Wright, M. D., Matthews, J. C., Silva, H. G., Bacak, A., Percival, C., & Shallcross, D. E. (2020). The relationship between aerosol concentration and atmospheric potential gradient in urban environments. *Science of The Total Environment*, 716, [134959]. <https://doi.org/10.1016/j.scitotenv.2019.134959>

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[10.1016/j.scitotenv.2019.134959](https://doi.org/10.1016/j.scitotenv.2019.134959)

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The relationship between aerosol concentration and atmospheric potential gradient in urban environments

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14 **Abstract**

15 Urban aerosol is a growing concern for people living within cities; aerosol have been implicated in
16 many ill health conditions, including that of the lung and of the heart. Atmospheric potential gradient
17 is a consequence of charge carried to the ionosphere through thunderstorms, and its value depends
18 on highly electrically mobile ion concentrations, hence local conductivity of the air. Ions attach to
19 aerosol in the atmosphere, reducing their mobility and therefore increasing the potential gradient,
20 and so potential gradient measurements have been suggested as a proxy for aerosol measurements.
21 Particle number count, size distribution and potential gradient were measured for two campaigns in
22 Manchester, U.K., and one campaign in Bristol, U.K. Using a factor based on size distribution to account
23 for preferential attachment at larger sizes provided the best relationship with potential gradient, but
24 particle count alone showed a weaker, but similar relationship. The increase in particle count caused
25 by annual bonfire and fireworks celebrations (November) was evidenced in both potential gradient
26 and particle numbers. Daily regression or correlation did not show a consistent relationship. In the
27 larger Bristol data set, increasing humidity led to a reduction of potential gradient, while increasing
28 particle number led to an increase.

1. Introduction

Air pollution is one of the main causes of global ill health and mortality, leading to an estimated 4.9 million deaths worldwide (GBD, 2018), and is of particular concern in urban areas due to the proximity of large populations to sources such as road traffic, domestic cooking and heating activities and industry. Air pollution is a complex and dynamic mixture of gases and both primary and secondary particulates, causing a range of adverse respiratory, cardiovascular, cancer and possibly also neurological outcomes (WHO, 2013; IARC, 2016; Xu et al., 2016; GBD 2018). Whilst identifying the health effects associated with specific components of this mixture is difficult, there is strong evidence for a causal association between exposure to 'fine' particulate matter, $PM_{2.5}$, and mortality (COMEAP, 2018; Apte et al., 2018). However, within $PM_{2.5}$ there are also a wide range of chemical components and particles of different sizes, which can vary significantly in time and by location, which may contribute differentially to health effects (COMEAP, 2018). The smaller size fractions, sub-micron ($< 1 \mu m$) and, in particular, ultrafine ($< 100 nm$) particles are of increasing concern, in part because of their high number concentrations, large specific surface area, and increased ability (relative to larger particles) to penetrate to, and hence deposit in, the distal parts of the lung (ICRP, 1994). There are also recent observations of translocation of a small but detectable fraction of the smallest particles into other organs, including the brain (Maher et al., 2016) and sites of vascular disease (Miller et al., 2017a, b). In the UK, there is no specific guideline or regulation relating to the ambient concentration of ultrafine particles, and whilst some modelled emissions factors for (ultrafine) particle number and mass exist, in 2018 there were only 3 monitoring sites dedicated to measuring submicron and ultrafine particle number concentrations and size distributions, all in South East England (AQEG, 2018).

The atmospheric potential gradient (PG) has been explored as a proxy for measurement of aerosol concentrations in historic data, and has been used to link macrophysical and microphysical processes (Silva et al 2015; Silva et al 2016; Harrison 2012). The ionosphere is kept highly charged due to thunderstorm activity and charged precipitation throughout the globe transferring charge from the

ground upwards (Rycroft et al 2007). The atmosphere is weakly conducting due to solar and cosmic rays and, near ground, ground based radiation ionising the air. The charge from the ionosphere travels back to ground through the ionised atmosphere completing a global electrical circuit, therefore a potential difference exists between the ground and the ionosphere which can be measured at ground level as PG.

