The spatio-temporal evolution of black carbon in the North-West European ‘air pollution hotspot’

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Abstract

Particulate black carbon has a range of negative impacts on health, the environment and climate, however despite this there are relatively few long-term studies on its ambient distribution as a tropospheric air pollutant. In order to address this lack of data, to help to provide greater insight into the spatio-temporal distribution of particulate black carbon and to assess potential influencing factors, a new, permanent suburban monitoring network was established with sites in four northwest European cities: London (UK), Leicester (UK), Amsterdam (the Netherlands) and Antwerp (Belgium). We report here an analysis of the first measurements made by the network over a twenty-seven-month period (01/01/2013 - 01/04/2015), alongside data from pre-existing comparator urban roadside (AURN Marylebone Road, London, UK) and rural background (AURN Auchencorth Moss, Scotland) sites. The temporal evolution of black carbon was investigated at each site, as were associations with other commonly monitored pollutants (*e.g.* O₃, NO_x, PM_{2.5}, PM₁₀) and wind fields. Results showed clear anthropogenic signatures across the diurnal, weekly and annual timeframes, and positive correlations were obtained between black carbon measurements and other common traffic-related pollutants, highlighting the importance of vehicle emissions as a major contributor to ambient black carbon concentration in northwest Europe. Average black carbon concentrations varied from 6.6 µg m⁻³ at the urban roadside, to 0.2 µg m⁻³ in the rural background, with suburban and urban background sites having average concentrations in the range of 1.0 – 2.4 µg m⁻³. Wind field analysis further highlighted the importance of road traffic as a source of black carbon and demonstrated the importance of local emission sources at the various receptor locations. Statistical analysis of data between sites generally indicated a weak correlation ($r_s = -0.03 - 0.68$, COD = 0.32 – 0.91), further highlighting the importance of local emissions in determining ambient black carbon concentration. It was also found that black carbon comprised a significant portion of total ambient particulate matter (PM), particularly at sites with the larger traffic volumes and during rush-hour (*e.g.* ~45% of PM_{2.5} at Marylebone Road), however, its contribution to total PM was found to decrease on days of high pollution, indicating the importance of other PM components when air quality is particularly poor.

Keywords

Black carbon; particulate matter; air quality; North-West Europe; health

Highlights

- Black carbon and traffic-related pollutants are strongly correlated at all measurement sites, indicating that road traffic is a significant black carbon source in suburban NW-Europe.
- Cross-site analysis demonstrates BC is a local, not regional, pollutant and a significant contributor towards suburban particulate matter (PM) mass concentration.
- BC contributions to PM are particularly high during periods of increased anthropogenic activity and during the winter months.
- The location of the measurement site and proximity of local sources are crucial in determining ambient BC concentration and in evaluating its impacts on human exposure.

Graphical Abstract

1. Introduction

Air quality has deteriorated around the world since the onset of industrialisation, especially over the last few decades, in line with a steep rise in global population and the prolific use of fossil fuel burning processes (Cramer, 1998; Fenger, 1999). Many studies have now linked a wide range of environmental impacts (Bond et al., 2013; Huang et al., 2011) and human health end-points with exposure to air pollutants, and in particular, with exposure to airborne particulate matter (PM), of which a significant fraction can comprise black carbon (BC) (Janssen et al., 2011; World Health Organisation, 2012; Petzold et al., 2013).

There is a lack of formal agreement on the exact meaning of the term ‘black carbon’, with definitions ranging from the somewhat general, *e.g. ‘an ideally light-absorbing substance composed of carbon’* (Petzold et al., 2013), to the more prescriptive, *e.g. ‘an insoluble, refractory aggregate of small carbon spherules [that] strongly absorb visible light’* (Bond et al., 2013). The principle sources of particulate BC in the atmosphere are biomass burning (Chen et al., 2017; Xu et al., 2018) and the incomplete combustion of fossil fuels within various anthropogenic processes, including road (Cross et al., 2010; Laborde et al., 2012; Vignati et al., 2010); air (Dodson et al., 2009; Masiol et al., 2016; Shirmohammadi et al., 2017) and water transport (Comer, 2015). Estimates suggest that the global BC emission rate is of the order 7,500 – 17,000 Gg yr⁻¹, with source contributions varying significantly by region (Bond et al., 2013).

As a fraction of fine PM, BC is able to remain airborne for periods of several days to several weeks (Druffel, 2004; IPCC, 2017) and as such has the ability to travel thousands of kilometers (Streets et al., 2001). From emission through transport, BC is known to mix with other airborne particles, contributing to transcontinental plumes of atmospheric brown clouds (Ramanathan and Carmichael, 2008).

It is well established that airborne particulate BC plays a major role in global radiative forcing, via both direct and indirect mechanisms. As well as being able to scatter electromagnetic radiation (Tegen and Heinold, 2018), BC is the strongest absorbing constituent of airborne PM (Druffel, 2004) and the second strongest absorbing constituent of the atmosphere as a

whole, after CO₂ (Bond et al., 2013; Husain et al., 2007; Jacobson, 2001). In addition, BC can alter the radiative properties of pre-existing ice particles and liquid droplets within clouds (Bond et al., 2013; Chýlek et al., 1996; Liu et al., 2002), and can alter the albedo of surface snow and ice (IPCC, 2017; Jacobson, 2001). Modelling simulations indicate that BC has a climate forcing impact of the order +1.1 Wm⁻² (Bond et al., 2013; Haywood and Shine, 1995; Husain et al., 2007; IPCC, 2017; Jacobson, 2001; Wang, 2004). BC is a major player in the global climate system and has also been linked with regional climate perturbation (Chameides et al., 1999; Menon et al., 2002).

