

NPL REPORT ENV 34

2018 ANNUAL REPORT FOR THE UK BLACK CARBON NETWORK

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Approved on behalf of NPLML by Richard Brown, Head of Metrology

EXECUTIVE SUMMARY

The Black Carbon Network provides data on ambient concentrations of primary particulate matter from predominantly combustion sources, using instruments that are inherently reliable and sensitive, with high time resolution. The data are a good surrogate for general primary air pollution, and are therefore much more responsive to control measures for sources such as vehicles than, for example, PM_{2.5} data. The network as currently organised also gives very clear quantification of urban and roadside increments. Observations of the main features and trends in the data are summarized below.

2018 urban annual mean Black Carbon concentrations on the Network (with the corresponding 2017 concentrations in brackets) ranged from 0.6 (0.7) $\mu\text{g.m}^{-3}$ at Glasgow Townhead to 2.8 (4.0) $\mu\text{g.m}^{-3}$ at Marylebone Road. Values between 0.1 (0.1) $\mu\text{g.m}^{-3}$ and 0.4 (0.5) $\mu\text{g.m}^{-3}$ were seen at the rural background sites Chilbolton, Detling and Auchencorth Moss. The network mean for Black Carbon concentration was 1.0 (1.1) $\mu\text{g.m}^{-3}$.

The annual mean UV component concentrations ranged from 0.0 (0.0) $\mu\text{g.m}^{-3}$ at Auchencorth Moss to 0.8 (0.9) $\mu\text{g.m}^{-3}$ at Strabane. Roadside sites showed many negative spikes in the UV component concentration, thought to be measurement artefacts. These could be caused by volatile components in fresh vehicle exhaust plumes, the internal timing of the Aethalometer measurement or use of an assumed Ångström Coefficient. The network mean for UV component concentration was 0.3 (0.3) $\mu\text{g.m}^{-3}$. The figures in brackets are again the corresponding concentrations for 2017.

The network design implemented in early 2012 allows urban and roadside increments in Black Carbon and UV component concentrations to be determined for London, Birmingham and Glasgow. In 2018, however, Birmingham Ladywood site was not operational thus only the London and Glasgow increments were calculated. The urban increment for Black Carbon was similar at both locations, (0.4 $\mu\text{g.m}^{-3}$ in London and 0.5 $\mu\text{g.m}^{-3}$ in Glasgow) while the roadside increment was related to road traffic volumes, 2.0 $\mu\text{g.m}^{-3}$ in London and 0.6 $\mu\text{g.m}^{-3}$ in Glasgow. Reductions in the roadside increment for Black Carbon have fallen in line with reductions in roadside increment for PM_{2.5} concentrations indicating similar emission sources. There was no significant urban or roadside increment in UV component concentration.

Day-of-the-week averages of the measurements show that the highest concentrations of Black Carbon were found on weekdays, with the weekends generally having lower values. Diurnal averages of Black Carbon broadly showed a commuter traffic-based signature, with the exception of Strabane, Ballymena and Dunmurry, in Northern Ireland, which showed their peak levels from 17:00hrs to 23:00hrs. This was indicative of local residential heating.

From the diurnal plots it can be seen that the main driver behind the UV component concentrations was domestic fuel use, with elevated concentrations in the evenings at the urban background and rural sites. Sites in Northern Ireland showed the largest evening effect due to the higher use of coal, wood and solid fuel for domestic heating. These sites also showed higher concentrations in the winter months and it can be seen that UV component concentrations were inversely proportional to ambient temperature. This provides extra evidence of domestic heating being a major emission source.

Comparisons between Black Carbon concentrations and Elemental Carbon concentrations showed good linear relationships between the measurements at the sites where both are measured: Chilbolton, North Kensington and Marylebone Road, with R² values of 0.85, 0.90 and 0.90 respectively. The slopes were 1.24, 1.01, and 1.03, with intercepts of -0.04, -0.03 and +0.06 $\mu\text{g/m}^3$ respectively. The data are all compatible with a single, linear relationship between the two metrics, with Black Carbon concentrations exceeding Elemental Carbon concentrations by a factor of about 1.05.

