at all sites. As such we expect the internal temperature of OPC-N2 to be similar to ambient, but we do not have any measurements to support this.

Section 2.2: Details of the sites and reference instruments at each should be provided in a table. As is, the descriptions are structured differently enough that it is difficult for a reader to appreciate the similarities and contrasts among the sites.

We have added the following table to section 2 as a summary of the different sites in the manuscript.

Table 1: Summary of the measurement sites. Full details available in the text. N/A signifies not available. Custom housing for the OPC-N2 as per description in text.

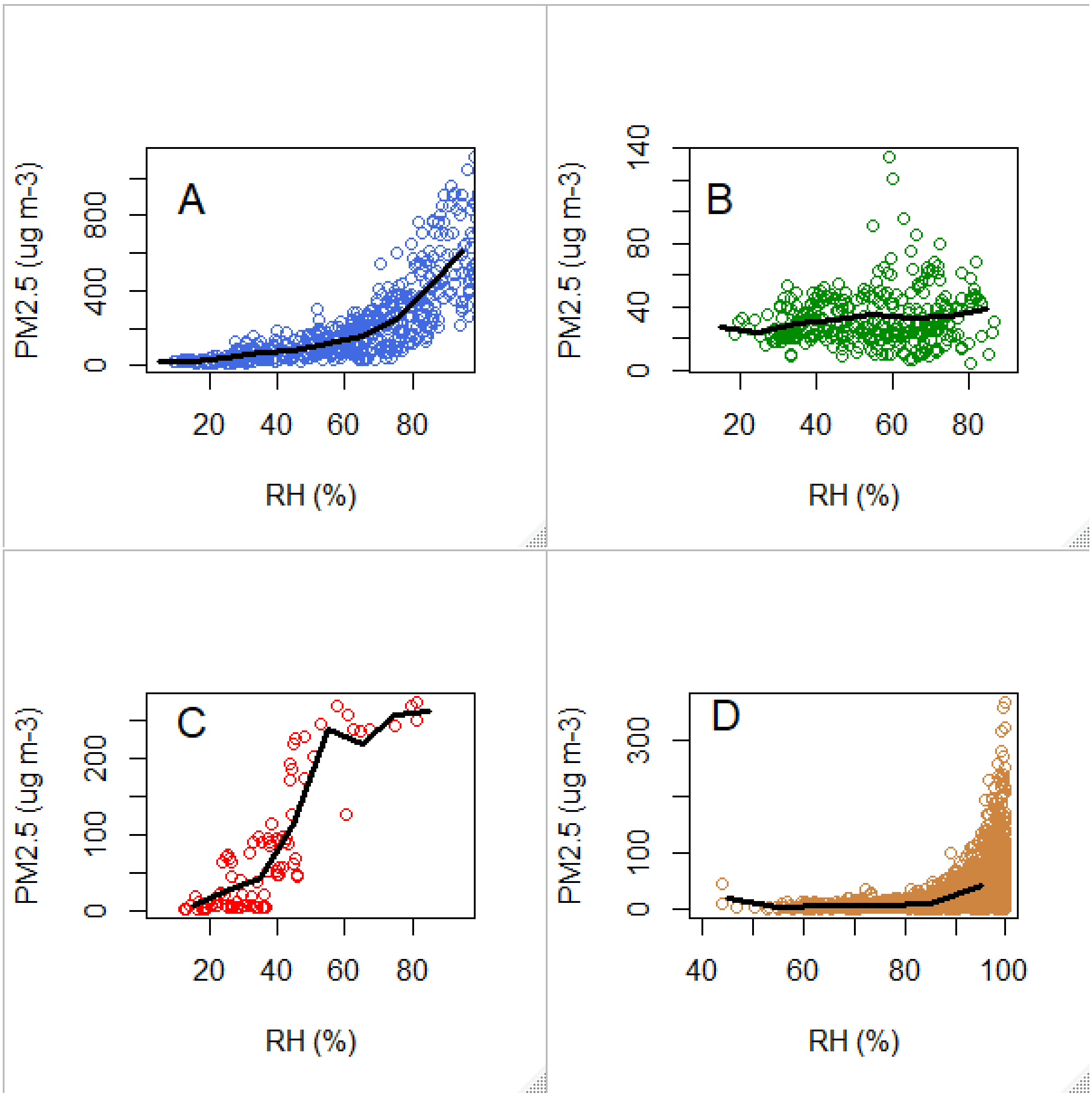
Location	Site Description	OPC-N2 housing	Reference instrument	Aerosol composition instrument
Birmingham BAQS, UK	Urban background	Custom	GRIMM	N/A
Birmingham Tyburn, UK	Urban background	Custom	TEOM-FDMS	N/A
Beijing, China	Urban background	Custom	TEOM-FDMS	AMS
Delhi, India	Urban background	Custom	TEOM-FDMS	ACSM
Nairobi, Kenya	Urban background	Custom	Gravimetric	N/A
Masaya volcano, Nicaragua	Volcano	AQMESH	N/A	N/A

Section 3.1: The relationship between RH and composition for both Delhi and Beijing should be discussed and possibly graphed. The assertions that “there is clear influence of RH on the measurements performed in Delhi” and that the “stepwise increase in the derived particle mass between a RH of 40-50% RH may point to deliquescence” in Beijing should be made only after quantitatively describing any role of RH-dependent variation in composition (due to connections with things such as wind patterns and photochemical production).

Later in the manuscript, we state that explore the relationship RH, aerosol composition and OPC-N2 reported mass concentrations in section 3.2 for Delhi and Beijing, as shown in Figs 3 and 4.

Figure 1: It would help to include curves representing the mean or median PM2.5 vs RH

We think that including curves of the mean PM2.5 vs RH could be misleading as we want to highlight the range of concentrations measured as function of RH. We have therefore added the mean reported PM2.5 mass concentration binned by RH to figures in supporting information, as shown below.



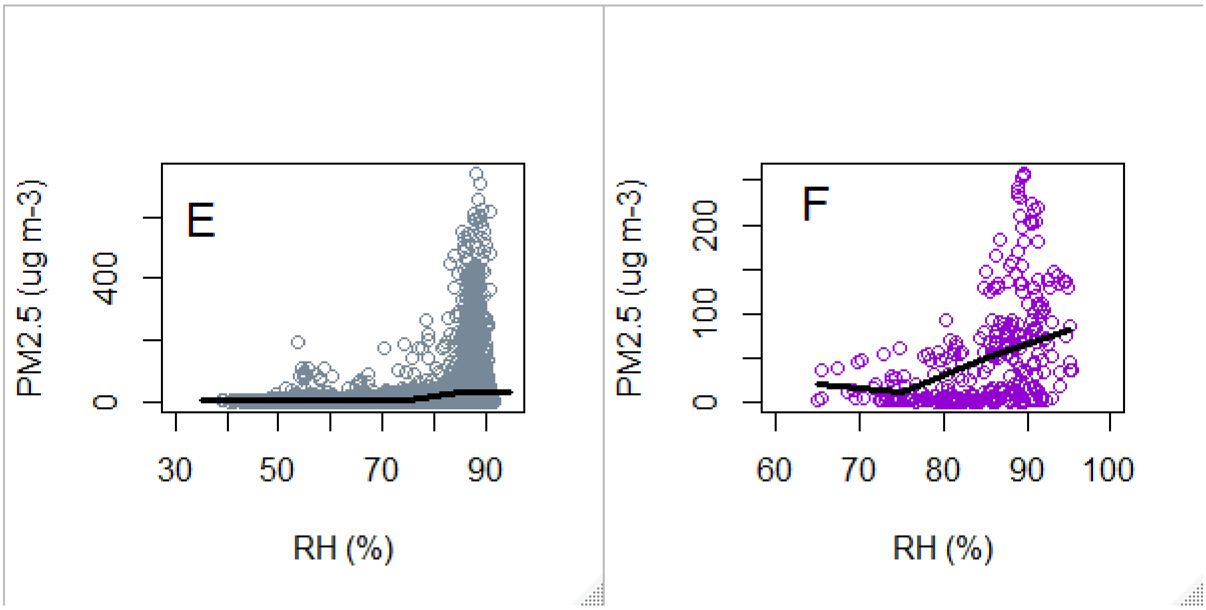


Figure S1: Plot of reported PM2.5 mass concentration by OPC-N2 against ambient RH for the whole measurement period in Delhi (A), Nairobi (B), Beijing (C), Bham BAQS (D), Nicaragua (E), Bham Tyburn (F). The black line is the mean reported PM2.5 mass concentration binned by RH. Note the different y- and x-axis scales.

Figure 2: There is a brief mention in the text that the OPC-N2 tends to underestimate PM2.5 at low RH, which partially explains the «1 OPC/TEOM ratios in Beijing (it is hard to say for the Birmingham sites because of the wide y-axis range used). But I'd like to know whether the authors have an explanation for the apparently strong dependence of the ratio on RH for RH < 40% where hygroscopic growth is probably not significant enough to be responsible. Could this be related to a confounding relationship between RH and composition?