It has been suggested that in comparison with standard PM_{2.5} and PM₁₀ metrics, BC is a more reliable indicator of harmful particulates and is a particularly important additional air quality indicator to evaluate the health risks of air dominated by primary combustion particles (Janssen et al., 2011). The negative health impacts of air pollution have been shown by numerous toxicological studies (Grahame and Schlesinger, 2010; Murray and Lopez, 1996; World Health Organisation, 2012), with research to link BC to various health endpoints currently on-going (e.g. Ljungman et al., 2019; Redaelli et al., 2019). From this, research evidence is emerging of a consistent association of BC with cardiovascular and respiratory related morbidity and mortality (Delfino et al., 2006; Hoek et al., 2000; Mar et al., 2000; Ostro et al., 2007; Peng et al., 2009; Rich et al., 2006; Zanobetti and Schwartz, 2006). More recently findings have been published suggesting that airborne BC particles have the ability to penetrate the placental barrier of pregnant women causing an increase in cases of low birth weights, premature births and miscarriages (Bové et al., 2019). To conduct such studies, and advance understanding of the effects of BC on health, it is crucial that comprehensive ambient BC measurements should be made over wide geographical areas and in environments where human exposure may be high.

Whilst BC emissions are reported to be decreasing in the US and across Europe (Mousavi et al., 2018, Kutzner et al., 2018) they are increasing in much of the rest of the world. Attempts to regulate black carbon emissions have been instigated in recent years on the global through to the local level (Brewer 2019). Diesel engines been a particular target in urban areas owing to their large (90%) contribution to the transport share of BC emissions (US EPA, 2017). A host

of regulations and mitigation policies have been introduced, from tighter vehicle emission testing standards e.g. EURO 6 European Emission Standards (Commission Regulation (EU) 2016/646 2016), and schemes to reduced diesel cars in cities such as low emission zones (Holman et al., 2015), to the complete phasing out of fossil fuel cars in the next 5-10 years (Brewer 2019). Residential biomass burning, another contributor in urban areas, has also been targeted with schemes such as the Clean Air Strategy in the UK (DEFRA, 2019) and the EPA's New Source Performance Standards for residential wood heaters as part of the Clean Air Act in the US (EPA, 2020). Owing to the emerging evidence of BC as pollutant of concern here have been a number of short-term studies investigating the role of BC in the Earth's atmosphere and its impact on air quality across the world including in Europe (Becerril-Valle et al., 2017; Reche et al., 2011), Asia (Chen et al., 2014; Wang et al., 2013), and the Americas (Healy et al., 2017; Peralta et al., 2019; Shirmohammadi et al., 2017). However, in order to better understand its spatio-temporal evolution and to ultimately help elucidate and legislate for its impacts on the environment and on human health, it is necessary to establish integrated permanent measuring networks for the production of long-term data sets. These networks have been slowly emerging over recent years with longer term measurements now available in some regions (Helin et al., 2018; Mousavi et al., 2018; Sun et al., 2020; Kutzner et al., 2018). To this end a novel network for the long-term monitoring of ultrafine particles and particle composition (including BC) was established across north-west Europe as part of the Joint Air Quality Initiative project (JOAQUIN; www.joaquin.eu).

The JOAQUIN monitoring network was designed to produce high quality data over long time-scales to help provide the required evidence-base to inform and drive health and environment air pollution policy in north-west Europe, which is considered a global 'hot-spot' for air pollution (see Figure 1). This paper follows on from work on ultrafine particles (Hofman et al., 2016) and biomass burning (Cordell et al., 2016) by presenting the first findings from the JOAQUIN network regarding BC across the north-west Europe region (Hama et al., 2017a; Hama et al., 2017b; Hama et al., 2018). The findings presented will go some way towards addressing the current gap in the literature for long-term continuous BC measurements over wide geographical areas and will provide a baseline for future studies to help assess the success of new air pollution reduction policies and measures introduced within the region.

These include the implementation of the Antwerp Low Emissions Zone (2017-2028; Lage-emissiezone Antwerpen, 2020), the Ultra-Low Emissions Zone in London (2019; Ultra Low Emission Zone, 2020) and widening of Low Emissions Zone restrictions in Amsterdam (2017-2030; Low emission zone, 2020).

Figure 1: Time averaged global tropospheric NO₂ column densities (top), with focus on North West Europe (top inset), as measured by the Ozone Monitoring Instrument (OMI), and time averaged global black carbon surface concentrations (bottom), with focus on North West Europe (bottom inset), as modelled by the MERRA-2 model over the study period and visualised using the NASA Giovanni tool (Giovanni, 2020).

2. Experimental

2.1 Monitoring Sites

The JOAQUIN network was established in four major north-west European cities, Eltham London (UK), Leicester (UK), Amsterdam (NL) and Antwerp (BE), as described in detail by Hofman et al. (2016). Each station was deployed in an urban/suburban background location, to ensure measurements were “representative of exposure of the general urban population” to air pollution in each respective city (AEA, 2010). For this study two additional sites from the UK Government Department for Food and Rural Affairs (DEFRA) Automated Urban and Rural Network (AURN), were also included to allow comparison of JOAQUIN urban background data with data derived from urban roadside and rural background air, which represent pollution extremes. The AURN sites employed were located on Marylebone Road in central London and at Auchencorth Moss near Edinburgh (Scotland). The locations of each JOAQUIN and AURN site employed within this study are shown in Figure 2; coordinates, average annual traffic intensities and distance to nearest road are shown in Table 1.

Figure 2: Location of each of the JOAQUIN (i.e. Eltham, Leicester, Amsterdam and Antwerp) and AURN (i.e. Marylebone Road and Auchencorth Moss) measurement sites employed during this study. Base-map obtained from Google Earth and overlain with the time averaged black carbon surface concentrations generated by the MERRA-2 model (visualised using the NASA Giovanni tool; Giovanni, 2020) over the study period.

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213 *Table 1: Monitoring site descriptive parameters*

Country	Location	Site Designation	Distance to nearest main road / m	Traffic intensity ¹ / vehicles day ⁻¹	Coordinates	
					Latitude	Longitude
England, UK	Eltham, London	Suburban Background	60	16,500 ²	51°27'09" N	0°04'14" E
England, UK	Marylebone, London	Urban Traffic	1	80,200 ²	51°31'21" N	0°09'17" W
England, UK	Leicester	Urban Background	140	22,500 ²	52°37'12" N	1°07'38" W
Scotland, UK	Auchencorth Moss	Rural Background	1,500	7,400 ²	55°47'32" N	3°14'34" W
The Netherlands	Amsterdam	Urban Background	64	15,000 ³	52°21'35" N	4°51'59" E
Belgium	Antwerp	Urban Background	30	29,500 ⁴	51°12'35" N	4°25'55" E

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1. Mean annual traffic intensity at the nearest main street
2. Year 2013 (Map Road traffic statistics - Road traffic statistics, 2020)
3. Year 2015: <http://www.verkeersprognoses.amsterdam.nl>
4. Year 2013 (Vlaamse Milieumaatschappij, 2020)

2.2 Instrumentation

Each of the JOAQUIN stations was instrumented with a Model 5012 Multi Angle Absorption Photometer (MAAP; Thermo Scientific, US). The MAAP determines particulate BC concentrations *via* the measurement of light absorption and scattering at 670 nm (Petzold et al., 2013). For the Eltham and Leicester sites, inlets with PM_{2.5} sharp-cut cyclones were employed, for the Amsterdam and Antwerp sites, TSP inlets were installed. All instruments operated at a sample flow rate of 1 m³/h.