Comparisons between particulate mass concentrations and Black Carbon concentrations showed that Black Carbon made up a significant proportion of the particulate mass concentration at roadside sites. At Marylebone Road the Black Carbon concentration comprised 12% of the PM₁₀ concentration and 18% of the PM_{2.5} concentration, and the latter result is the highest decrease measured at this site (from 27% in 2017). At Birmingham Keeley Street Black Carbon formed 10% and 15% of PM₁₀ and PM_{2.5} respectively, while Glasgow High Street showed a similar proportion of BC to PM to Birmingham.

Monthly means of Black Carbon concentrations were examined over the period 2009 to 2018 to evaluate trends. Marylebone Road, Belfast Centre, North Kensington and Dunmurry have shown a significant downward trend in Black Carbon concentrations. The most dramatic downward trend is at Marylebone Road, which has shown steadily reduced Black Carbon concentrations year on year since 2011, with concentrations reducing by more than a factor of two over 10 years. This overall drop in concentration was likely to be due to the increased number of low emission (hybrid) buses in the London bus fleet and stricter emission controls on London taxis, HGVs, lorries and vans (Euro III to Euro IV). However there is evidence that this effect was reduced in 2017 and 2018 compared to previous years. Low emission buses now make up 36% of the fleet.

The Marylebone Road UV component concentration showed a statistically significant upward trend over the period 2009 to 2018. This trend should be treated with caution due to the low concentrations involved and is probably an indirect consequence of the reduced Black Carbon concentrations over the last 5 years. The Aethalometer measures the UV component by the difference between the BC and UV channel. As Black Carbon signal has fallen it has become easier to determine the small UV component signal. It is unlikely that the UV component emissions across London have risen in the last 5 years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which were not dominated by traffic, have remained stable. There was also a significant downward trend in the Dunmurry data.

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1.0 INTRODUCTION

1.1 GENERAL

The National Physical Laboratory (NPL) in partnership with the Environmental Research Group at King's College London was awarded the contract to restructure and run the UK Black Smoke Network by the Department for Environment, Food and Rural Affairs (Defra) in September 2006. The current network layout of 14 sites has been in place, with only minor site changes, since a new contract was issued by Defra in 2012. At this time the focus of the Network was changed to provide targeted monitoring of the major urban conurbations of London, Birmingham and Glasgow, plus additional sites to monitor specific emission sources. In January 2017 a new contract was issued by the Environment Agency to run the network, continuing with this structure. Since then the only site change was the closure of Birmingham Tyburn Background in March 2017 due to sale of the land. A replacement site, Birmingham Ladywood, did not make measurements in 2018. Therefore for the monitoring year 2018 there were 13 sites which recorded data.

1.2 BLACK CARBON

Black Carbon (BC) is a measure of airborne soot-like carbon (in $\mu\text{g.m}^{-3}$) based on the optical absorption of specific wavelengths by particulates collected on a filter. Ideally it is a similar metric to Elemental Carbon (EC), a measure of soot-like carbon determined by thermo-optical (chemical) techniques, though in practice the EC fraction of total carbon depends strongly on the method chosen. BC has a close relationship to the Black Smoke measure monitored by the network and its predecessors for many decades before the installation of the Aethalometers¹, though again this can be affected by the instruments and circumstances.

BC is typically formed through the incomplete combustion of fossil fuels, biofuel, and biomass, and is emitted in both anthropogenic and naturally occurring soot. Black carbon warms the planet by absorbing heat in the atmosphere and by reducing albedo (the ability to reflect sunlight) when deposited on snow and ice. Black Carbon stays in the atmosphere for periods of days to weeks.

The terminology to be used for 'Black Carbon' data continues to receive attention within, for example, the Global Atmosphere Watch aerosol special advisory group. This is mainly concerned with highlighting the assumptions used to convert optical data to mass concentration data. The term "Equivalent Black Carbon" is formally recommended for data which simply converts an aerosol absorption coefficient to a mass concentration, such as is done here. The procedures used for the Black Carbon Network are described within this report.