The global PG can be affected by climate, as thunderstorm activity increases, or by space weather (Rycroft et al 2012; Silva and Lopes 2017). Locally the atmospheric PG can be affected by the production of ions, such as from traffic (Jayaratne et al 2015) or high voltage transmission lines (Matthews et al 2010; Wright et al. 2014); or it can be affected by charging in clouds, thunderstorms, dust and other meteorological processes (Yaniv et al 2016). Measurement of atmospheric PG is becoming more commonplace to provide insight into meteorological and space phenomena (Nicoll et al 2019).

The presence of aerosol near the surface of the earth acts as a sink of small ions in the atmosphere, when ions attach to aerosol the mobility of the charge carrier is significantly decreased and so the conductivity of the air is decreased. If the air conductivity is reduced then the PG will increase in that region, as a consequence of Ohm's law. This has been demonstrated through a relationship between measured aerosol concentration and PG (e.g. Yaniv et al 2016; Kubicki et al 2016), and used to explain a correlation with gaseous pollutants and PG (Silva et al 2016) and to infer the presence of aerosol from PG measurements alone (Aplin 2012). Therefore, measurement of PG could contribute towards improving the currently sparse spatial distribution of monitoring stations probing the number concentration of submicron particles, albeit indirectly, without the requirement for costly and/or high maintenance instrumentation as is currently employed in existing networks (AQEG, 2018). An investigation of the atmospheric PG and smoke measurements in historic data at Kew Gardens, U.K., allowed new insights into the levels of pollution at the time through the extrapolation of a theoretical PG in a smoke free environment (Harrison 2006). Comparison of this limit to that of maritime air

implied the presences of aerosol not measured by the existing apparatus. Therefore, it is important to clarify the nature of the relationship between measured PG and aerosol number concentrations in the urban environment, where number concentrations are generally higher and hence the coupling between the two quantities should be strongest, and where most public exposure (in terms of particle number) occurs.

The aims of this research are to use measurements aerosols and PG in two datasets to evaluate the use of PG as a tool to understand aerosol properties within a city. Previous work has shown that PG will increase with increased pollution, specifically aerosol concentration, due to a reduction of small ions (Silva et al 2016; Conceição et al 2016), and that increasing humidity will cause hygroscopic growth in aerosol and therefore increase small ion loss and increase PG (Silva et al 2015). We would expect to see a diurnal cycle in aerosol concentrations due to traffic patterns, which should be evident in PG measurements (Silva et al, 2014; Matthews et al 2019). Additionally, while we know that meteorological conditions can affect PG, particularly in disturbed weather (Harrison and Nicoll, 2018; Matthews et al 2019) we would like to test if effects can still be seen in a large data set without data filtered for fair weather fields.

2. Theory

2.1 Potential gradient, air conductivity and Ion-aerosol attachment

PG is related to air conductivity σ at the Earth's surface through the Air-Earth current, J_z , according to Ohm's law (Eq. 1), while the conductivity is a result of the number and electrical mobility (μ) of the airborne charge carriers (Eq. 2). If ions are lost to aerosol, then there is a significant reduction in the average mobility of the charge carriers, reducing the conductivity.

$$PG = J_z / \sigma \quad (1)$$

$$\sigma_{\pm} = e \sum n_{\pm} \mu_{\pm} \quad (2)$$

In equation 2, the subscript $\pm$ denotes each polarity, and e is the elementary charge. The ion-aerosol attachment process is described by ion-aerosol population balance equations, e.g.:

$$\frac{dn_{\pm}}{dt} = q_{ion} - \alpha n_{+}n_{-} - n_{\pm} \int_D \sum_{q=0}^{\infty} \beta_{\pm, \pm q}(D) N_{\pm q}(D) dD - n_{\pm} \int_D \sum_{q=1}^{\infty} \beta_{\pm, \mp q}(D) N_{\mp q}(D) dD \quad (3)$$

Where q_{ion} is the air ionisation rate, α is the ion recombination coefficient, n the concentration of small ions, N the concentration of aerosols and D the diameter of the aerosols. Within the population balance equation, $\beta_{\pm}$ are ion-aerosol attachment coefficients which modulate ion-aerosol interactions and hence aerosol influence on PG. In general, $\beta_{\pm}$ increases with increasing aerosol diameter (Hoppel and Frick 1986).