BC concentration at the Marylebone Road and Auchencorth Moss AURN sites was measured using a Magee Aethalometer (model AE22), which measures the light absorption of carbon particles at two wavelengths (DEFRA, 2015): 880nm (IR), which shows the quantitative concentration of BC and 370nm (UV), which shows the presence of organic compounds (DEFRA, 2015). Whilst some differences in absolute BC values are observed between MAAP measurements at 670 nm and aethalometer measurements at 880 nm, it has been shown that measurements have excellent correlation. For example, Hyvärinen et al. (2011) found MAAP and aethalometer data to be in very good agreement, with a strong linear correlation ($y = 1.0249x$; $R^2 = 0.9729$) across the measurement range. A similarly strong correlation between measurements made by the two instruments was reported by Helin et al. (2018) (*i.e.* $R^2 = 0.91$ and 0.93), but with an offset in absolute values (*i.e.* $y = 0.72x + 21$ and $y = 0.66x + 87$). In another study, Park et al. (2012) found > 70% of sample measurements using the two techniques were within ± 2 precision intervals and > 80% within ± 3 precision intervals of each other. In this instance, no additional corrections were made to BC measurements, *e.g.* for location specific mass absorption cross-section (MAC) (Saturno et al., 2017; Helin et al., 2018).

All JOAQUIN and AURN sites were additionally instrumented with NO_x, O₃, PM_{2.5} and PM₁₀ monitors, particle counters (TSI 3783) for total particle number concentration (TNC) in the size range 7 – 1,000nm (except for Auchencorth Moss), and equipment to measure meteorological parameters as detailed in Hofman et al. (2016). In addition, SO₂ was measured at Eltham, Antwerp and Marylebone Road, and 29 different non-methane hydrocarbons (NMHCs; Automatic Hydrocarbon Network - Defra, UK, 2020) were measured at Eltham and Marylebone Road by automated Gas Chromatography (Perkin Elmer).

2.3 Campaign Period

The study focuses on the initial period of operation of the JOAQUIN network and presents the first BC results collected over the first twenty-seven-month period (01/01/2013 - 01/04/2015). A mobile monitoring campaign was conducted in parallel with this intensive measuring period, which involved the temporary deployment (*ca.* 1-month) of an identically equipped mobile unit to each of the JOAQUIN sites to facilitate instrument cross-validation (see Hofman et al., 2016 and Hama et al., 2017a for details); results of the cross-validation can be found in Figure A1 of the supplementary information.

2.4 Data Validation

All BC data were collected at a temporal resolution of 1-minute and were subjected to a ratification process whereby they were screened for errors, anomalies and instrument downtime and maintenance intervals before being averaged to 30-minute values for analysis. Analysis was conducted using the open-source *openair* tools (Carslaw, 2015) in the statistical computing software, R (R Development Core Team, 2015).

3. Results and Discussion

3.1 Overall air quality across the network sites

The concentrations of gaseous and particulate air pollution varied considerably across the measuring sites in the period 01/01/2013 - 01/04/2015. This can be seen in Figure 3, which gives the mean diurnal evolution of commonly measured pollutants averaged by hour over the sampling period at all sites, and Figure A2 and Table A1 of the Supplementary Material, which show corresponding average concentrations over the total sampling period at all sites. In brief, Marylebone Road had the highest levels of all measured pollutants, except for O₃, corresponding to its roadside location and high traffic load (Table 1). The measurements suggest that O₃ chemistry is limited by volatile organic compound (VOC) emissions at this site, which is typical of urban centers where VOC/NO_x ratios are low, causing O₃ to increase with VOC concentration but decrease with increasing NO_x (Butenhoff et al., 2015). The reverse occurs in suburban and rural areas such as Auchencorth Moss, which are NO_x-limited. Auchencorth Moss experienced the lowest levels of all pollutants except for O₃. The (sub)

urban background sites were relatively similar for all pollutants, with Antwerp showing slightly higher levels of NO_x and PM and lower levels of O₃, which is consistent with the sites' higher traffic count and road proximity. A more detailed description of the common air pollutant parameters across the study sites can be found in Section A1 of the Supplementary Material.

Figure 3: Time-averaged diurnal evolution of (a) NO, (b) NO₂, (c) O₃, (d) SO₂, (e) PM₁₀ and (f) PM_{2.5} at the measurement sites over the study period, 01/01/2013 – 01/04/2015. Data for each site have been aggregated into hourly bins, averaged over the study period and normalised to the respective mean of the dataset in order to account for the range of concentrations observed between sites. Note: NO_x measurements at Auchencorth Moss were terminated on 03/09/2013 and PM₁₀ measurements at Leicester were terminated on 23/09/2013.

3.2 BC concentration across and between the network sites

The concentration of BC averaged over the total sampling period at all sites is shown in Figure 4. As with the other air pollutants measured, the roadside site (Marylebone Road) had the highest levels of BC, with a sampling period mean of 6.6 (± 4.9) µg m⁻³. The (sub) urban

background sites of Eltham, Leicester and Amsterdam were all similar to one another, with sampling period means of $1.2 (\pm 1.2)$, $1.4 (\pm 1.3)$ and $1.0 (\pm 0.8) \mu\text{g m}^{-3}$, respectively (Table 2). Antwerp at $2.4 (\pm 1.9) \mu\text{g m}^{-3}$ had a somewhat higher mean BC concentration, corresponding to higher traffic numbers and proximity to a nearby heavily trafficked road (Table 1). Antwerp also has a relatively large diesel fleet (*e.g.* with respect to nearby Amsterdam; 61% versus 17% of vehicles, respectively), which could also be a contributing factor to somewhat higher ambient BC concentrations (European Environment Agency, 2015). Average BC measured at Auchencorth was notably lower at only $0.2 (\pm 1.9) \mu\text{g m}^{-3}$. The findings recorded here are in line with the 2015 UK annual report on BC networks: the average for 2015 was recorded at be $1.3 \mu\text{g m}^{-3}$ across all UK measurement sites, with Marylebone Road reaching at maximum of $5.1 \mu\text{g m}^{-3}$ and Dunmurry Kilmakee (a rural background site) having an average concentration of $0.3 \mu\text{g m}^{-3}$ (Butterfield et al., 2016).