1.3 MEASUREMENT METHOD

1.3.1 Aethalometer instrument and data processing

Aethalometers quantify Black Carbon on filter samples based on the transmission of light through a sample. The sample is collected onto a quartz tape, and the change in absorption coefficient of the sample is measured by a single pass transmission of light through the sample, measured relative to

¹ QUINCEY, P. A relationship between Black Smoke Index and Black Carbon concentration. *Atmospheric Environment*, 2007, **41**, 7964–7968

a clean piece of filter. The system evaluates changes in two optical sensors (sample and reference), with the light source both on and off, such that independent measurements of the change in attenuation of the sample are produced for averaging periods of typically five minutes. The absorption coefficient for material added during the period, σ [m^{-1}], is calculated from the attenuation change, the filter area, and volume of the sample. This is converted to a Black Carbon concentration for the period, as a first approximation, using a mass extinction coefficient [$16.6 \text{ m}^2 \cdot \text{g}^{-1}$] chosen by the manufacturer to give a good match to Elemental Carbon. In practice this mass extinction coefficient will vary with factors such as particle size, sample composition and quantity of material already on the filter, as discussed below.

The AE-22 Aethalometers run on the Network operate at 2 wavelengths, 880 nm and 370 nm. The 880 nm wavelength is used to measure the Black Carbon (BC) concentration of the aerosol, while the 370 nm wavelength gives a measure of the “UV component” of the aerosol. At wavelengths shorter than about 400 nm, certain classes of organic compounds (such as polycyclic aromatic hydrocarbons, and also certain compounds present in tobacco smoke and smoke from wood burning) start to show strong UV absorbance. The UV component can therefore in principle be used as an indicator of wood and solid fuel emissions.

The UV component concentration presented in this report is obtained by subtracting the measured BC concentration from the concentration measured by the 370 nm source with a mass extinction coefficient of $39.5 \text{ m}^2 \cdot \text{g}^{-1}$, which is the coefficient for this wavelength corresponding to the value used at 880 nm for “ideal” black carbon material. The UV component is not a real physical or chemical material, but a parameter based on UV absorption due to the mix of organic compounds measured at this wavelength. This metric termed ‘UVPM’ is expressed in units of ‘BC Equivalent’.

It is well known that the assumption of constant mass extinction coefficient does not hold as the filter spot darkens, leading to nonlinearity in the Aethalometer response. The effect of this nonlinearity means that the Aethalometer has reduced sensitivity to black carbon at high filter tape loadings. To correct for this nonlinearity, the model developed by A Virkkula² has been used to correct for increased attenuation due to spot darkening during sampling. This uses the simple equation:

$$BC_{corrected} = (1 + k \cdot ATN) \cdot BC_{uncorrected} \quad \text{Eqn 1}$$

where ATN is the light attenuation by the filter spot, and k is a parameter determined for each filter spot such that continuity between adjacent filter spots is greatly improved. All of the Black Carbon and UV component results in this report have been corrected by this method.

1.3.2 Sampling

At all sites, ambient air was drawn into the sampling system through a standard stainless steel rain cap mounted on the end of a vertical stainless steel tube. Size selection of the sampled aerosol was made by a PM_{2.5} cyclone placed close to the inlet of the Aethalometer. All of the tubing before the cyclone is constructed from stainless steel. Sampling has been standardised across the network by using this size selective inlet before the Aethalometer, which was not possible with the Black Smoke method.

² VIRKKULA, A. et. al. A Simple Procedure for Correcting Loading Effects of Aethalometer Data. *Journal of Air and Waste Management Association*, 2007, **57**, 1214-1222

2.0 NETWORK INFRASTRUCTURE

The following sections present the design of the Network, describe its operation and the changes to the Network in 2018.

2.1 NETWORK SITES AND DESIGN

Figure 1 shows the locations of the Aethalometers during 2018. The sites are located to target the measurement of traffic emissions of Black Carbon in urban areas, and of solid fuel and biomass emissions in Northern Ireland & Cardiff.