The reviewer asks some pertinent questions. It should be noted that aerosol with high hygroscopicity, typically those with high mass ratios of inorganic salts, can take up water significantly at RHs less than 50%, for example sulphates and nitrates. However, we cannot rule out correlations between RH and composition that are not related to hygroscopicity. It is possible that the low RH correlates with low particle concentrations independent of hygroscopicity.

Figure 2: Clarify in the caption that the k fits are shown in color.

We have changed the caption to read:

*"**Figure 2:** Humidograms with corresponding κ fit (shown in colour) for Delhi (A), Beijing (B), Bham BAQS (C), Bham Tyburn (D). Note the different y-axis and x-axis scales. The two-stage correction factor described in Section 3.3.1 has not been applied for these humidograms."*

Page 11, line 14: It is not necessarily true that any enhancement at RH < 79% is not due to ammonium sulfate because the particles may be in a metastable state.

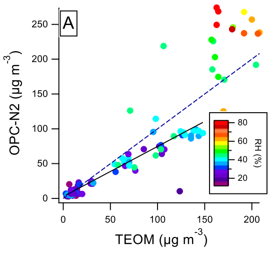
The reviewer is correct that ammonium sulphate could be in a metastable state and hence not efflorescent. We have rephrased the passage to reflect this:

“From Figs. 3 and 4, it appears that this effect was occurring at RH above 40 %, below the deliquescence point of ammonium sulphate (79 %) indicating the ammonium sulphate component of the aerosol was in a metastable state. Aerosols with multi-component mixtures are observed to deliquesce earlier than the deliquescence points of the individual components e.g. Pope et al. (2010). It is noted that the nitrate component of the aerosols have a smoother continual take up of water with respect to RH (Gibson et al., 2006;Hu et al., 2010).”

Figure 3b: The sulfate content should be presented as a fraction and not as an absolute concentration. It is not surprising that sulfate mass concentration increases with increasing total (TEOM) concentration. But that is unimportant for the OPC-N2 vs. TEOM comparison and only the fractional contribution matters.

We note that the sulphate concentrations were obtained using an aerosol mass spectrometer, which measures PM1, while the TEOM measures PM2.5. therefore, this fraction is only an estimate as a substantial fraction of sulphate (PM1-2.5) missing.

But the reviewer has made a valid point and we have made the change to Fig 3b as shown below and updated the text to reflect this change



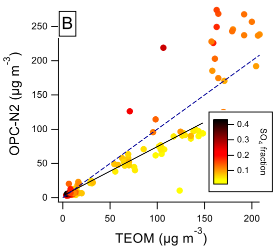


Figure 3: Reported OPC-N2 uncorrected PM2.5 mass concentrations against TEOM PM2.5 mass concentration measurements coloured by ambient RH (A) and fraction of sulphate to the total PM2.5 mass (B) in Beijing. The straight line indicates the linear regression fit for concentrations below 150 µg m-3. The dashed line is 1:1.

Section 3.3: The manuscript suggests that the OPC-N2-based PM2.5 is higher than the reference and that using the measured density of 1.28 rather than the assumed density of 1.65 would not explain the discrepancy. Why not? It seems this change would improve agreement, whereas the manuscript seems to suggest it would not.

While there are periods where the reported concentration by OPC-N2 was considerably higher than by the TEOM (reference) in Delhi, generally the ratio of OPC/TEOM was below 1, as shown in Figure 2A. Therefore, using a lower particle density would not improve the agreement between the OPC and reference instrument. To clarify this point, we have changed the text to read:

"Previous measurements of aerosol density during winter in Delhi at midday were on average 1.28±0.12 g cm-3 (Sarangi et al., 2016), which is lower than that applied by the OPC-N2 (1.65). Generally, the OPC/TEOM ratio was below 1 (Fig 2A) and so this would not fully explain this discrepancy."

Page 13, line 9: What does "lose structure" mean here? The amplitudes of the daily peaks are lower than earlier in the measurement period, but why should that indicate there is a problem? This is especially important because it is the basis for the suggestion that the lifetime of the sensor in Delhi is only a month. And furthermore, even if the sensitivity of the OPC-N2 decreased due to dirty optics, the authors should not attempt to extrapolate to all low cost sensors as they do here.

The reviewer has made a valid point and we agree that we do not have enough evidence to suggest that there was a problem with sensor. Consequently, we have edited this paragraph to remove this discussion.

Please see our response to the last comment from Reviewer 1.

Page 14, line 11: Does "though within a large range" refer to 0.1 – 0.12? If so, I don't think of that as a large range. And if not, reword this so it is clearer.

We were referring to the large scatter OPC/TEOM in Fig 2C and not that calculated kappa values.

To clarify this point, we have changed the text to read:

"The calculated κ value for each month in Birmingham had little variability (0.1-0.12) from October to January and this suggests that on average the bulk hygroscopicity of the aerosols was consistent, though within a large range (as indicated by the large spread in OPC/TEOM in Fig 2c)."

Section 3.4, first paragraph: Rather than relying on trajectories, it seems statistical tools or machine learning could provide at least some improvement in correction accuracy using things like weather observations, day of the week, time of day…

We applied this method of back trajectories as a proxy for particle composition (and hence hygroscopicity) as it was successfully applied by Di Antonio et al. (2018) for their dataset, to see if could also be applied to our dataset. Statistical tools may be able to improve the correction accuracy but are outside the scope of this work.

Page 16: Use a word other than global, which implies something based on more than three urban background sites during only relatively short periods of the year.

We chose to refer to this calculated kappa value as global as it covered urban background sites across 2 continents. The reviewer does make a valid point, and we have added the following sentence to clarify:

"From Fig. 8, a κ of 0.33 (assuming a uniform particle density of 1.65 for all sites) was calculated, slightly higher than the average of 0.3 suggested for continental regions (Pringle et al., 2010). Consequently, we refer to the calculated κ from Fig 8 as global κ but note that it was calculated from 3 urban background sites on two continents. We applied this κ (0.33) to correct the OPC-N2 data at all sites, as well the average for continental regions (0.3), with the results summarised in Table 3.*"*

References

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Effect of aerosol composition on the performance of low-cost optical particle counter correction factors

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Abstract. There is considerable interest in using low-cost optical particle counters (OPC) to supplement existing routine air quality networks that monitor particle mass concentrations. In order to do this, low-cost OPC data needs to be cross-comparable with particle mass reference instrumentation, and as yet, there is no widely agreed methodology. Aerosol hygroscopicity is known to be a key parameter to consider when correcting particle mass concentrations derived from a low-cost OPC, particularly at high ambient Relative Humidity (RH). Correction factors have been developed that apply κ-Köhler theory to correct for the influence of water uptake by hygroscopic aerosols. We have used datasets of co-located reference particle measurements and a low-cost OPC (OPC-N2, Alphasense), collected in four cities in three continents, to explore the performance of this correction factor. We report evidence that the elevated particle mass concentrations, reported by the low-cost OPC relative to reference instrumentation, is due to bulk aerosol hygroscopicity under different RH conditions, which is determined by aerosol composition and in particular the levels of hygroscopic aerosols (sulphate and nitrate). We exploit measurements made in volcanic plumes in Nicaragua, that are predominantly composed of sulphate aerosol, as a natural experiment to demonstrate this behaviour in the ambient atmosphere, with the observed humidogram closely resembling the calculated pure sulphuric acid humidogram. The results indicate that the particle mass concentrations derived from low-cost OPCs during periods of high RH (> 60 %) need to be corrected for aerosol hygroscopic growth. We employed a correction factor based on κ-Köhler theory and observed corrected OPC-N2 $PM_{2.5}$ mass concentrations to be within 33% of reference measurements at all sites. The results indicated that an *in situ* derived κ (using suitable reference instrumentation) would lead to the most accurate correction relative to co-located reference instruments. Applying literature κ in the correction factor also resulted in improved performance of OPC-N2, to be within 50 % of reference. Therefore, for areas where suitable reference

instrumentation for developing a local correction factor is lacking, using a literature κ value can result in a reasonable correction. For locations with low levels of hygroscopic aerosols and RH, a simple calibration against gravimetric measurements (using suitable reference instrumentation) would likely be sufficient. Whilst this study generated correction factors specific for the Alphasense OPC-N2 sensor, the calibration methodology developed is likely amenable to other low cost PM sensors.