Similar BC levels to those reported herein have also been recorded across other parts of Europe by various researchers. Becerril-Valle et al. (2017) took measurements around the Madrid area and at a rural location in southern Spain. The mean BC concentration recorded by the authors over a year period (2014 – 2015) at their urban traffic site ($3.70 \pm 3.73 \mu\text{g m}^{-3}$) was somewhat lower than that measured at Marylebone road in this study. Measurements made by Becerril-Valle et al. (2017) in the urban background ($2.33 \pm 2.96 \mu\text{g m}^{-3}$) were more similar to those made here in Antwerp than the other urban background locations in our study, and their measurements made under rural background conditions were considerably higher than at Auchencorth Moss ($2.61 \pm 5.04 \mu\text{g m}^{-3}$). Helin et al. (2018) reported mean BC concentration levels in Finland that were higher at a street canyon site ($1.69 \pm 1.520 \mu\text{g m}^{-3}$) than at two residential areas ($0.88 \pm 1.5 \mu\text{g m}^{-3}$ and $1.04 \pm 2.13 \mu\text{g m}^{-3}$) over the period 2015 - 2017, but still notably lower than at the roadside site of Marylebone Road in this work. The traffic site in Bern, Switzerland, studied by Reche et al. (2011) also had lower BC concentrations ($3.5 \pm 1.3 \mu\text{g m}^{-3}$) than those observed at Marylebone road herein, demonstrating the extreme levels of traffic derived pollutants at this central London site.

Figure 4: Box and whiskers plots of the 30-minute average black carbon data ($\mu\text{g m}^{-3}$) recorded at the measurement sites over the study period, 01/01/2013 – 01/04/2015. The mean point is given by grey crosshairs, the median point is given by solid grey circles, the interquartile range is given by the blue boxes and the (first and third quartile) range is given by the whiskers (grey lines) (outliers excluded for clarity). To allow direct comparison, data for the urban and suburban background sites (i.e. Eltham, Leicester, Amsterdam and Antwerp) are given on the same scale, but owing to the differences in range, data for Marylebone Road and Auchencorth Moss are given on different scales.

Table 2: Descriptive statistics for black carbon measurements at the network sites, over the period 01/01/2013 – 01/04/2015 (except where noted)

BC / $\mu\text{g m}^{-3}$	Eltham ¹	Marylebone	Leicester ²	Auchencorth	Amsterdam	Antwerp
Range	0.0 – 12.1	0.1 – 48.2	0.0 – 16.1	0.0 – 5.1	0.1 – 9.6	0.1 – 19.5
Q1	0.5	2.7	0.6	0.1	0.5	1.1
Q3	1.5	9.3	1.7	0.3	1.3	3.1
Mean	1.2	6.6	1.4	0.2	1.0	2.4
St Dev	1.2	4.9	1.3	0.2	0.8	1.9

1. BC measurements initiated on 03/04/2014

2. BC Measurements initiated on 12/04/2013

In terms of temporal evolution, BC exhibited similar patterns to NO₂ at the Amsterdam, Antwerp, Eltham and Leicester sites, with peaks at ~6 – 7 am during the weekdays (Figure 5), coinciding with traffic rush hour, a trend which is missing or diminished at the weekends, suggesting vehicle emissions are a major contributor to BC pollutant levels in these suburban and urban environments, in-line with previous similar studies (Becerril-Valle et al., 2017; Helin et al., 2018; Reche et al., 2011). The BC concentration at Marylebone road, again exhibited similar temporal behaviour to other pollutants measured at the site, *i.e.* remaining elevated all day after the early morning rush hour peak and only began to dip in the early evening (18:00). As discussed above, this trend is likely owing to the high levels of vehicular activity within the highly active London ‘tourist hotspot’. Other studies that have looked at traffic dominated sites have observed similar trends, including Helin et al. (2018) at the street canyon location included in their study and Becerril-Valle et al. (2017) at an urban traffic site.

At the sites with lower traffic intensities (*i.e.* Eltham, Leicester, Amsterdam and Auchencorth Moss), BC evening peaks were observed to occur somewhat later than those sites with larger traffic volumes (*i.e.* Marylebone Road and Antwerp). Also, at all sites, except Marylebone Road, BC evening peaks at the weekend were significantly larger than their morning counterparts, with morning peaks at some sites absent all together. This likely results from a combination of lower traffic intensities at the weekend and the contribution of non-traffic related sources (*e.g.* domestic burning activities) to ambient BC concentrations in the evening and at weekends. Looking in more detail at the evolution of the evening BC peak in the suburbs of Eltham on a Saturday across the four seasons (shown in Figure A3 of the Supplementary Material), it is clear that there is a higher ambient BC loading in the colder months at the weekend. More specifically, the average hourly peak concentration of BC on a Saturday evening during the winter (2.1 µg m⁻³) is roughly double the peak concentration measured in spring (1.0 µg m⁻³) and summer (1.1 µg m⁻³), and over two and a half times greater during the autumn (2.8 µg m⁻³). Moreover, the timing and shape of the evening peak also shifts with season, with the spring and summer evening peaks being less distinct, occurring at 21:00 hrs and 23:00 hrs, respectively, and the clear autumn and winter peaks occurring at 21:00 hrs and 19:00 hrs, respectively. Indeed, there is a distinct shift in peak timing in the winter to earlier in the evening after UK ‘daylight saving’ had taken place and in

377 coincidence with fewer daylight hours. In combination, the change in peak BC values over the
378 seasons, the timings of the peaks and their shapes and widths, indicate the occurrence of
379 increased domestic burning activities taking place around the Eltham site during the colder
380 months.