To simplify the complex mathematical description of the ion-aerosol interactions for a dynamic aerosol time series, an effective attachment coefficient, β_{eff} , can be obtained from the aerosol size distribution (Hoppel 1985, Horrak et al 2008), using

$$\beta_{eff} = \frac{\int \beta_{\delta}(D) N(D) dD}{N_T} \quad (4)$$

Where $\beta(D)$ is a function of the diameter,

$$\beta_{\delta}(D) = \sqrt{\frac{D-1nm}{D+5nm}} \cdot \frac{D}{40nm} \cdot 10^{-6} \quad (5)$$

As in urban areas typically the number of particles of increasing size tends to peak around 100 nm (Asmi et al., 2011), whilst the effective attachment rate increases with size, there will be a peak attachment effect at a given size, calculated from the size distribution and effective ion-aerosol attachment rate, AR (equation 6), which would be typically between 200 and 300 nm.

$$AR = \beta_{eff} N \quad (6)$$

A consequence of the ion balance equation is that effect of aerosol on PG can be quantified with respect to an aerosol limit (Eq. 7), when the number concentration of aerosol is known (Harrison 2006; Conceição 2016).

$$PG = \frac{J_z \beta_{eff}}{2q_{ion}Ze} N + PG_0 \quad (7)$$

2.2 Aerosol hygroscopicity and PG

The size distribution of aerosols can be affected by relative humidity (RH) if the particles are sufficiently hygroscopic. As discussed previously the size distribution of aerosol affects the rate of ion attachment, and hence the measured PG in the atmosphere. The effect of relative humidity on PG due to hygroscopic growth was described in a basic formulation by Silva et al (2015), Eq. 8, where k_B is the Boltzmann coefficient, T the temperature, and κ an effective hygroscopicity parameter relating to the hygroscopic properties of the polydisperse aerosol,

$$PG \approx \frac{\pi J_z k_B T}{e^2 q_{ion}} ND_{a,0} \left(1 + \kappa \frac{RH/100}{1-RH/100} \right)^{1/3} \quad (8)$$

3. Experimental Methodology

Three experimental campaigns were undertaken, where atmospheric PG and aerosol number concentrations were measured in urban environments. The first two were located at Manchester University between 13/07/2015 and 07/08/2015 and between 30/10/2015 and 12/11/2015, and the third at the University of Bristol between 23/05/2016 and 11/08/2016. In the first Manchester campaign, particle number concentration (PNC) data was supplemented by aerosol size distribution measurement.

3.1 Manchester Campaigns

In both Manchester campaigns, measurements were taken at a sampling station approximately 20 m above ground level. PNCs were obtained with a Condensation Particle Counter (CPC, Model 5.403, Grimm Aerosol) and size distributions with SMPS (TSI, Models 3082 Electrostatic Classifier, 3081 Differential Mobility Analyzer and 3775 CPC, July-August data only) for aerosol diameters 18-514 nm. For PNC data aerosol was sampled through 2 m conducting 6 mm internal diameter tubing, while for

SMPS data aerosol was sampled through a larger metal tube of ~ 8 m length with high sample flow rate, as other instrumentation was sampling concurrently as part of a different study. Losses within these sampling tubes were not assessed in the current study because these are likely to affect predominantly smaller particles with relatively low ion-aerosol attachment rates. PG data was obtained using an electric field mill meter (JCI 131, Chilworth) on an outdoor balcony, where a sonic anemometer recorded the local wind speed and direction. This meteorological data was supplemented by wind speed and direction data from the Whitworth Observatory, an open rooftop site close to the aerosol and PG measurement location. The local wind speed will be affected by the street geometries but provide the local conditions under which the aerosol and PG measurements were taken, while the rooftop measurements are more indicative of city-wide conditions (both measurement sites are described in Matthews et al, in press). Rain measurements were provided from METAR recordings from Manchester Airport. The balcony was in a shielded area of the University, which will distort the value of PG, and so absolute values of PG are not presented here, rather the mean PG for each measurement period was calculated and the data normalised to this mean. As rainfall can have a strong influence on urban PG (Matthews et al 2019), data 1 hour before and after rainfall (as determined from METAR records) was discarded. To negate potential bias introduced into the calculated mean because of missing data (either deliberately removed due to rain, instrument zeroing/maintenance, intermittent instrument failure or the start/end times of measurements) the data for each second of the day across the measurement period were averaged, to compile an averaged daily cycle, which is presented in Fig. 1 below and used to calculate the daily and overall means. The second campaign was chosen to incorporate November 5th, around which time bonfires are traditionally lit and fireworks set off at a number of events in the UK, previously shown to enhance particle concentration and significantly alter aerosol chemistry (Reyes-Villegas et al., 2018).