1.0 Introduction

Advances in miniaturization technology have led to the development of many different kinds of low-cost air pollution sensors, ranging from passive gas samplers to miniaturized versions of reference instruments (Lewis et al., 2018;Jayaratne et al., 2018). The term low-cost is relative and typically refers to the sensor being at least an order of magnitude cheaper than corresponding reference instrumentation (Lewis et al., 2018). Monitoring of key air pollutants (e.g. $PM_{2.5}$, NO_x and O_3) has traditionally been performed via reference standard or equivalent monitors at fixed monitoring stations. However, this approach can lack the necessary spatial coverage to properly assess personal exposure due to significant spatial heterogeneity in the concentration of air pollutants in urban areas (de Nazelle et al., 2017). Low-cost sensors are an attractive option due to their reduced costs and portability, making them viable for mobile or highly spatially resolved measurements, to complement existing monitoring frameworks. This has led to low-cost sensors becoming a common feature of an increasing number of air pollution monitoring operations (Snyder et al., 2013;Morawska et al., 2018).

The trade-off with using low-cost sensors are that they are currently not as accurate, precise, selective or sensitive when compared to research or regulatory grade instrumentation (Mead et al., 2013;Lewis et al., 2018;Lewis et al., 2016;Smith et al., 2017;Crilley et al., 2018;Borrego et al., 2016;Popoola et al., 2016). Consequently, low-cost sensors of air pollutants need to be carefully characterised to ensure they meet the specific requirements of the intended application (Castell et al., 2017). In their review, Morawska et al. (2018) concluded that low-cost sensors were fit for purpose for many applications, such as supplementing routine air quality measurements and engaging the public and community groups. However, there is still work needed if low-cost sensors are to be used for accurate exposure measurements, or in the future for compliance monitoring, which is of particular interest in under-monitored low and middle-income countries (LMICs). LMICs typically have high urban air pollution yet the resources and infrastructure are sometimes lacking to support continuous classical reference air quality measurements (Pope et al., 2018). One of the challenges with using low-cost sensors in this setting is there is currently no agreed methodology for the evaluation of their accuracy and precision and their subsequent calibration (Lewis et al., 2018).

A key pollutant for air monitoring networks is airborne particulate matter (PM) due to their well-established detrimental physical health effects (Cohen et al., 2005;Landrigan et al., 2018). In particular, exposure to fine particles ($PM_{2.5}$, particles with an aerodynamic diameter less than 2.5 µm) is known to have multiple disease pathways (Landrigan et al., 2018). Recently, short term exposure to $PM_{2.5}$ has been linked to short term cognitive decline (Shehab and Pope, 2019). $PM_{2.5}$ mass regulatory limits are based on the dry particle mass concentration and regulatory-grade particle mass instrumentation dry the aerosol before measurement to record the dry aerosol mass concentrations. Low-cost optical particle counters (OPC) measure particle diameter and number concentrations by light scattering and convert this to particle mass concentrations by assuming particle sphericity and a uniform density. Low-cost OPCs typically do not dry the aerosol before measurement, and this can result in over-estimation of the dry particle size (compared to that which would be determined after drying) under high RH conditions (Crilley et al., 2018;Jayaratne et al., 2018;Di Antonio et al., 2018), thought to be related to the uptake of water by hygroscopic aerosol. Consequently, the reported PM mass concentrations by low-cost OPC are of the wet particle mass concentration and these need to be converted to a dry particle mass concentration in order to have comparability with regulatory standards and reference instrumentation. One solution to measuring the dry mass of particles would be to add a pre-conditioning drying step prior to the OPC, but this would result in higher costs, greater power consumption and less instrument portability thereby reducing the unique selling points (USPs) of low-cost sensor devices.

Recently, a methodology to correct the wet particle mass concentrations to dry mass concentrations was proposed by Crilley et al. (2018) based upon the κ-Köhler theory (Petters and Kreidenweis, 2007). κ-Köhler theory describes the relationship between particle hygroscopicity and volume by a single value, κ, and can be adapted to relate particle mass to average bulk hygroscopicity at a given RH. Crilley et al. (2018) calculated κ values representative of ambient bulk hygroscopicity using co-located reference instruments to derive a correction factor for the derived OPC-N2 (Alphasense) particle mass concentrations. Application of this *in situ* correction factor by Crilley and co-workers notably improved the OPC-N2 reported PM$_{2.5}$ and PM$_{10}$ mass concentrations, to be within 33 % of the reference instrumentation at urban background location with high ambient RH. Subsequently, Di Antonio et al. (2018) proposed a similar method that applied κ-Köhler theory to correct the particle size distribution measured by the OPC-N2, and then calculated the particle mass fraction concentration using this corrected particle size. Using this approach Di Antonio and co-workers also observed notable improvement in the OPC-N2, to be within 43 % of reference PM$_{2.5}$ mass concentrations. Di Antonio and co-workers assumed κ values for their correction factor, based upon the assumed major hygroscopic components of the aerosol mix (ammonium sulphate and sodium chloride), which may not be realistic considering the complex multi-compositional nature of urban particles.

It is clear that the aerosol hygroscopicity is a key parameter to consider when correcting particle mass concentration derived by a low-cost OPC (Crilley et al., 2018;Di Antonio et al., 2018). Aerosol hygroscopicity is dependent on the chemical compounds present, and consequently the derived correction factor may vary from location to location due to differences in particle bulk composition, shape and density. To investigate this, we utilised datasets containing co-located particle measurements from reference instruments and a low-cost OPC collected in four cities in three continents: Birmingham, UK; Nairobi, Kenya; Delhi, India and Beijing, China. Across these four cities, the airborne particle composition and range of ambient RH varied considerably allowing for the exploration of sensor performance in response to these factors (composition and RH) and how it affected the calculated correction factors. We also report measurements taken near a volcano in Nicaragua, at a location that received regular volcanic plumes that contained particles that were typically chemically homogenous. The aim of this paper was to investigate whether a universal correction method can be reasonably applied to reported particle mass concentrations by low-cost OPC across a wide variety of locations.

2.0 Method

The datasets used in the current work were acquired in several different field campaigns, at different times, but the same type of low-cost particle sensor was deployed at all five locations, the OPC-N2 manufactured by Alphasense. This sensor has been described in detail in Sousan et al. (2016) and Crilley et al. (2018) and can be considered as a miniaturized optical particle counter. The measured particle number concentration by the OPC-N2 is converted via on board factory calibration to particle mass concentrations for PM_1, $PM_{2.5}$ and PM_{10} size fraction according to European Standard EN481 (OPC-N2 manual). Data collection with the OPC-N2 followed the procedures outlined in Crilley et al. (2018) at the four urban locations via a Raspberry Pi employing the py-opc Python library (Hagan et al., 2018). Uncorrected PM mass concentrations were used without any modification. In Birmingham, Delhi, Beijing and Nairobi the OPC-N2 was housed within a custom-built with the same inlet length (12 cm 3/8” dia. stainless steel tubing, see Crilley et al. (2018) for details) ~~was used for each OPC-N2~~and was placed outside. At Nicaragua, the OPC-N2 was part of the commercially available AQMesh system (Air Monitors), with uncorrected PM concentrations extracted.

2.2 Measurement Locations

Each site in this study, other than the volcanic Nicaraguan site, are classed as urban background for their respective cities. We have focused on $PM_{2.5}$ mass concentrations in this study, as this particle size fraction was measured by reference instrumentation at all study sites. We also note that we used a different OPC-N2 sensor at each site.~~.~~ Previous work has shown that co-located multiple OPC-N2 have an inter-unit precision of 22±13 % for PM_{10} mass concentrations (Crilley et al. 2018). A summary of the measurement location is provided in Table 1.

Table 1: Summary of the measurement sites. Full details available in the text. N/A signifies not available. Custom housing for the OPC-N2 as per description in text.

Location	**Site Description**	**OPC-N2 housing**	**Reference instrument**	**Aerosol composition instrument**
Birmingham BAQS, UK	Urban background	Custom	GRIMM	N/A
Birmingham Tyburn, UK	Urban background	Custom	TEOM-FDMS	N/A
Beijing, China	Urban background	Custom	TEOM-FDMS	AMS
Delhi, India	Urban background	Custom	TEOM-FDMS	ACSM
Nairobi, Kenya	Urban background	Custom	Gravimetric	N/A
Masaya volcano, Nicaragua	Volcano	AQMESH	N/A	N/A

2.2.1 Birmingham, United Kingdom

The OPCs were deployed at two urban background locations in Birmingham. The first was the Birmingham Air Quality Supersite at Elms Road (BAQS, 52.4554 N, 1.9286 W), located with the University of Birmingham campus (Alam et al., 2015) and will be referred to as Bham BAQS throughout. The second site was the Tyburn Road air monitoring station, part of the UK Automatic Rural Urban Network (AURN), referred to as Bham Tyburn throughout. This dataset has been previously described in Crilley et al. (2018), and the current work focuses on the long-term measurements (October 2016 till February 2017) at Bham BAQS, using the OPC-N2 that gave the most complete time series. The reference instrument for $PM_{2.5}$ mass concentrations measurements at Bham Tyburn was a Tapered Element Oscillating Microbalance with a Filter Dynamic Measurement System (TEOM-FDMS). The Bham Tyburn dataset is used to compare to other sites that had the same reference instrument (Beijing and

Delhi, See Sections 2.2.2 and 2.2.3). The reference instrument at Bham BAQS was a GRIMM Portable Aerosol Sampler (model 1.108) that was serviced and calibrated before the measurements. The GRIMM is an OPC type device similar to the low cost sensors, but does contain a pre-conditioning step that reduces the internal RH of the device. Previous work demonstrated that the GRIMM was not affected by RH, based on co-located measurements with a TEOM-FDMS (Crilley et al., 2018).