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Figure 5: Time-averaged diurnal evolution of black carbon for each day of the week for each measurement site over the study period, 01/01/2013 – 01/04/2015. From top: (a) Eltham, (b) Marylebone Road, (c) Leicester, (d) Auchencorth Moss, (e) Amsterdam and (f) Antwerp. Data for each site has been aggregated into hourly bins and averaged over the study period. Note: black carbon measurements at Eltham and Leicester were initiated on 03/04/2014 and 12/04/2013, respectively.

As can be seen in Figure 6, BC levels are notably lower at weekends (with diminished, or absent rush hour signatures), owing to reduced road traffic activities, in-line with previous literature (Barmpadimos et al., 2011; Bhugwant et al., 2000; Butterfield et al., 2016). Figure 6 also demonstrates that BC levels at each of the sites investigated here were lower during spring and summer and higher throughout autumn and winter. This is most likely a combined result of meteorological conditions and changes in local emissions. Primarily, BC levels will have been affected by reduced mixing and poor dispersion of local sources owing to colder meteorological conditions in the autumn and winter months (Singh et al., 2018; Butterfield et al., 2016). Indeed, it is well known that at colder near ground temperatures over land surfaces, the boundary layer/mixing height is reduced and so with it, is the volume into which pollutants are dispersed (Chan and Wood, 2013); to date numerous studies have shown that boundary layer height and pollutant concentrations are anti-correlated (Schäfer et al., 2006; Su et al., 2018; Xiang et al., 2019). Further, during winter months, domestic fuel burning (Cordell et al., 2016) and transport related activities taking place in urban areas are likely to be greater (Žibert and Pražnikar, 2012). For example, 5-year average road traffic statistics compiled by the UK Department for Transport (Road traffic statistics (TRA), 2020) show that vehicle use on urban roads is highest between September and December, presumably as inclement weather dissuades individuals from seeking alternative transport for local journeys (Bergstroom and Magnusson, 2003).

The high spatio-temporal variability in BC concentrations noted here is distinct and comparable to concomitant UFP and PN_1 (also shown to be important traffic-related pollutants) observations made by the same network (Hofman et al., 2016).

Figure 6: Temporal evolution of black carbon concentrations at each measurement site for (left) each hour over 24 hours, (middle) each month over 1 year and (right) each day over 1 week, over the study period, 01/01/2013 – 01/04/2015. Data for each site has been averaged over the study period and normalised to the respective mean of the dataset in order to account for the range of concentrations observed between sites. Note: black carbon measurements at Eltham and Leicester were initiated on 03/04/2014 and 12/04/2013, respectively.

To investigate potential spatio-temporal relationships for BC between pairs of study sites, the Coefficient of Determination (COD) and Spearman's rank correlation coefficient (r_s) were derived (Table 3, 4). The UK sub/urban background areas of Leicester and Eltham exhibited the highest similarity in terms of ambient BC concentration (COD = 0.32 and r_s = 0.62), followed by Amsterdam and Antwerp (COD = 0.43 and r_s = 0.68), which may be expected owing to the relative proximity of these respective pairs to one another, commonality in terms of type and magnitude of BC source and region of origin. Unsurprisingly, the lowest level of spatio-temporal correlation was between the background station at Auchencorth Moss and both the urban traffic site of Marylebone Road and the urban background site of Antwerp, where traffic density was particularly high (Table 1). In general, the high COD and low r_s statistics suggest ambient BC across northwest Europe are not particularly well related on a regional scale.

Table 3: Spearman rho correlation coefficients ($p < 0.05$) of 30-minute black carbon between pairs of measurement sites, over the period 01/01/2013 – 01/04/2015 (except where noted).

	Eltham ¹	Marylebone	Leicester ²	Auchencorth	Amsterdam	Antwerp
Eltham¹	1					
Marylebone	0.249	1				
Leicester²	0.615	0.266	1			
Auchencorth	0.251	-0.032	0.187	1		
Amsterdam	0.454	0.192	0.388	0.235	1	
Antwerp	0.453	0.359	0.366	0.156	0.684	1

1. Black carbon measurements initiated on 03/04/2014

2. Black carbon measurements initiated on 12/04/2013

Table 4: Coefficients of divergence (COD) of 30-minute black carbon between pairs of measurement sites, over the period 01/01/2013 – 01/04/2015 (except where noted).

	Eltham ¹	Marylebone	Leicester ²	Auchencorth	Amsterdam	Antwerp
Eltham¹	0					
Marylebone	0.667	0				
Leicester²	0.322	0.655	0			
Auchencorth	0.718	0.910	0.756	0		
Amsterdam	0.353	0.694	0.369	0.715	0	
Antwerp	0.446	0.521	0.421	0.831	0.430	0

1. Black carbon measurements initiated on 03/04/2014

2. Black carbon measurements initiated on 12/04/2013

In order to investigate potential contributions of regional scale BC to local levels of BC pollution, annual (2014) gridded concentration fields (Seibert et al., 1994) were calculated using 3-hour interval HYSPLIT back trajectory data (Air Resources Laboratory- HYSPLIT- Hybrid

Single Particle Lagrangian Integrated Trajectory model, 2020) and BC measurements from two sites (Figure 7). The suburban London site in Eltham was chosen for the UK and the urban background site in Amsterdam (using Rotterdam as the closest receptor site with available data) for mainland Europe.

Figure 7: Annual (2014) BC concentration field data plot using HYSPLIT smoothed and gridded back trajectories for Eltham (a) and Amsterdam (b)

The analysis shown in Figure 7 indicates that on average throughout a typical year (2014), higher ambient concentrations of BC in both London and Amsterdam are associated with air masses associated with central and eastern Europe, and for London, with the north of the UK.

When compared with the MERRA model output for annual BC surface concentrations (Figure 1), it becomes apparent that although ambient BC levels appear to be dominated by local sources (with temporal variations poorly correlating between sampling sites), European regional transport could also be an influencing factor.