Urban and traffic increments were targeted by having a rural background, an urban background and a roadside / kerbside siting combination across each conurbation. These site combinations are shown in Table 1. Note that Chilbolton is used as a Rural Background site for both Birmingham and London. The existence of two background sites on roughly opposite sides of London allows additional information to be gained by considering periods with different wind direction.

Conurbation	Site Name	Site Classification
Glasgow	Auchencorth Moss	Rural Background
	Glasgow Townhead	Urban Background
	Glasgow High Street	Traffic
Birmingham	Chilbolton	Rural Background
	Birmingham Ladywood	Urban Background
	Birmingham Keeley Street	Traffic
London	Chilbolton	Rural Background
	North Kensington	Urban Background
	Marylebone Road	Traffic
	Detling	Rural Background

Table 1 Sites to measure emissions of Black Carbon from traffic and urban sources

The Birmingham Tyburn Background site shut on 1st March 2017 was relocated to Birmingham Ladywood site, however the replacement site did not make any measurements in 2018.

Five other sites make up the Network. These sites measure specific non-traffic emission sources and are listed in Table 2.

Site Name	Site classification	Typical Emission Source
Belfast Centre	Urban Background	Urban background
Lisburn Kilmakee	Urban Background	Solid fuel use / Urban background
Strabane	Urban Background	Solid fuel use
Ballymena	Urban Background	Solid fuel use
Cardiff Centre	Urban Background	Urban background

Table 2 Sites to measure non-traffic related emission sources

The sites making up the Network are shown on a map in Figure 1.