3.2 Bristol campaign

A JCI 131 field mill (Chilworth) and Maximet 500 (Gill) weather station were positioned on the roof of the University of Bristol School of Chemistry between 2016 and 2017. Details of the setup are given in Matthews et al (2019). The atmospheric PG will be distorted by the presence of buildings and the non-flat environment meaning that absolute PG values are not reported, but to infer details of aerosol content relative PG value changes can be used, and so the PG measured at site, not corrected for geometric distortion, is used in this analysis. Rain measurements were not measured on site for this analysis, though measurements at the We the Curious science centre, Anchor Road, Bristol (see Matthews et al 2019) indicate that there was rainfall on 47 days during the measurement period. For the Bristol campaign, we did not remove rain in the analysis to assess whether the influence of aerosol can still be detected in the whole data set. Particle number concentration was determined using a TSI 3010 CPC with an upper limit of 10,000 particles cm^{-3} . A dilution system was put in place to increase this range to 14,000 cm^{-3} . The performance of the TSI 3010 CPC with dilution system was compared to a TSI 3007 CPC on six occasions during the campaign.

4. Results

4.1 Manchester results

The time series of PG, PNC and AR is shown in Fig. 1(a) and 1(b) for both Manchester measurements, and the daily average cycle shown in Fig. 1(d) and 1(e). The SMPS measured for a small subset of the July measurements, and the PNC and AR calculated from the SMPS size distribution are shown in 1(c). The PG visually looks to be a good fit with the AR in both Fig. 1(a) and between 1(c) and 1(d), with a peak in PG corresponding to the rush hour at 08:00 UT, though the minima in the daily averaged cycle is overnight (03:00 UT) for both PG and (CPC-derived) PNC, but not for AR. This may be due to a shift in the size distribution of particles, which display higher mean diameter overnight (Fig. 2(b)) and hence, because of higher aerosol attachment coefficients for larger aerosol, AR remains higher despite PNC in general being lower overnight. Note that small aerosols in the range between the Grimm CPC

lower detection limit (5 nm) and the SMPS lower size limit as operated here (18 nm) will be included in Fig. 1(d) and 1(e) but not 1(c), and this is one possible reason why for the same data collection period the diurnal variation appears different. The Carnegie curve is also displayed in Fig. 1(d) and 1(e), which is the global background potential gradient derived by thunderstorm activity throughout the globe (Harrison 2013). The Carnegie Curve is not likely to be apparent in any daily measurement but can be seen in the diurnal average of the summation of multiple sites (Nicoll et al 2019) and in single rural sites when there is no known source or aerosols or space charge (e.g. Matthews 2012; Lopes et al 2017). The relationship between the Carnegie curve and PG does not look to be strong in this analysis, with pollutant influence dominating.

Fig. 2. Time series of (a) normalised potential gradient (PG), attachment rate (AR), and particle number count (PNC) measured by condensation particle counter (CPC) and scanning mobility particle sizer (SMPS) in July 2015, and (b) PG and PNC (CPC) measured in November 2015 and average daily time series of (c) PNC measured by SMPS, geometric mean diameter (GMD) and AR, (d) PG and PNC (CPC) in July 2015, and (e) PG and PNC (CPC) measured in November 2015.