2.2.2 Beijing, China

The measurements in Beijing formed part of the Air Pollution and Human Health in a Chinese mega-city (APHH-Beijing, www.aphh.org.uk), a joint UK-China program addressing air quality in Beijing (Shi et al., 2019). The measurements took place at the Chinese Academy of Science Institute of Atmospheric Physics (IAP) tower campus (39.9735 N, 116.3723 E), located in the northern suburbs of Beijing. The OPC-N2 sampled on top of a shipping container, with a height of approx. 2.5 m from 5th-9th December 2016. In addition, co-located ground level measurements with a TEOM-FDMS set to measure $PM_{2.5}$ mass concentrations and an Aerodyne Aerosol Mass Spectrometer (AMS) were obtained (Xu et al., 2019).

2.2.3 Delhi, India

The measurements in Delhi were part of the Air Pollution and Human Health in an Indian mega-city (APHH-Delhi, www.urbanair-india.org), a joint UK-India project tackling air pollution in Delhi. The sampling location was Indian Institute of Technology Delhi (IITD) main campus in Hauz Khas (28.5464 N, 77.1913 E), located in the southern suburbs of New Delhi. The instruments were located on the roof (4 stories) of Block IV at IITD. The inlet for the co-located $PM_{2.5}$ TEOM-FDMS was approximately 5 m from the OPC-N2 at the same sampling height. On-line measurements of inorganic aerosol concentrations were provided by an Aerodyne Aerosol Chemical Speciation Monitor (ACSM), located nearby on the IITD campus in Block V, at the same sampling height (Gani et al., 2019).

2.2.4 Nairobi, Kenya

The measurements in Nairobi are previously reported in Pope et al. (2018). These measurements are part of the 'A Systems Approach to Air Pollution' program (ASAP East Africa, www.asap-eastafrica.com). In the current work, we used the urban background data that were collected on the rooftop of the American Wing building at the University of Nairobi (1.2801 S, 36.8163 E). The sampling inlet was at a height of 17 m above ground level with unobstructed airflow in all directions. The measurement period is from 2 February to 24 March 2017. Calibration of the OPC was carried out in situ using a standardized gravimetric approach by collocation with of the OPC and an Anderson dichotomous impactor (Sierra Instruments Inc., USA).

2.2.5 Masaya volcano, Nicaragua

Masaya is an active volcano that is currently degassing and due to its low altitude (600 m a.s.l.) the volcanic plume causes persistent gas and PM air pollution in nearby populated areas. The presented results are part of the first study of high temporal and long-term measurements of PM and SO_2 concentrations in several populated areas near Masaya volcano. Here we will discuss the results from station 789, set up in Pacaya community (11.9553°N, 86.3013°W, 870 m a.s.l.) 15 km to the west of Masaya volcano. Because it is located at a higher altitude than the degassing crater, the volcanic plume frequently grounds there. The station was set up at Susie Syke private clinic, on a post approximately 6 m above ground level, where it was not obstructed by vegetation, buildings or other objects. The site is not believed to be influenced by firewood burning. It is located ~100 m from a paved highway (busy during morning and evening rush hours) but is upwind of it during the predominant weather conditions. Measurements were performed using an 'AQMesh' pod, a commercially available sensor package. SO_2 concentrations were measured by an Alphasense B4-series electrochemical sensor; while particle concentrations were measured by the OPC-N2. Note uncorrected particle mass concentrations were extracted from the AQMesh. The AQMesh was operational between 27 February 2017 and 15 December 2017. Gaps in the data time series are due to power outages.

2.3 Description of applied correction factor

The methodology for the applied correction factor has previously been described in detail by Crilley et al. (2018). Briefly, the correction factor uses κ-Köhler theory to relate the particle mass to hygroscopicity for a given RH (Pope, 2010), according to Eq (1):

$$a_w = \frac{\left({}^{m}/_{m_o} - 1\right)}{\left({}^{m}/_{m_o} - 1\right) + \left(\frac{\rho_w}{\rho_p}\kappa\right)} \tag{1}$$

Where a_w is the water activity (a_w = RH/100), m and m_o are the wet and dry (RH=0 %) particle mass, respectively while p_w and p_p are the density of the dry particles and water, respectively. The value for κ, which relates the bulk aerosol composition to hygroscopicty, can be determined by a non-linear curve fitting of a humidogram, calculated using the ratio of a wet/dry particle mass as a function of water activity (a_w, RH/100). Throughout, we used the raw mass concentrations as reported by the OPC-N2. We used the reference instrument measurements (TEOM-FDMS and GRIMM, as indicated in Section 2.2) as the dry particle mass while the raw OPC-N2 is the wet particle mass concentration. The TEOM-FDMS employs a Nafion dryer and therefore measures dry particle mass concentration (Grover et al., 2006). Equation (1) can be rearranged to calculate the correction factor C according to Eq. (2):

$$C = 1 + \frac{\frac{\kappa}{\rho_p}}{-1 + \frac{1}{a_w}} \tag{2}$$

To calculate the particle mass concentration from measured particle number size distribution, spherical particles of a uniform density and shape are assumed by the OPC, which is not strictly true for airborne particles in an urban atmosphere but is considered a standard approximation. For full details see Crilley et al. (2018). The OPC-N2 assumes the ambient particle density to be 1.65 g cm^{-3} across all size bins to derive the particle mass concentrations from the measured particle number concentrations (Crilley et al., 2018), therefore we have used this density for the dry particles (ρ_p) in Eq. (2). We assume that the particle density is uniform across the particle size distribution measured by the OPC-N2. Furthermore, we assume that both the OPC-N2 and reference instrument are responding to same dry aerosol mass for the all particles below the size cut-off on the reference instrument. We also note that we assume both the OPC-N2 and reference instrument responses are linear over the range of measured concentrations at each site. The raw particle mass concentration derived by the OPC-N2 are corrected according to Eq. (3):

$$PM_{corr} = \frac{PM_{raw}}{C} \tag{3}$$

3 Results and Discussion

A wide range of ambient particle concentrations and relative humidity was observed across the different measurement locations, with an overview provided in Table 2~~1~~. Typically, low particle concentrations were observed in Birmingham and Nairobi with higher levels of humidity in Birmingham compared to Nairobi (Table 2~~1~~). Meanwhile a high particle load was observed in Delhi and Beijing, as would be expected for winter in these two cities. In Delhi, there was a wide range of humidities observed (10-100 %), while in Beijing it was relatively dry (mean of 36±15 %) during the measurement periods. The observed difference in particle load and composition between sites allows for the effect of relative humidity on the OPC-N2 measurements and the applied correction factor to be explored in the following sections.

Table 2~~1~~: Summary of measurement datasets. Reported OPC-N2 $PM_{2.5}$ mass concentrations are uncorrected. For the Nicaragua measurements there was no co-located reference instrumentation. Only one 24 hr average gravimetric $PM_{2.5}$ concentration was available for Nairobi, presented with stated measurement uncertainty.

Site	**Date**		**RH (%)**	**OPC-N2 $PM_{2.5}$ (µg m^{-3})**	**Reference $PM_{2.5}$ (µg m^{-3})**
Birmingham	Oct 2016-Feb 2017	Average ± SD	89±10	38 ± 54	11 ± 8
		Range	44 - 99	0.3 - 566	0.5 - 63
Beijing	Dec 2016	Average ± SD	36±15	74±82	75±66
		Range	13 - 81	3 - 274	2.7 - 208
Delhi	Jan-Feb 2018	Average ± SD	59±25	218±228	168±76
		Range	9 - 100	12 - 1113	50 - 478
Nairobi	Feb-Mar 2017	Average ± SD	51±18	32±16	27.6±6.8
		Range	16 - 89	4 - 135	
Nicaragua	Feb-Dec 2017	Average ± SD	77±11	21±55	NA
		Range	39 - 91	0.5 - 742	

3.1 Effect of RH on measurement by OPC-N2 at all sites

To explore if there was evidence for an artefact on the OPC-N2 derived $PM_{2.5}$ mass concentrations due to RH, we plotted the reported $PM_{2.5}$ mass concentrations by the OPC-N2 as a function of RH (Fig 1). From Fig 1, there is clear influence of RH on the measurements performed in Delhi and Birmingham, evidenced by the observed exponential increase in particle mass with RH (Figs 1a and 1d). At Beijing, the observed stepwise increase in the derived measured particle mass between a RH of 40-50% RH may point to deliquescence of a predominant PM component (Fig 1c), explored further in later sections. What is evident from these three sites (Beijing, Birmingham, Delhi) was there was a large spread in derived $PM_{2.5}$ mass concentrations at high RH, which likely reflects the heterogeneous nature of the particle composition and hence hygroscopicity.