3.3 Relationships between measured pollutants

The potential relationships between pollutants, including BC, measured at each site during the study period were investigated using Spearman's rank correlation matrices with hierarchical cluster analysis (Figure 8 and Table 5) and simple linear regression analyses (Figures A4 and A5 of the Supplementary Material). As expected, at each site PM_{2.5} and PM₁₀ were highly correlated, as were typical traffic pollutants such as NO₂ and total NMHCs, including aromatic species (where measured).

Figure 8: Correlation matrices to show the relationships between air pollutants at each measurement site over the study period, 01/01/2013 – 01/04/2015. The correlation between two data sets is given by the respective ellipse shape, colour and data value (Spearman's rank correlation). Dendrograms provided show the hierarchical clustering in the data.

Table 5: Spearman's rank correlation coefficients ($p < 0.05$) of black carbon versus a range of commonly measured pollutants at each site, over the period 01/01/2013 – 01/04/2015 (except where noted).

Co-pollutant	Eltham ¹	Marylebone	Leicester ²	Auchencorth	Amsterdam	Antwerp
NO ₂	0.83	0.88	0.72	0.23 ³	0.78	0.83
PM _{2.5}	0.64	0.59	0.54	0.54	0.69	0.56
PM ₁₀	0.60	0.62	0.36 ⁴	0.49	0.60	0.56
TNC	0.66	0.86	0.66	N/M	0.38	0.69
Total HC	0.76	0.79	N/M	0.30	N/M	N/M
Total Aromatics	0.83	0.83	N/M	0.40	N/M	N/M

N/M = Not measured

1. Black carbon measurements initiated on 03/04/2014

2. Black carbon measurements initiated on 12/04/2013

3. NO_x measurements terminated on 03/09/2013

4. PM₁₀ measurements terminated on 23/09/2013

Across the monitoring network, the most consistent correlation of BC occurs with NO₂ ($r_s = 0.72$ - 0.88) with the exception of the rural site at Auchencorth Moss, for which no relationship was observed ($r_s = 0.23$), most likely owing to the minimal local traffic at this site and the relatively rapid reactivity of NO₂ with respect to timescales of PM evolution during transport (Figure 7).. The BC vs NO₂ correlation was strongest at Marylebone Road ($r_s = 0.88$), which further supports local traffic being the major source of BC emissions in the study area, as this site has the highest traffic flow and closest proximity to a road. Marylebone Road and the surrounding area is particularly tourist intensive with many shops and attractions, which receive deliveries multiple times each day from heavy duty vehicles (generally fueled by diesel), a major and increasing source of PM in the urban centre (Fenger, 1999; Hao and Liu, 2016; Reche et al., 2011).

At the Marylebone Road site there was also a very strong correlation ($r_s = 0.86$) observed between BC and total ultra-fine particle number concentrations, with traffic being the major UFP source in such close proximity to a road (Hofman et al., 2016). Strong relationships between BC and TNC were also observed for Antwerp, Leicester and Eltham ($r_s = 0.69$, 0.66 and 0.66 respectively) but the association was weak at Amsterdam (0.38), presumably owing to road proximity, lower levels of traffic, and potentially, the presence of the city's Low Emissions Zone and relatively smaller diesel fleet, as discussed in Section 3.2. It should be noted however, that work by Holman et al. (2015) on LEZs in European cities noted that there was only a slight decrease in traffic related pollutant concentrations in Amsterdam between 2008 (pre-implementation) and 2010 (post-implementation). At the Amsterdam site, TNC concentrations have also been shown to be impacted by Schiphol airport, with a 34% increase in TNC observed when winds originated from the direction of Schiphol. Overall, it was estimated that the airport contributed an estimated 5.44% to TNC during the same monitoring period as that reported on here (Hofman et al 2018). TNC was not measured at Auchencorth Moss. There was also a moderate correlation between BC and $PM_{2.5}$ or PM_{10} across all sites.

BC was also well correlated with the total NMHC concentration at both Marylebone Road and Eltham where these species were measured ($r_s = 0.69$ and 0.83, respectively), and the total concentration of gaseous aromatic species (0.76 and 0.84, respectively). It is well established that hydrocarbons are major constituents of vehicle exhaust emissions (Häsänen et al., 1981) and that increased BC emissions are linked with higher aromatic content of fuel (Karavalakis et al., 2015). As such the high correlation between total NMHC/aromatic species and BC further highlights the importance of road transport as a source of BC in the study area.

3.4 Effect of wind speed and direction on BC

Average wind speed and direction data (Figure A6 in the Supplementary Material) indicate a small bias towards south westerly winds at each site with the exception of Marylebone Road. However, as can be seen from the polar plots (Figure 9) and polar annulus plots (Figure 10), the effect of wind speed and direction on BC varied considerably by site, with no clear similarities between each location, indicating prevalence of site-dependent influences.

547

548 Eltham experienced higher concentrations of BC when wind speeds were low ($< 3 \text{ ms}^{-1}$) and
549 generally from the north-east and south-east (Figure 9a), potentially owing to the proximity
550 of a major arterial road for the capital (A2, $< 1 \text{ km}$) and a railway station ($\sim 1 \text{ km}$). There may
551 also have been some contribution to the north-eastern BC signal from shipping activity
552 associated with the Port of London, with shipping previously being identified as a source of
553 BC (Comer, 2015). As shown in Figure 10a, the easterly BC signal observed at Eltham exhibited
554 a clear diurnal pattern (in-line with Figure 3), suggesting local transport activities as the major
555 source of BC (most likely the A2 road) at this suburban site. A local (Figure 9a) nocturnal
556 source of BC to the west of the site can also be seen in Figure 10a. This signal most likely
557 results from night-time domestic fuel burning activities (with some contributions from
558 transport) within the large number of houses located in the center of Eltham directly west of
559 the measurement site (Streets and Aunan, 2005).

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(a) London, Eltham

(b) London, Marylebone

(c) Leicester

(d) Auchencorth Moss

(e) Amsterdam

(f) Antwerp

Figure 9: Relationship between mean black carbon concentrations ($\mu\text{g m}^{-3}$) and wind direction and wind speed (m s^{-1}) at each measurement site over the study period, 01/01/2013 – 01/04/2015.