Fig. 1(b) shows the PNC and the PG on the same axis scales for the shorter November period. It shows clearly the increase in particle concentrations throughout each day as fireworks and bonfires are lit, with clear peaks in the evening. The daily average cycle shows that the daily peak in PNC has shifted from the 15:00 to 18:00 UT traffic related peaks in the summer, to a peak at 22:00 UT. Firework displays start in the evening and will continue into the night, the increase in PNC is a result of these localised sources being dispersed to the measurement location. Tracer experiments in Manchester have shown that material released 2 km away can be detected, and night-time boundary layer conditions will provide a stable atmosphere, keeping pollutants at high concentrations (Matthews et al in press). The change of conductivity of a parcel of air due to the introduction of aerosol is a process that can typically take a few minutes to hours (Conceição et al, 2016) which is a similar time scale to the transportation of material over a km scale (Matthews et al, in press). However, new ions are created throughout the passage of the aerosol plume at a constant rate due to radioactive and cosmic ray processes. It is also the case that new aerosol from combustion processes are smaller and may grow over time, increasing the attachment rate to ions, so it may be expected that high aerosol concentrations due to distant sources, as in the November measurements, may have an enhanced effect on the PG level. The effect of very close sources on PG is not known from this analysis, and measurements using co-located field mills and particle counters roadside would be recommended to determine whether there is an immediate effect on PG by ion-aerosol attachment or space charge effects when there is an influx of very high aerosol concentration.

Fig. 2. Average particle size distribution measured in 10 days in July and (b) average size distributions of the four 6 hourly time periods during the same measurement period.

Frequency domain analysis of the atmospheric PG and the effective attachment rate (AR) were calculated using Lomb Scargle periodograms (Lomb 1976). Lomb Scargle periodograms were calculated from 9 days data between 21/07/2015 and 30/07/2015, aerosol size distributions were provided in 90s samples, so PG was averaged to 90 s samples for comparison. The PG shows daily and half daily peaks, indicative of traffic related activity. This is consistent with previous studies; Matthews et al (2019) compared rural measurements south of Bristol, UK to urban measurements in Bristol and which both showed a daily peak in the Lomb-Scargle periodogram, the half day cycle was stronger in the urban site. Silva et al (2014) strongly identified both the half day and whole day cycle, as well as a weekly signal, in a dataset of 10 years of PG from a Bendorf electrograph in Lisbon, Portugal. The asymptotic spectral amplitude, S of the periodograms as a function of sampling time T to an exponent was calculated using a power law polynomial fit. The periodograms for AR and PG are visually very similar and have a power law fit of 1.65, which is close to the 5/3 Kolmogorov theoretical exponent for turbulence spectra. This would imply that both PG and the aerosol concentrations are both influenced by atmospheric turbulence (Conceição et al 2018).

Fig. 3. Lomb-Scargle periodograms for (a) AR and (b) PG measured in Manchester in July 2015.

Fig. 4. Daily regression coefficients for PG v particle count as measured by CPC and SMPS, and AR, for measurements in Manchester in 2015.

Daily regression coefficients were found for the July data (Fig. 4) which show that the correlation is not consistent by day, but that the AR is the best correlated measurement. However, the correlation coefficients for PNC are similar, and so PNC alone may be used investigate the relationship between PG and aerosols in an urban environment.

Fig. 5. Histograms of the distribution of minute average (a) PG and (b) PNC where 23% of measurements were above 14000 cm⁻³ which are not indicated in the scale.

Longer term relationships between PNC and PG were explored using 3 months of data recorded on the roof of the School of Chemistry. The PG dataset as a whole was investigated, without removing data during rainfall or for a fair-weather condition, which will weaken any relationship between PG and PNC (Matthews et al 2019, Nicoll and Harrison 2018). Fig. 5 shows histograms of (a) the PG and (b) the PNC at the site, the distribution of PG at the site compares well to PG distributions measured at other sites in the UK with predominantly positive PG with a small amount of negative fields (Bennett and Harrison 2008, Matthews et al 2010). The PNC distribution has a peak at ~6000 cm⁻³. Due to equipment limitations, the upper limit of PNC is restricted to 14000 cm⁻³, and 23% of measurements fall above that limit. The histogram shows that the peak of the number probability density distribution is around 6000 particles cm⁻³, though the daily mean will vary. Due to the loss of higher counts in the data, non-parametric statistics will be used for PNC data analysis. The daily mean PG and median PNC were calculated for the entire data set. The daily mean of PG shows considerable variability, which will be related to prevalent meteorological conditions and local aerosol and ion production.