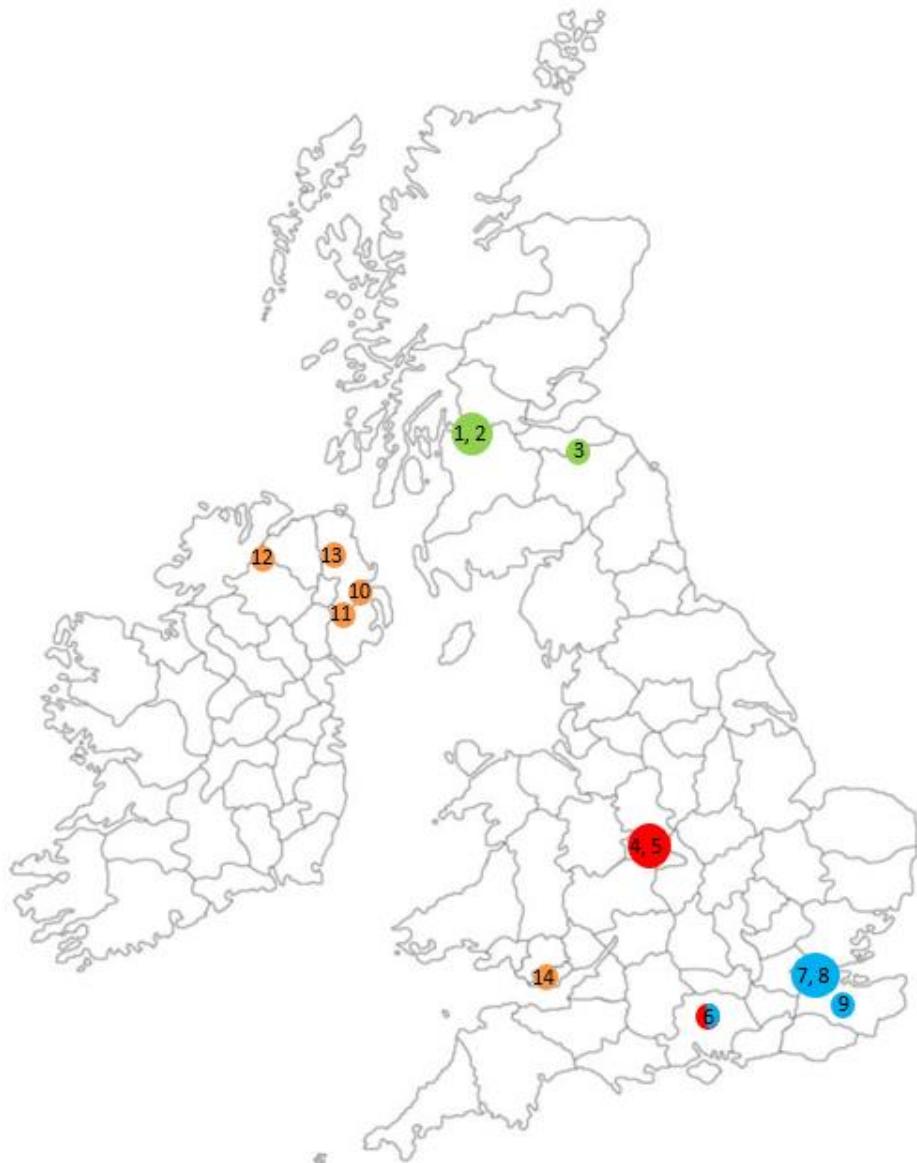


Figure 1 Sites on the BC Network during 2017

Key for Figure 1:

Emission source	Key	Site Name
Glasgow Urban Area	1	Glasgow High Street
	2	Glasgow Townhead
	3	Auchencorth Moss
Birmingham Urban Area	4	Birmingham Keeley Street
	5	Birmingham Ladywood
Birmingham Urban Area + London Urban Area	6	Chilbolton
London Urban Area	7	North Kensington
	8	Marylebone Road
	9	Detling
Solid Fuel Use	10	Belfast Centre
	11	Lisburn Kilmakee
	12	Strabane
	13	Ballymena
Domestic Emissions	14	Cardiff Centre

2.2 NETWORK OPERATION

The operation of the Network was set up to mirror that of the AURN, to include a Central Management and Control Unit (CMCU) and a Quality Assurance and Quality Control Unit (QA/QC). The Environmental Research Group at King's College London (King's) carries out the CMU activities. These activities include the routine collection of data from site, initial data validation and instrument fault finding, routine liaison with the Local Site Operators (LSO) and the Equipment Support Unit (ESU). The QA/QC activities are performed by NPL and include: site audits, inter-laboratory performance schemes, data ratification and reporting.

As the Aethalometer produces real-time continuous data, it was decided to perform remote data collection and diagnostics at each site via a modem to maximise data capture and minimise LSO costs. A summary of this activity is outlined below.

Measurements are collected from the sites on the Network on a daily basis. Measurements of Black Carbon, UV carbon, flow, raw attenuation signals and tape life since the last data collection are requested from the Aethalometer and automatically loaded into King's database. The 5 minute mean measurements are averaged to 15 minute means in line with measurements made using gaseous and particulate monitors on the AURN. A valid 15 minute measurement is only calculated where two or more valid 5 minute measurements exist in that 15 minute period. A range of sensibility checks are undertaken at this point to ensure measurements are above zero and below a maximum limit ($100 \mu\text{g m}^{-3}$); the flow data is also checked to ensure it is 4 l/min ($\pm 10\%$).

The data from each site is assessed using a range of algorithms/criteria, which determine whether the site requires a manual check; this is 'risk-based' data checking and provides a method for improving the efficiency of the manual checking procedure. The list of algorithms/criteria examine whether:

- Data warning flags have been attached to the data, either from the instrument or from the sensibility checks during processing.
- Data checking resulted in any notes or actions on the previous day.
- There are any services, local site operator visits or audits being undertaken the previous day.
- The data show unusual instability.
- The data capture over the previous 24 hours is less than 90 %.
- The site was not manually checked the previous day, in which case a manual check will be performed.

If any of these tests produce a positive result, the site is included in a list of sites to be examined manually. Where necessary, this manual validation is undertaken using the MONNET software package every working day; a screen shot of the 5 day data checking graph is shown in Figure 2. This shows the Black Carbon and UV carbon measurements and the flow measured by the instrument. Where NO_x measurements are available from the site (such as North Kensington and Marylebone Road) these are included as a method of assessing the impact of local traffic emissions. Further manual checks are made comparing the measurements between sites across the network to identify any outliers.