Meanwhile at Nairobi, the derived concentrations by the OPC-N2 appeared to be independent of RH. Typically during the dry season in Nairobi, airborne mineral dust comprises a large fraction of $PM_{2.5}$ (35% annual mean, (Gaita et al., 2014), which is known to have low hygroscopocity. Furthermore, we note that the measurements in Nairobi were performed during the dry season and as a result the ambient RH was typically less than 85%, the RH value identified in Crilley et al. (2018) where the OPC-N2 becomes significantly sensitive to RH. This combined with the low hygroscopocity of the aerosol in Nairobi, was the likely reason why there was little evidence for an RH artefact observed for the OPC-N2 (Pope et al., 2018). Therefore, a simple calibration against gravimetric measurements is likely to be sufficient in locations with low RH and low proportion of hygroscopic aerosols, such as Nairobi.

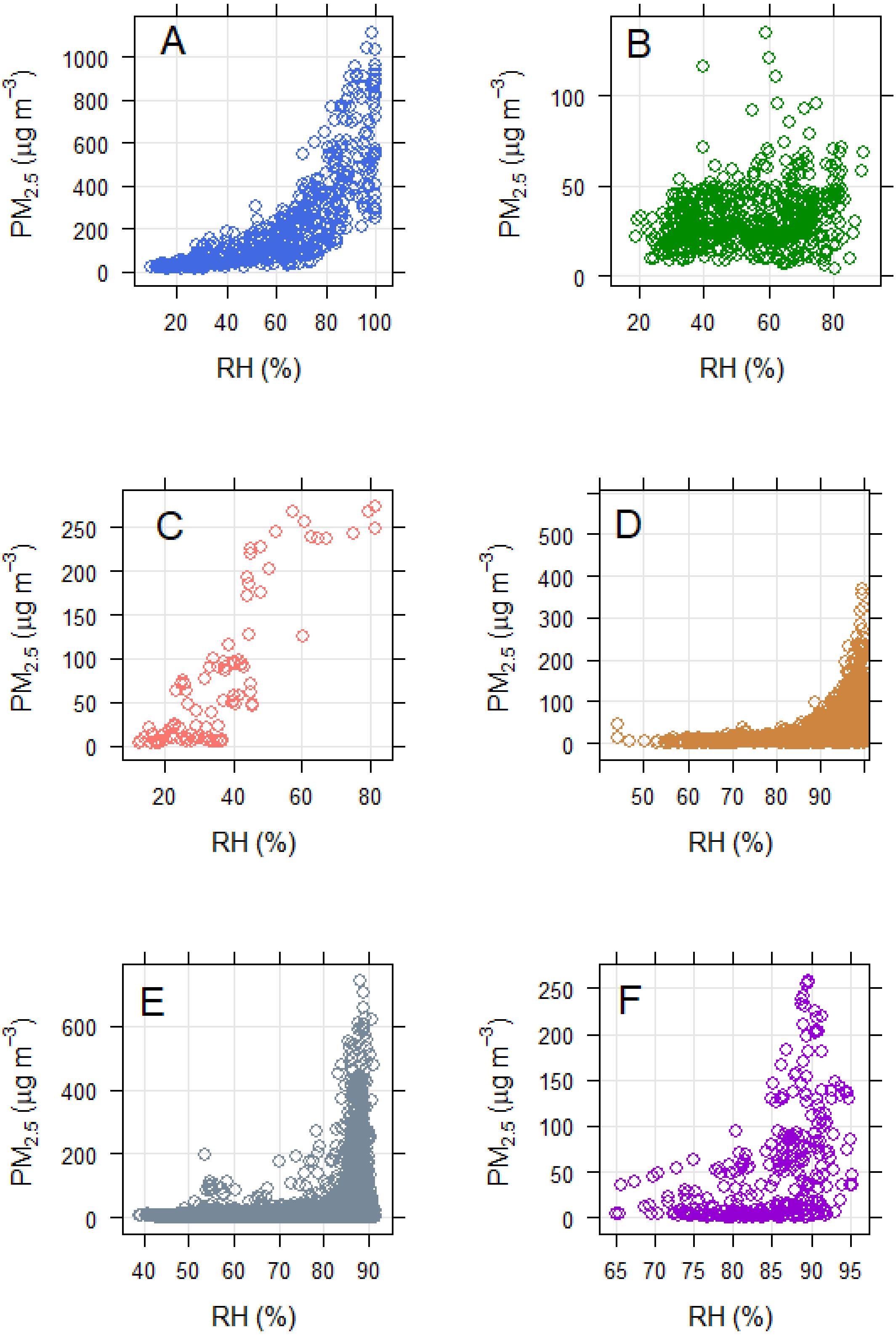

Figure 1: Plot of reported PM$_{2.5}$ mass concentration by OPC-N2 against ambient RH for the whole measurement period in Delhi (A), Nairobi (B), Beijing (C), Bham BAQS (D), Nicaragua (E), Bham Tyburn (F). Note the different y- and x-axis scales.

As posited in Crilley et al. (2018), the RH artefact on the OPC-N2 was likely related to the ambient aerosol bulk hygroscopicity. Therefore, we plotted humidograms for sites from Fig. 1 with evidence for an RH effect where reference particle mass concentration data was available (i.e. Birmingham, Delhi and Beijing; Table 2~~1~~), shown in Fig 2. In Beijing, there was insufficient data at high RH due to the short time period of sampling (4 days), and the factors affecting the response of the OPC-N2 explored in more detail in Section 3.3. A quasi-exponential increase in the ratio of OPC-N2 to reference instrument concentrations at high RH was observed at Birmingham and Delhi, as would be expected if the aerosols were undergoing hygroscopic growth (Fig. 2). Using κ-Köhler theory (Petters and Kreidenweis, 2007), calculated κ were 0.1 and 0.16 for Birmingham and Delhi, respectively. These κ values are typical for continental regions with high organic loadings (Pringle et al., 2010). While high organic loadings would be expected for Delhi during winter, there is also significant loadings of hygroscopic aerosols such as sulphate and nitrate (Gani et al., 2019) and this is explored in more detail later. Differences in aerosol composition would likely explain why the calculated κ at Bham BAQS (0.1) was lower than that observed at Bham Tyburn (0.38-0.41, Crilley et al. 2018). Previous work in Birmingham demonstrated that the proportion of ammonium sulphate and nitrate decreases in winter compared to summer (Yin et al., 2010), and may explain the observed lower κ value over winter. What becomes evident from Figure 2 is that different κ values were observed at each site. If the aerosol composition was broadly similar at each site, we would expect the same κ value. This suggests that the aerosol composition varies significantly over the different measurement sites. The scatter in OPC/TEOM observed in Fig 2 as function of RH was likely due to temporal variability in aerosol composition due to changing sources and sinks (both local and regional).