Fig. 6. Time series of atmospheric measurements between May and September 2016 on the roof of the School of Chemistry showing (a) the mean atmospheric PG and median PNC, (b) the Spearman Rank correlation coefficient calculated for each day, (c) mean temperature and humidity measured on those days and (d) average rainfall measured at a different site.

Due to the upper limitation on PNC, the Spearman rank Correlation Coefficient was calculated instead of linear regression for 1 s measurements of PG and PNC measured in the same location on the roof of the School of Chemistry. Fig. 6(b) shows a time series of each day, and – as with the Manchester data shown in Fig. 5, there is no consistent pattern showing the relationship between PG and PNC. There was no apparent relationship between the daily correlation coefficient with daily averages of PNC, PG, humidity or temperature.

Box plots were plotted of the minute averages for PG against increasing PNC, and PG against humidity to test the relationships discussed in section 2.1. Fig. 7(a) shows the PG increasing as PNC increases, while Fig. 7(b) shows a decrease in PG as RH increase. The increase in PG as PNC increases is as would be expected from the known theory and previous measurements. Referring back to Eq. 7, the PG in the limit of $PNC = 0$ is a positive value at approximately 10 V m^{-1} .

Fig. 7. (a) Box plots of increasing PNC and PG (b) Box plots of increasing RH and PG

The change in PG with RH can be considered in 3 ranges, below 60% the effect on PG would be dominated by ion mobility changes, but there is not sufficient measurements in this range to see any

clear affect. Between 60 and 90%, hygroscopic growth would be the dominant mechanism affecting the relationship, and there is a very small increase in PG between 75 and 90%. At the higher humidities, the air is highly saturated, which may be accompanied by generation of space charge, charged clouds overhead and charge separation. In this region, effects due to hygroscopic growth could be easily masked by the large changes in PG that can occur under charged clouds, in precipitation and due to space charge (Bennett and Harrison 2007; Matthews et al 2019).

PG and PNC measurements were taken in Bangkok during 2018, and the relationship also showed a reduction in PG with increasing humidity (Matthews et al, in press); PG median values were calculated for 10% size bins of RH between 50% and 100%, with a reduction in median values in each increasing bin. In Bangkok the primary source of sub-micron aerosol is likely to be traffic related, and therefore contain a significant hydrocarbon fraction. Hydrocarbon based aerosol will be less susceptible to hygroscopic growth than aerosol from clean air (Silva et al 2015) and therefore the low growth rate in the hygroscopic region may be an indication that the aerosol in Bristol has a low hygroscopic growth factor and is therefore likely to be hydrocarbon based. However, direct measurement of the aerosol composition or hygroscopic growth factor using a technique such as Hygroscopicity-Tandem Differential Mobility Analyser system would be required to confirm this hypothesis (Massling et al 2011).

5. Discussion and conclusions

The use of PG as an indicator of air-pollution within an urban area remains difficult in real time due to the large number of other variables that can affect electric fields in urban areas. The predominant effect discussed here is that of aerosol loading causing a reduction of conductivity due to smaller ions being reduced by attachment. In Eq. (3) the dominant terms in a high aerosol environment will be the aerosol-attachment terms. However, where there is a source of ions the ion production term q may not be constant and can often cause a charge imbalance. Ions can be produced by electrical infrastructure such as sub-stations and high voltage power lines where one polarity may predominate

(Ling et al 2010; Wright et al 2014), which in turn can affect measured PG due to clouds of space charge (Matthews et al 2010) and increase the charge state on aerosols (Matthews et al 2015). Traffic has also been shown to produce bipolar ions, which may not create areas of space charge but can affect the conductivity in the air affecting the measured PG (Ling et al 2010; Maricq et al 2006; Lee et al 2012).