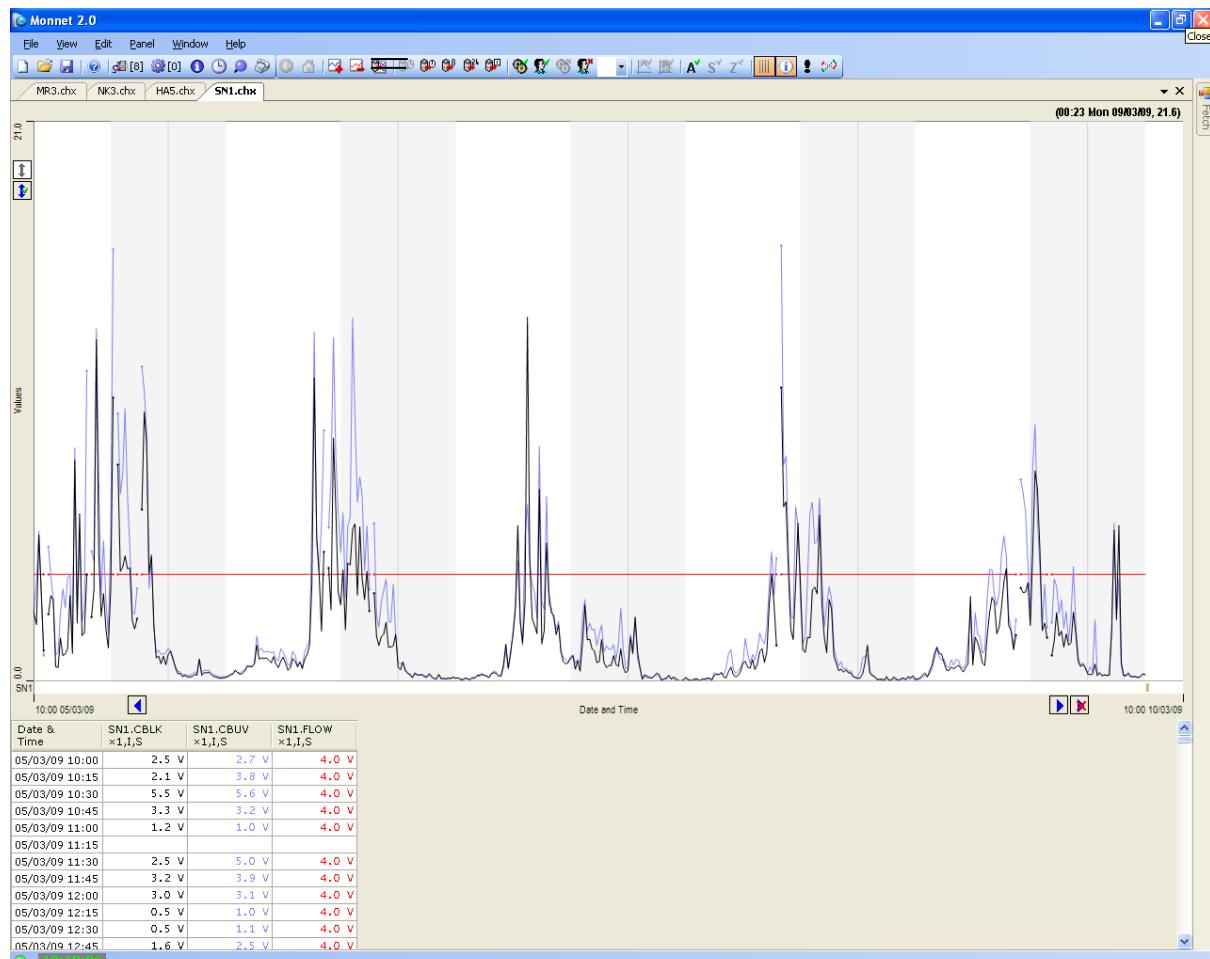


Figure 2 MONNET data checking graph

Issues raised during the manual data checking are noted in the database, this information is retained and passed to NPL to inform the ratification process. Occasionally, issues raised during data checking require an intervention from either the LSO or the ESU. If this is the case a visit request is sent to either the LSO or ESU. The reports generated from these visits are processed at King's and stored according to the site that they pertain to. The directory is mirrored to the web server and accessible via a password protected web portal for access during ratification.