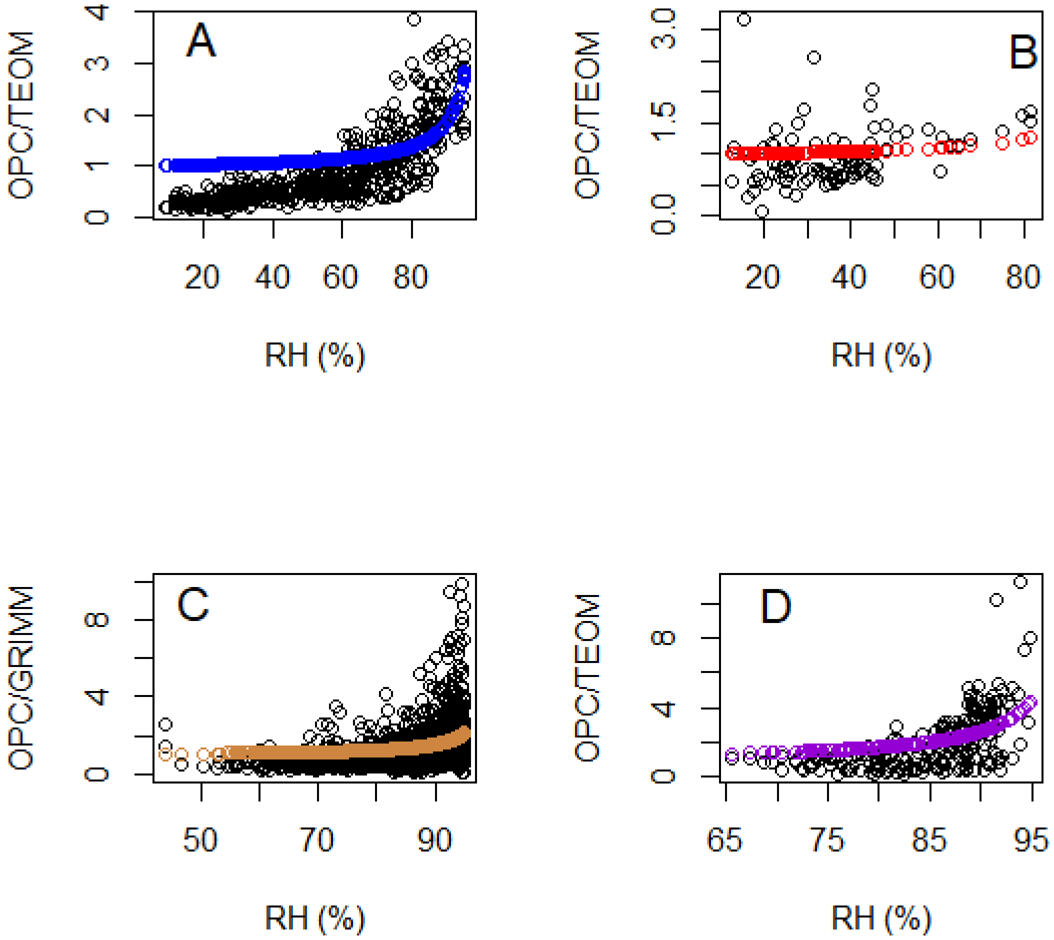

Figure 2: Humidograms with corresponding κ fit (shown in colour) for Delhi (A), Beijing (B), Bham BAQS (C), Bham Tyburn (D). Note the different y-axis and x-axis scales. The two-stage correction factor described in Section 3.3.1 has not been applied for these humidograms.

3.2 Effect of aerosol composition on OPC-N2 RH correction factor

3.2.1 Mixed aerosol composition (Urban)

To explore the effect of aerosol composition on correction factor for the OPC-N2, we focus first on the Beijing and Delhi measurements, as co-located on-line aerosol composition data was available at these two sites. During the measurements in Beijing, there were periods when the OPC-N2 and TEOM were in reasonable agreement, typically at lower PM concentrations as observed by the regulatory grade equipment. Fig. 3a demonstrates that at concentrations below ca. 150 µg m^{-3}, there was linear relationship between reported OPC-N2 and the TEOM concentrations (r^2 of 0.85), with a slope of 0.72. When the $PM_{2.5}$ mass concentrations were above 150 µg m^{-3}, the relationship appeared to deviate from linearity, but these were also the times when the RH was higher (>50 %, Fig. 3a). Generally, the times of high RH also corresponded to times of relatively high sulphate concentrations (Fig. 3b) and to a lesser extent high particle nitrate concentration (Fig. S1, Supporting Information). Both nitrate and sulphate aerosol have high hygroscopicities (Petters and Kreidenweis, 2007).

Similar trends were also observed in the Delhi measurements. Generally, at low RH there appears to be a linear relationship between the reported OPC-N2 and TEOM concentrations (Fig. 4a), but this deviates from linearity at high RH, similar to that observed in Beijing. For periods where the ambient RH in Delhi was less than 50 %, we observed that the OPC-N2 generally recorded $PM_{2.5}$ mass concentrations half that of the TEOM (slope of 0.48 and r^2 of 0.55). This was broadly similar to that observed in Beijing (Fig. 3a) and suggests that the OPC-N2 generally under-reports $PM_{2.5}$ mass concentrations at low RH (<50 %). Also apparent from Fig. 4a, there were times in Delhi when the RH was high (>80 %) and yet the $PM_{2.5}$ mass concentrations by the OPC-N2 were of a similar relationship to the TEOM at low RH.

We therefore plotted the relationship of OPC-N2 and TEOM $PM_{2.5}$ mass concentrations coloured by the sum of sulphate and nitrate concentrations (Fig. 4b) and generally, when the concentration of these species and RH were high, we observed notably higher OPC-N2 concentrations relative to the reference. Both sulphate and nitrate are highly hygroscopic aerosols and this therefore suggests that the high readings by the OPC-N2 relative to the TEOM in Beijing and Delhi were due to water uptake by hygroscopic particles as suggested by Crilley et al. (2018). From Figs. 3 and 4, it appears that this effect was occurring at RH above 50 %, below the deliquescence point of ammonium sulphate (79 %) indicating the ammonium sulphate component of the aerosol was in a metastable state. Aerosols with multi-component mixtures are observed to deliquesce earlier than the deliquescence points of the individual components e.g. Pope et al. (2010). It is noted that the nitrate component of the aerosols have a smoother continual take up of water with respect to RH~~suggesting that there was uptake by other aerosols such as nitrate (Hu et al., 2010)~~. ~~Nitrate aerosols also have a smoother continual take up of water~~ (Gibson et al., 2006;Hu et al., 2010). ~~, and this points to nitrate aerosols contributing at RH below 79 %. Aerosols with multi-component mixtures are observed to deliquesce earlier than the deliquescence points of the individual components e.g. Pope et al. (2010). Overall, these results from Beijing and Delhi suggest that the applied calibration depends not just on the ambient RH but also aerosol composition.~~

Figure 3: Derived OPC-N2 uncorrected $PM_{2.5}$ mass concentrations against TEOM $PM_{2.5}$ mass concentration measurements coloured by ambient RH (A) and fraction of sulphate to the total $PM_{2.5}$ mass ~~sulphate concentration~~ (B) in Beijing. The straight line indicates the linear regression fit for concentrations below 150 µg m^{-3}. Dashed line is 1:1.

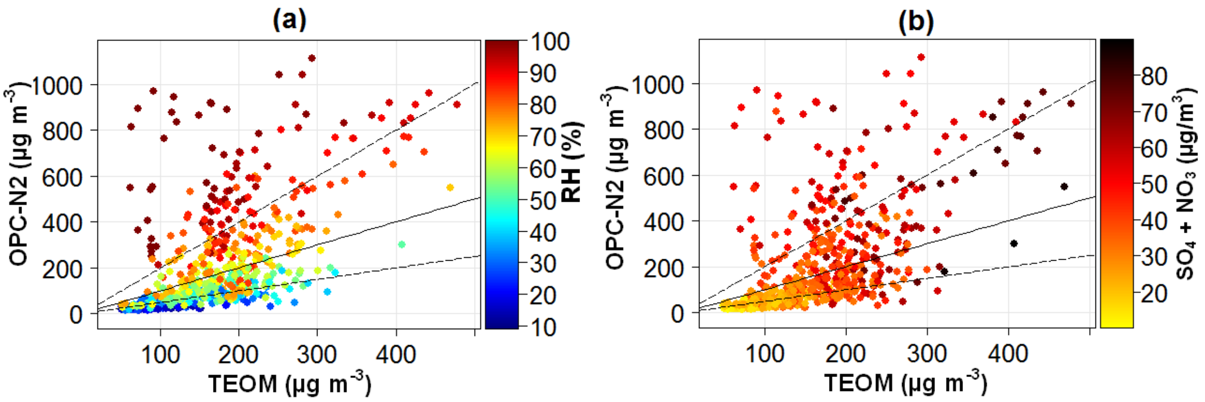

Figure 4: Derived OPC-N2 uncorrected $PM_{2.5}$ mass concentrations against TEOM $PM_{2.5}$ mass concentration measurements coloured by **(a)** ambient RH and **(b)** sum of particle sulphate and nitrate concentration in Delhi. The solid line is 1:1, while the dashed lines are 0.5:1 and 2:1.

3.2.2 Homogenous aerosol composition (volcano plume)

Fresh volcanic plumes are typically dominated in composition by sulphuric acid and therefore these plumes offer an opportunity to explore using k-Köhler theory to develop the correction factor in a substantially homogenous aerosol mix under ambient conditions. If the RH artefact is due to aerosol hygroscopicity, then the resultant humidogram using data collected by the OPC-N2 in the plume should resemble that for sulphuric acid. To derive the volcanic plume humidogram, shown in Figure 5, the following steps were taken. The plume was identified at station 789 when the collocated gas phase SO_2 measurement was greater than 20 ppm. The aerosol within the plume was assumed to be composed solely of sulphuric acid with a corresponding κ value of 1.19 (Wexler and Clegg, 2002). The dry mass of the volcanic particles were calculated using Eq. (1), with RH input from the collocated measurements at site 789. The derived humidogram was compared to the pure sulphuric acid humidogram calculated using E-AIM model I (Fig. 5). The observed agreement between model and measurements strongly points to particle hygroscopic growth driving the high particle mass concentrations observed by the OPC-N2 at high RH.