568 *Figure 10: Relationships between mean black carbon concentrations ($\mu\text{g m}^{-3}$) and wind*
 569 *direction by time of day at each of the measurement sites over the study period, 01/01/2013*
 570 *– 01/04/2015.*

At Marylebone Road, wind of relatively low speed ($< 2 \text{ ms}^{-1}$) from the north and north-west of the monitoring site led to the highest observed BC levels (Figure 9b), a result of several busy roads in the vicinity, including the A501 located several meters from the sample inlet with an average traffic flow of $\sim 80,200$ vehicles per day. There are also two intersections west of the site where vehicular tire and brake abrasion and engine acceleration are frequent, significantly adding to BC emissions (Reche et al., 2011). Another major influencing factor at Marylebone Road is the asymmetric street canyon in which the monitoring site resides. Formed by the surrounding buildings, such a street canyon (height-to-width ratio of ~ 0.8 (Reche et al., 2011)) can affect wind speed and direction and result in pollutant levels over double that of background concentrations (Murena and Favale, 2007). In a modelling study (Buccolieri et al., 2018), it was found that tree cover in the Marylebone Road street canyon also had an impact on pollution levels, with the vegetation affecting wind velocity and turbulence. The authors demonstrate that in the presence of the trees, perpendicular winds act to trap pollution in the Marylebone Road street canyon, decreasing dispersion and increasing pollutant concentrations (owing to recirculation zones); yet parallel winds act to lower pollution levels on the ground and increase pollution levels on upper floors of buildings where aerodynamic effects outweigh those of deposition. Unlike the other sites investigated here, there is no diurnal pattern in the polar annulus for Marylebone Road, with BC levels of roughly similar intensity during typical 'working' hours of the day (ca. 06:00 – 19:00 hrs), as was seen in Figure 6.

As shown in Figure 9c, wind speeds were relatively moderate ($< 15 \text{ ms}^{-1}$) from all directions at the Leicester site except for the north-west, which similarly correlated with higher concentrations of BC ($\sim 2 \mu\text{g m}^{-3}$). The polar annulus plot for Leicester shown in Figure 10c again demonstrates the bimodal diurnal evolution of BC at this site (as seen in Figure 6), but with higher evening/nighttime concentrations when winds derive from the north-west and south-west quadrants. Activities at East Midlands airport and Radcliffe-on-Soar coal-fired power station (including evening/nighttime power generation), which are located adjacent to one another $\sim 27 \text{ km}$ to the north-west of the city, may be contributing to these heightened BC levels in Leicester. Previous studies have illustrated the impact of aviation on ambient BC levels (Masiol and Harrison, 2014; Shirmohammadi et al., 2017) with one study (Dodson et

al., 2009) reporting that aircraft emissions contributed ~24-28% towards BC concentration at a study site in Rhode Island. Figure 9c also shows BC levels were higher when wind speeds were low, indicating a local source of BC near the Leicester site, most likely the two nearby A-roads, University Road and Welford Road, located within ~25 and 125 m of the site, respectively. Further potential contributors to ambient BC in Leicester include city centre and residential/domestic activities from the north-west and motorway traffic, with the major M1 motorway and M1/M69 interchange to the south-west of the site and the Leicester Forest East service station to the west.

As shown previously, BC levels at Auchencorth Moss were relatively low compared to other sites, however, directional patterns are nevertheless evident in the corresponding polar and polar annulus plots (Figures 9d and 10d). As can be seen in Figure 9d, levels of BC are highest at the rural background site when wind speeds were low ($< 5 \text{ ms}^{-1}$) to moderate ($< 15 \text{ ms}^{-1}$) to the east and south-east, most likely owing to the presence of local roads (e.g. A703 located ~1 km east of the site) and villages in these directions. In-line with Figure 6, the polar annulus in Figure 10d shows that concentrations of BC peaked later in the day (~ 16:00 – 22:00 hrs) when winds derived from the south-eastern quadrant, potentially a result of local domestic fuel usage during the evening.

BC concentrations at the Amsterdam site were highest when winds derived from the east and north-east (Figure 9e) and exhibited a clear bimodal signature over the average 24-hour period (Figure 10e). With Amsterdam city centre and its various arterial roads lying to the north-east of the site, it is evident that inner city anthropogenic and road transport activities are a major contributor to ambient BC concentrations in the suburbs, despite the existence of a major low emissions zone and electric tram network (Panteliadis et al., 2014). It is also likely that there is some contribution to suburban BC in Amsterdam from shipping related activities (van der Zee et al., 2012; Comer, 2015) around the Port of Amsterdam and its connecting canals, which has an annual activity of ~17,000 ship movements (Port of Amsterdam, 2018). As BC is often co-emitted with SO_2 by shipping and port related processes, source contributions from maritime activities in Amsterdam could be better elucidated in the future by combining concomitant high-time resolution BC, SO_2 and meteorology

measurements (van der Zee et al., 2012). There was no evidence from the wind field data analysis to suggest that activity at Schiphol airport (~9km from the measurement site) was a major contributor to ambient BC level in central Amsterdam.

The second highest average BC concentrations were measured in Antwerp, with greatest values when wind speeds were low ($< 5 \text{ ms}^{-1}$) and from the south and south-east (Figure 9f). As shown in Figure 9f, average BC concentrations also exhibited a bimodal diurnal pattern when winds derived from the south-eastern quadrant, with peak times aligning with typical peaks in commuter activity (*i.e.* ~07:00 and after 18:00), as also shown in Figure 6. These findings most likely arise from the presence of a major arterial road (N184) within 30 meters of the site and the surrounding city ring road, indicating road transport to be the major contributor to average BC concentration in this instance.

3.5 Contribution of BC to PM

The combination of BC and PM monitoring is particularly useful in assessing sources of ambient air pollution (Reche et al., 2011). Four of the sites employed within this investigation measured the contribution of BC to $\text{PM}_{2.5}$ (Eltham, Marylebone, Leicester and Auchencorth Moss), the remaining two sites measured the contribution of BC to TSP (Amsterdam and Antwerp). In order to investigate the temporal evolution of the contribution of BC to fine PM, BC measurements made in Eltham, Marylebone, Leicester and Auchencorth Moss were expressed as fractions of measured $\text{PM}_{2.5}$. In the absence of TSP mass concentration measurements at the Amsterdam and Antwerp sites, PM_{10} was used here as a surrogate and fractional contributions of BC to PM_{10} were determined (with the assumption that the results will comprise an upper limit, with TSP typically $> \text{PM}_{10}$ across various European environs; *e.g.* Monn et al., 1995). Across all the sites there appears to be a similar trend (Figures 11 and 12) with BC concentration positively correlating with PM mass (Figure 8), as was observed in previous studies (Yoo et al., 2011). The hourly contribution of BC to PM is largest during rush hour, with the sites of highest traffic intensity having the greatest contribution of BC to PM, *i.e.* for $\text{PM}_{2.5}$ Marylebone Road ~45%, Eltham ~18%, Leicester ~18% and Auchencorth Moss ~5% and for PM_{10} (as a surrogate for TSP) Antwerp ~11% and Amsterdam ~6%, further highlighting the importance of traffic sources to ambient BC and PM at key times of the day.