3.0 QUALITY ASSURANCE AND QUALITY CONTROL (QA/QC)

Quality Assurance and Quality Control activities cover two main areas: site audits and instrument performance. The first addresses sampling issues and the second ensures the consistency and accuracy of the measurement of Black Carbon concentration.

3.1 SITE AUDITS

Table 3 gives the site audit dates and serial numbers of the Aethalometer audited.

Site	Date	Serial No.
Auchencorth Moss	19/07/2018	862
Ballymena	03/08/2018	849
Belfast	01/08/2018	864
Birmingham Keeley Street	11/07/2018	860
Birmingham Ladywood	-	-
Cardiff Centre	25/08/2018	869
Chilbolton	08/08/2018	851
Detling	10/08/2018	855
Dunmurry Kilmakee	01/08/2018	861
Glasgow High Street	18/07/2018	868
Glasgow Townhead	18/07/2018	856
Marylebone Road	09/08/2018	866
North Kensington	09/08/2018	867
Strabane	02/08/2018	848

Note: The Birmingham Ladywood site was not operational at the time of the audits

Table 3 Site Audit Visits

3.1.1 Sampler Leak Rate and Calibration of Sample Flow

The leak rate for Aethalometers was measured by simultaneously measuring the flow rate at the input and exhaust of the analyser and required the use of two calibrated flow meters.

The absolute value of the inlet flow measured during the leak test was used to calibrate the sample flow of the instrument.

Both flow meters used were calibrated against National Standards. When taking into account the repeatability of the measurements in the field, the flow inlet and exhaust flows were measured with an uncertainty of $\pm 2.5\%$, expressed with a level of confidence of 95%.

According to the manufacturer, the maximum acceptable leak rate is 20%. The highest value measured was at North Kensington, but as this was still within the manufacturer's limits, and the inlet flow rate was acceptable, no further action was taken. Black Carbon concentrations were not corrected for leak rate, but the leak rate is included in the uncertainty budget.

Table 4 gives the measured leak rates and sample flows for each site:

Site	Leak Rate, %	Indicated Flow, lpm	Inlet Flow, lpm
Auchencorth Moss	2.5	4.0	4.115
Ballymena	10.0	4.0	4.073
Belfast	12.6	4.0	3.877
Birmingham Keeley Street	11.0	4.1	4.144
Cardiff Centre	15.2	4.0	4.060
Chilbolton	8.4	4.0	3.827
Detling	-	4.0	4.199
Dunmurry Kilmakee	8.3	4.0	4.113
Glasgow High Street	16.9	4.0	4.004
Glasgow Townhead	16.2	4.0	3.906
Marylebone Road	13.6	4.0	3.872
North Kensington	19.0	4.0	4.200
Strabane	15.7	4.0	3.780

Note:

The Birmingham Ladywood site was not operational and at Detling there were no measurements due to problem with data acquisition during the audits.

Table 4 Aethalometer leak rates and sample flows

3.1.2 Instrument Performance

The best simple indication of instrument performance can be gained by examining the zero noise of the Aethalometer, as this gives an indication of the optical and electrical stability over the measurement period. This was carried out by generating nominally particle-free air using a High Efficiency Particle (HEPA) filter and examining the reported concentrations over an extended period of time. The Aethalometer concentration fell quickly to a stable value around zero, with variations due to noise in the optical system and electronics. Figure 3 shows a typical Aethalometer response to this test running on the normal time base of 5 minutes. The data are from the Chilbolton site.