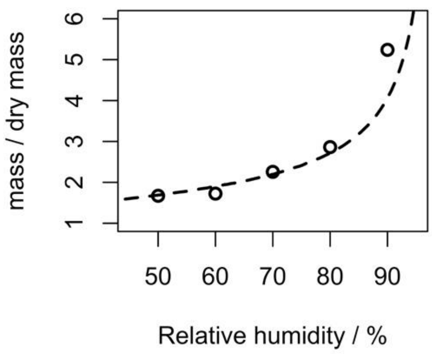

Figure 5: Comparison of humidograms from pure sulphuric acid and that observed in the Nicaragua volcanic plume. Circles: Nicaragua plume aerosol. Dashed line: modelled sulphuric acid humidogram from the E-AIM model.

3.3 Evaluation of the OPC-N2 performance in Delhi and Birmingham

During the measurements in Delhi, the OPC-N2 typically over-reported the $PM_{2.5}$ mass concentrations relative to the reference (Fig. 6A). The OPC-N2 assumes a uniform particle density of 1.65 g cm^{-3} in the particle counts to mass conversion, and this density may be inappropriate for Delhi aerosol during winter. Previous measurements of aerosol density during winter in Delhi at midday were on average 1.28±0.12 g cm^{-3} (Sarangi et al., 2016), lower than applied by the OPC-N2. Generally, the OPC/TEOM ratio was below 1 (Fig 2A) and so this would not fully explain this discrepancy~~and so this would not explain this discrepancy~~. From Fig. 6A, higher $PM_{2.5}$ mass concentrations relative to the reference were reported by the OPC-N2 towards the start of the measurement period (Fig. 6A), generally during the times when the RH was high (e.g. 25-29th Jan, Fig. 6B). We also note that the reported concentrations by the OPC-N2 towards the end of the measurements in Delhi (11th Feb onwards, Fig. 6A), were in better agreement with reference. The cause of this change in performance is unclear but could reflect lower RH or changes in aerosol composition. ~~We also note that the reported concentrations by the OPC-N2 appear to lose structure towards the end of the measurements in Delhi (11th Feb onwards, Fig. 6A), which may point to deterioration in the OPC-N2 performance (possibly due to dust coating within the instrument). This may be the reason why the OPC-N2 reported lower concentrations relative to the reference towards the end of the measurement period. As the OPC-N2 measured for 24 days in total, this suggests that the lifetime of low cost particle sensors in Delhi may only be of the order of a month.~~

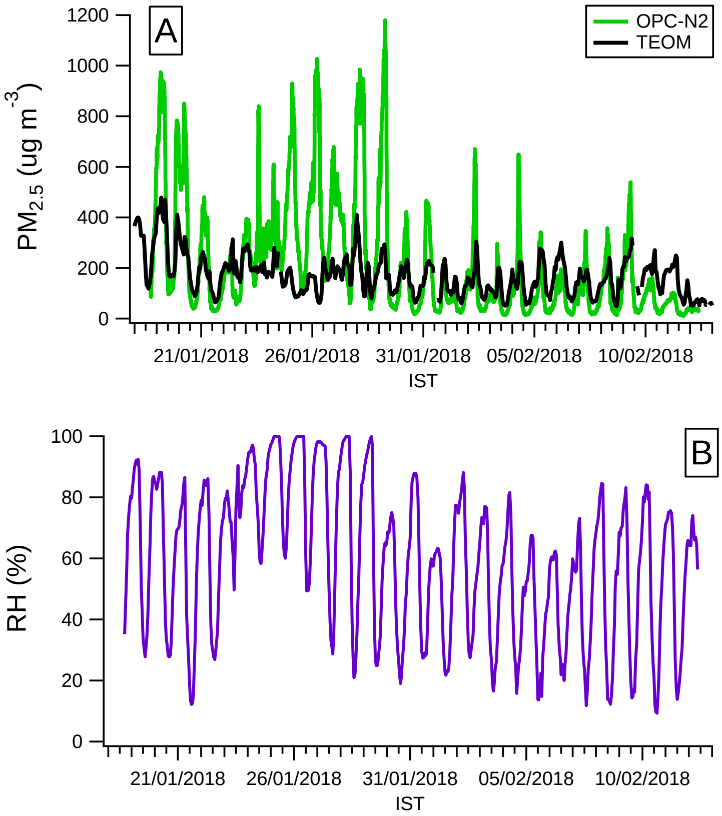

Figure 6: Time series of reported OPC-N2 and TEOM-FDMS $PM_{2.5}$ mass concentrations (A) and ambient RH (B) at IIT Delhi.

As the reported OPC-N2 concentrations at Birmingham and Delhi demonstrated an artefact due to RH (Fig. 2), we applied the correction factor from Crilley et al. (2018), using the *in situ* locally derived κ values. In addition, the κ for ammonium sulphate (0.61) was also used, as Di Antonio et al. (2018) suggested it may be more representative for urban aerosols. The results of the correction factors, relative to the co-located reference instruments are summarized in Table 3~~2~~. Compared to the uncorrected OPC-N2 concentrations, application of correction factor with both κ resulted in improved performance of the reported concentrations relative to the reference. However, the using the locally derived κ resulted in the best correction of the OPC-N2, to be within 33 % of the reference measurements, compared to using the ammonium sulphate κ (Table 3~~2~~).

Table 3~~2~~: Slopes of uncorrected and corrected $PM_{2.5}$ mass concentrations from the OPC-N2 relative to the reference instruments (r^2 in brackets). Intercepts were not constrained to zero. There were four OPC-N2 measuring at Bham Tyburn and the range is presented.

		CORRECTED	
	UNCORRECTED	*Locally derived κ*	*κ for NH_4SO_4*
BHAM BAQS	3.5 (0.24)	1.3 (0.44)	0.5 (0.24)
DELHI	1.73 (0.33)	1.1 (0.60)	0.55 (0.60)
BHAM TYBURN	2.5-3.5 (0.64-0.67)	0.98-1.33 (0.82-0.85)	0.72-0.98 (0.84-0.86)

We also observed that ratio of OPC-N2/GRIMM concentrations was low at high RH at Bham BAQS (Fig. 2C). The spread in OPC/GRIMM ratios observed at high RH at Bham BAQS is indicative of a wide range of aerosol composition with differing hygroscopocity over the 4 months. The calculated κ value for each month in Birmingham and found little variability (0.1-0.12) from October to January and suggest that on average the bulk hygroscopicity of the aerosols was consistent, though within a large range (as indicated by the large spread in OPC/TEOM ratio in Fig 2c).

3.3.1 Two-stage correction methodology for datasets with wide range of ambient RH

For the Delhi dataset, the observed wide range of ambient RH may have affected the correction factor (Fig. 2), as particle hygroscopic growth would be limited at low RH. Consequently at low RH, defined as RH <60 %, a linear correction factor may be more appropriate. From Fig. 2, there appeared to be a linear relationship between the OPC-N2 and TEOM $PM_{2.5}$ measurements for RH less 60 %. We therefore calculated a linear correction factor for the OPC-N2 relative to the TEOM when the ambient RH was less than 60 % (Fig. S3, Supporting Information) and applied it the reported OPC-N2 $PM_{2.5}$ concentrations. Using this normalised OPC-N2 concentrations, the humidogram was replotted (Fig. 7), and the corresponding κ fit calculated (Fig. 7). Using this normalised OPC-N2 measurements, the κ line better matches the observed OPC/TEOM (Fig. 7) compared to using the uncorrected OPC-N2 data (Fig. 2). The calculated κ value from Fig. 7 was 0.45, which may be considered more realistic considering the high sulphate and nitrate loading in Delhi (Fig. 4). Using the κ from Fig. 7, we corrected the normalised OPC-N2 $PM_{2.5}$ concentrations via Eq. (2) and (3). The time series of the corrected OPC-N2 concentrations is shown in Fig. S4 (Supporting Information), and the application of this two-stage correction method resulted in the OPC-N2 being in good agreement with the reference instrument (slope of 1.1, r^2 of 0.61). However, we note that this is similar to the agreement observed when the OPC-N2 was corrected without the two-stage approach (Table 3~~2~~). However, the two-stage approach resulted in more physically realistic humidograms and κ (Fig. 7) for Delhi and this approach may be more appropriate for locations that experience a wide range of ambient RH.

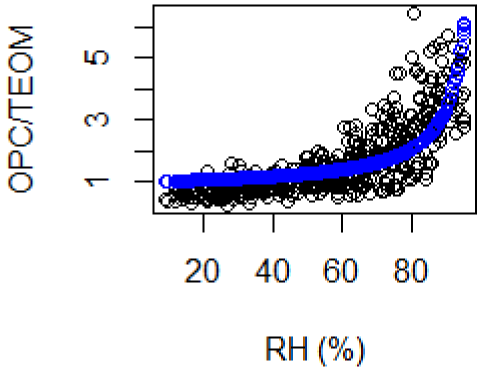

Figure 7: Humidograms for Delhi using normalised OPC-N2 $PM_{2.5}$ mass concentrations with corresponding κ fit.