This is further reflected in the contribution of average BC to PM during each weekday (Figures 11 and 12), with the ‘weekend effect’ clearly evident across the sites, yielding a lower contribution of BC to PM when traffic volumes are lighter and anthropogenic habits associated with the weekday cycle are absent or diminished (Barmpadimos et al., 2011; Bhugwant et al., 2000). The only exception to this pattern occurs in Antwerp on Saturdays, where there is a peak in road traffic in the afternoon and late evening, resulting a higher BC to PM ratio in the afternoon.

Figure 11: Temporal evolution of the mean black carbon fraction of $PM_{2.5}$ at four measurement sites for each hour over one week (upper panel), each hour over 24 hours (lower left panel), each month over 1 year (lower middle panel) and each day over 1 week (lower right panel), over the study period, 01/01/2013 – 01/04/2015. Note: black carbon measurements at Eltham and Leicester were initiated on 03/04/2014 and 12/04/2013, respectively.

Figure 12: Temporal evolution of the mean black carbon fraction of PM_{10} (as a surrogate for TSP) at the Amsterdam and Antwerp sites for each hour over one week (upper panel), each hour over 24 hours (lower left panel), each month over 1 year (lower middle panel) and each day over 1 week (lower right panel), over the study period, 01/01/2013 – 01/04/2015.

The contribution of BC to PM_{10} (as a surrogate for TSP) was observed to increase during the winter, most likely owing to increased vehicle usage for transport and, to a minor extent (~5 – 6% for these locations; Cordell et al., 2016), increased fuel use for domestic heating (Dodson et al., 2009; Žibert and Pražnikar, 2012). In Amsterdam and Antwerp, the fractional contribution of BC to PM was lowest during spring, with increases in agriculture related secondary PM potentially a contributing factor (Twigg et al., 2015; Air Quality Expert Group, 2018). The fractional contribution of BC to $PM_{2.5}$ at the Marylebone Road, Eltham and Leicester sites was also lowest during spring, although the variation between seasons in Eltham and Leicester was less pronounced.

It is worth noting that the apparent lower contribution to particle mass observed at Antwerp (and in Amsterdam to some degree), despite the higher proportion of diesel vehicles, is due to the fact that for Antwerp and Amsterdam the contribution of BC to PM_{10} is determined,

whereas for the other sites the contribution to $PM_{2.5}$ is described. BC is composed almost entirely of particles $<1\ \mu m$ (Gong et al., 2016) and thus its contribution to PM_{10} is significantly lower than to $PM_{2.5}$.

The contribution of BC to PM during periods of ‘high pollution’ was also investigated at each site, where a ‘high pollution’ measurement is defined as an instance when either $PM_{2.5}$ exceeded $25\ \mu g\ m^{-3}$ for Eltham, Marylebone Road, Leicester and Auchencorth Moss, or when PM_{10} exceeded $40\ \mu g\ m^{-3}$ for Antwerp and Amsterdam (*i.e.* the European annual average limits under directive 2008/50/EU). Results consistently showed that BC at each site comprised a lower fraction of PM on days when pollution is considered ‘high’, with the greatest difference seen at sites with the greatest traffic volume, *i.e.* Marylebone Road and Antwerp. On average the percentage contribution of BC to $PM_{2.5}$ on high pollution days at Marylebone Road was $\sim 29\%$, rising to $\sim 40\%$ on low pollution days; in Antwerp BC contributed on average $\sim 9\%$ of PM_{10} (as a surrogate for TSP) when pollution was high and $\sim 21\%$ when pollution was low. The smallest differences were observed in Leicester and Amsterdam, with the average contribution of BC to $PM_{2.5}$ and PM_{10} (as a surrogate for TSP), respectively on high and low pollution days varying by $< 1\%$. These results imply a greater relative importance for other components of PM at traffic dominated sites when air quality deteriorates, in-line with findings by Yin and Harrison (2008), who report on a significant increase in nitrate contribution to both $PM_{2.5}$ and PM_{10} when average daily PM_{10} mass concentration exceeds $50\ \mu g\ m^{-3}$.

4. Conclusion

The capabilities of a new, permanent sub/urban background network to monitor black carbon across the North-West European air pollution hotspot have been demonstrated, with sites in Amsterdam, Antwerp, London and Leicester. This study provides an important insight into the spatio-temporal distribution of BC with results demonstrating a high degree of correlation between BC and other traffic-related pollutants, indicating that road traffic is a significant (but not exclusive) BC source in the region; other sources are likely to include domestic activities (*e.g.* heating) and non-road transport, such as aviation and shipping. The importance of road traffic sources to ambient BC concentrations, and hence the contributions of BC to total PM

across all sites, was further highlighted by the presence of clear diurnal, weekly and seasonal patterns in the data, with peaks in concentration occurring at typical rush-hour times (in the morning and evening), during the working week and during colder months. When investigating the relationship between BC and other pollutants and meteorological factors, it was clear that relationships varied by site, indicating the location of the measurement site and location and proximity of local sources, are fundamentally important factors in determining ambient BC concentration and in evaluating its impacts, *e.g.* on human exposure. Cross-site analysis further suggested that local sources are a major factor in determining ambient BC concentration, but back trajectory and model analysis highlighted the association between air mass transport across Europe contributing and higher ambient BC concentrations at sites in the UK and mainland Europe. Analysis also highlighted that BC is a significant contributor towards suburban PM mass concentration, particularly during periods of increased anthropogenic (*e.g.* vehicular) activity (*e.g.* rush-hour) and during winter months.

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