The assumption that changes in PG are caused by changes in aerosol content requires the assumption that the Air-earth current remains constant (Eq. 1). The current measured at ground is a summation of four components: the conduction current caused by the Earth's electrical circuit, a displacement current due to changes in field above (such as that caused by clouds of space charge), a turbulent current caused by the movement of space charge to ground through turbulent processes, and a precipitation current caused by charged precipitation (Bennett and Harrison 2006). The fair-weather conduction current is affected by space weather phenomena and can undergo large fluctuations due to solar disturbances (Elhalel et al 2014). The addition of reliable instrumentation to measure the conduction current at the surface of an urban environment would provide additional information on the Earth's circuit and allow calculation of the local conductivity, while the measurement of ion mobility spectra and ion concentrations of both polarities would provide an accurate measurement of the local conductivity (Wright et al 2014). Where PG measurements alone may prove to be reliable, robust and low maintenance, the advantage of running such a full suite of instrumentation requiring regular maintenance above known aerosol measurement spectra is difficult to see.

Within the Manchester data set, data was omitted from the analysis due to precipitation, to limit the effects of non-fair weather fields. Rain data was removed one hour before and after rainfall, as well as during, to account for the distance between rain and PG measurement and the effect that charged clouds overhead can have before and after rainfall. A true fair-weather criteria would account for non-precipitating electrified clouds, and low wind speed conditions that can allow build-up of space charge

(Harrison and Nicoll, 2018), but the removal of rain data was considered sufficient in removing most of the noise from the signal.

The Bristol data set was a much larger data set and enabled us to investigate whether the effect of pollutant aerosol could be seen within the dataset above the natural changes in PG caused by non-fair weather conditions so no attempt to isolate non-fair weather days was made. It is known that urban PG is affected strongly by weather effects, which is demonstrated well in Bangkok during monsoon season (Matthews et al, in press) but also in Bristol (Matthews et al 2019) where disturbed weather effects show large extremes of PG that are measured within the city scale (~ 1 km) but are different at a regional scale (~ 20 km). It was also shown in the Bristol data in Matthews et al (2019) that not all extremes of PG variation were lost when using rainfall measurements from a different site in the city. While the increase in PG at very high humidities may be caused by non-fair-weather fields, therefore masking the effect of hygroscopic aerosol growth, at lower humidities an effect due to hygroscopic growth could be seen. The rising PG with aerosol concentration in the Bristol data, even including non-fair-weather fields, shows that – in a long term (several weeks) data set – the effect of pollution on PG is discernible in the full data set.

The measurements shown in Matthews et al 2019 show differences in the local and regional scale of PG measurements. Non-fair weather conditions show large changes in PG over the local (~ 1 km) scale, in fair weather, the fields are more similar. A global network of potential gradient measurements may also provide information on the global background, if local influences can be statistically removed (Nicoll et al 2019).

PG has been measured in two UK cities alongside PNC to evaluate the use of PG as a proxy for PNC. Both PG and PNC had strong daily and half daily cycles, consistent with traffic related effects of pollution. High aerosol concentrations during bonfire events in Manchester caused a change in the diurnal cycle of both aerosols and PG compared with summer measurements at the same location, with a high aerosol concentration overnight elevating night-time PG.

Analysis of a longer data set in Bristol, with no removal of data due to non-fair-weather conditions, showed less evidence of a correlation between PG and PNC on a daily basis, but, histograms of PG against PNC measurements did show an increasing trend. The measurement of atmospheric PG remains an interesting tool to understand aerosol behaviour in cities, but is most useful when analysed alongside meteorological data that can identify fair weather conditions.

Acknowledgments

This work was funded by Leverhulme Trust grant (RPG-2014-102) and NERC Grant number NE/K01501X. We thank Ed Aldred for assistance during the Bristol measurements. PG Data from these sites will be stored in the GloCAEM database, which is found at: <http://catalogue.ceda.ac.uk/uuid/bffd0262439a4ecb8fadb0134c4a4a41>.

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