3.4 On the global applicability of correction factors

The results so far point to the need to know aerosol composition in order to accurately apply a suitable correction factor, in agreement with previous work (Di Antonio et al., 2018;Crilley et al., 2018). However, to determine aerosol composition at the necessary time resolution would require expensive co-located equipment measuring aerosol composition (e.g. an Aerosol Mass Spectrometer) and somewhat negating the USP of a low cost monitor. Di Antonio et al. (2018) suggest air mass origin (via HYSPLIT) could provide compositional information in order to determine the appropriate κ value to use in the correction. We therefore examined the long-term Birmingham dataset for times when the applied correction factor over/under-corrected the OPC-N2 mass concentrations relative to reference instrument. However, unlike Di Antonio et al. (2018) we could not find any consistent patterns with respect to air mass origin and the performance of the correction factor. Furthermore, in order to apply a correction factor via this approach would require significant post-processing time. This therefore raises the question if it remains a 'low-cost' option.

To remain a low-cost option, a simple correction that can be applied to the OPC-N2 irrespective of aerosol composition changes is needed, though this may decrease the accuracy of the correction factor. For many locations around the world, ambient $PM_{2.5}$ mass concentrations are measured using gravimetric-based techniques (e.g. filters or TEOM) for regulatory purposes. Consequently, we have focused on developing a simple correction factor using TEOM data as a reference. To explore if this was viable we plotted OPC/TEOM ratio for all sites where this was available (Bham Tyburn, Delhi, Beijing) on one plot (Fig. 8). Note we used the two-stage correction for the OPC-N2 measurements in Delhi as described in Section 3.3.1 for Fig. 8. We did not apply this correction to the Bham Tyburn data as the RH was >60 %.

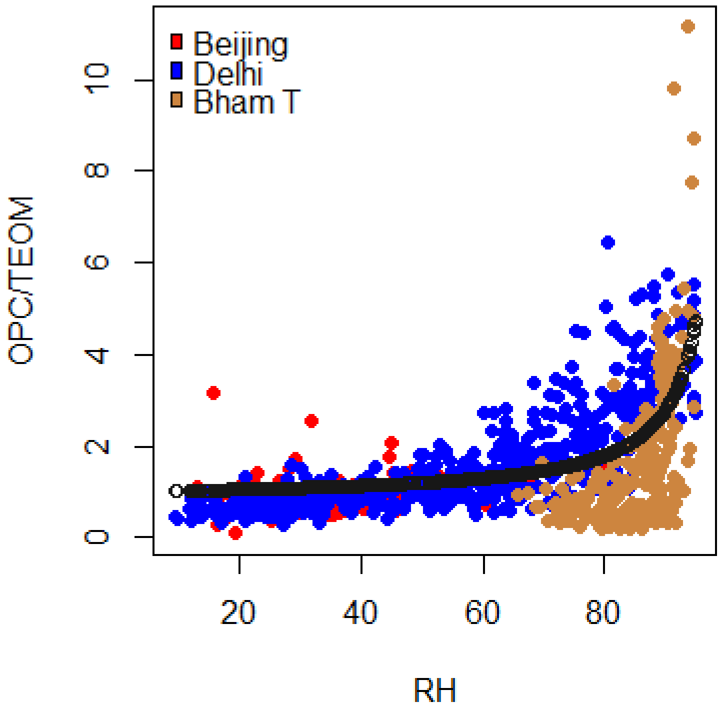

Figure 8: Humidogram using OPC-N2 data where there was dry reference mass (TEOM), coloured by location. The resultant κ fit (black) was generated using data from all three sites.

From Fig. 8, a κ of 0.33 (assuming a uniform particle density of 1.65 for all sites) was calculated, slightly higher than the average of 0.3 suggested for continental regions (Pringle et al., 2010). Consequently, we refer the calculated κ from Fig 8 as global κ but note it was calculated from 3 urban background sites on two continents. We applied this κ ~~of (~~0.33) to correct the OPC-N2 data at all sites, as well the ~~global~~ average for continental regions (0.3), with the results summarised in Table 4~~3~~. Variation in the κ values generally resulted in changes to the slope, while the correlation co-efficient remained similar. While the *in situ* derived κ resulted in a reasonable correction of the OPC-N2 relative to the reference (±10-30 %, Table 4~~3~~), using the κ from Fig. 8 (0.33) was comparable. Correcting the datasets using the global κ of 0.33 resulted agreement with reference instruments of within 50 % at all sites, with the Beijing, Delhi and some of the Bham Tyburn corrected OPC-N2 being within 20 % of reference (Table 4~~3~~). The only site with a notably poorer agreement using the global compared to the *in situ* κ was Bham BAQS, and this may be because the locally derived κ for the Bham BAQS measurements (0.1) was different compared to the other sites. This notwithstanding we do note that using a κ of 0.33 resulted in a significant improvement in accuracy compared to the uncorrected OPC-N2 derived particle mass concentrations at Bham BAQS (slope of 3.5, Table 3~~2~~). Overall, when considering the most appropriate correction or κ the results from Table 3 suggest that a locally derived κ, based on an *in situ* calibration with reference instrumentation, is preferable. However, the global κ derived using data from the three urban background locations in this study (0.33, Fig. 8) gave comparable results to the *in situ* derived correction (Table 4~~3~~). Therefore, it suggests that using this κ value or suitable literature values for urban background sites may be acceptable should there be no reference instruments available for calibration.

Table 4~~3~~: Comparison of performance local and the global correction factors for correcting OPC-N2 using the Crilley et al. (2018) method, shown as a slope relative to the reference instrument, with r^2 values given in parentheses. Intercepts were not constrained to zero.

SITE	IN SITU DERIVED CORRECTION	GLOBAL (κ = 0.33)	CONTINENTAL AVERAGE (κ= 0.3)
BHAM TYBURN	0.98-1.33 (0.82-0.85)	1.1-1.5 (0.82-0.85)	1.2-1.6 (0.82-0.85)
DELHI	1.1 (0.60)	0.80 (0.61)	0.84 (0.61)
BHAM BAQS	0.96 (0.42)	0.54 (0.45)	0.57 (0.45)
BEIJING	1.35 (0.87)	0.85 (0.85)	0.87 (0.85)

4.0 Conclusions

Recent work has demonstrated that aerosol hygroscopocity is likely the key parameter to consider when correcting particle mass concentration derived by a low-cost OPC, particularly at high ambient RH. Consequently, correction factors have been developed that apply κ-Köhler theory to correct for the influence of water uptake by hygroscopic aerosols. In the current work, we explored the performance of this correction factor using datasets with reference instruments and a low-cost OPC (OPC-N2) co-located in environments that had differing aerosol composition, particle load and ambient RH. We observed evidence that the enhanced high concentrations reported by the OPC-N2 relative to reference instrumentation during periods of high RH was related to the amount of hygroscopic aerosols (sulphate and nitrate) and RH, as expected if the bulk aerosol hygroscopicity was driving this response. This was most clearly observed during measurements in volcanic plumes in Nicaragua, where the observed humidogram closely resembled the calculated pure sulphuric acid humidogram. This agreement between model and measurements strongly points to particle hygroscopic growth driving the high particle mass concentrations observed by the OPC-N2 during times of high ambient RH.

The results indicate that the particle mass concentration measurements reported by low-cost OPC during periods of high RH (>60 %) need to be corrected for aerosol hygroscopic growth. We employed the correction factor method outlined in Crilley et al. (2018) to account for this and observed corrected OPC-N2 $PM_{2.5}$ mass concentrations to be within 33 % of reference at all sites. The choice of applied κ was found to be critical. The results from the current work indicate that an *in situ* derived κ (using suitable reference instrumentation) leads to the most accurate correction relative to co-located reference instruments. The *in situ* derived κ would also likely be dependent on the time of year if there were any local seasonality to the bulk aerosol composition, and this would need to be considered when determining appropriate calibration procedures.

An average κ of 0.33 was calculated using measurements from three urban locations around the globe (Beijing, Birmingham, and Delhi). Applying this global κ in the correction factor notably improved reported OPC-N2 $PM_{2.5}$ mass concentrations relative to uncorrected, to be within 50 % of reference measurements at all sites. Therefore, for areas where suitable reference instrumentation for developing a local correction factor is lacking, using a literature κ value can result in a reasonable correction. For locations with low levels of hygroscopic aerosols and

RH (such as Nairobi), a simple calibration against gravimetric measurements (using suitable reference instrumentation) would likely be sufficient. Whilst this study generated correction factors specific for the Alphasense OPC-N2 sensor, the calibration methodology developed is likely amenable to other low cost PM sensors.

Data Availability

Data available on request.

Author Contributions

LRC and FDP conceived the study. LRC, AS, LJK and FDP performed the data analysis. LRC, LJK, MDS, MSA, SY, MF, PF, YS, SG, JA, SA, RW, EI, DN, MG and FDP contributed to data investigation and curation. LRC wrote the original draft, with all other co-authors contributing to review and editing.

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