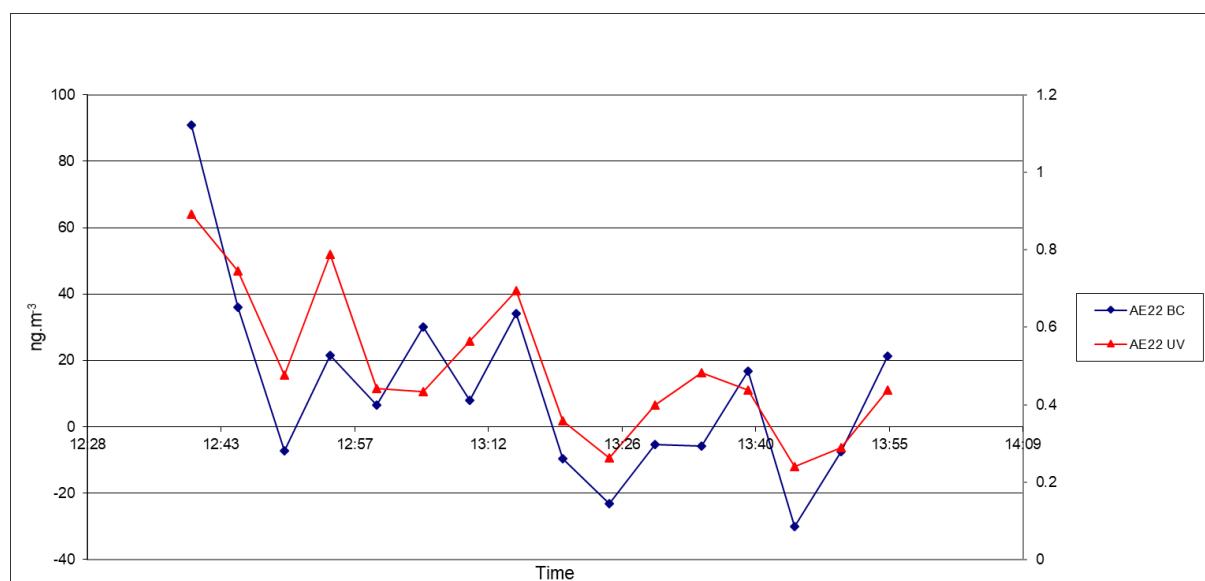


Figure 3 Chilbolton Aethalometer sampling HEPA filtered air

It should be noted that the concentrations normally reported by the Network are in $\mu\text{g.m}^{-3}$ and the above concentrations are in ng.m^{-3} . The zero noise was calculated as the standard deviation of the recorded concentrations multiplied by the student t-factor for the number of measurements. The results for each site are given in Table 5.

Site	BC noise, $\mu\text{g.m}^{-3}$	UV Channel noise, $\mu\text{g.m}^{-3}$	Annual Average BC, $\mu\text{g.m}^{-3}$
Auchencorth	0.02	0.01	0.13
Ballymena	0.04	0.02	0.78
Belfast	0.12	0.05	1.15
Birmingham Keeley Street	0.04	0.11	1.77
Cardiff	0.01	0.00	0.69
Chilbolton	0.02	0.01	0.41
Detling	0.06	0.04	0.44
Dunmurry	0.04	0.01	0.73
Glasgow High St	0.04	0.02	1.23
Glasgow Townhead	0.07	0.05	0.64
Marylebone Road	0.07	0.04	2.84
North Kensington	0.13	0.22	0.77
Strabane	0.04	0.02	1.00

Note: There are no data for Birmingham Ladywood as the site was not operational in 2018

Table 5 Hour-equivalent Zero Noise of BC and UV component channels

From previous experience, values at or below $1.0 \mu\text{g.m}^{-3}$ are considered acceptable and the results for all sites fall well within this limit. The noise value is expected to be higher for the audit test than is applicable in normal operation, due to the short averaging times, which mean that short-term humidity changes caused by air conditioning are more apparent. It should be noted that the UV Channel in Table 5 is not the UV component concentration, but the result taken directly from the UV channel. Section 1.3.1 gives a description of how the UV component is calculated.

QA/QC methods for Aethalometers are being developed internationally (see section 7.2 for more details), and are continually being reviewed and improved for the network.

4.0 MEASUREMENT UNCERTAINTY

4.1 SAMPLE VOLUME

From measurements at the site audit the sample volume can be determined, on average, with an uncertainty of $\pm 13.9\%$, expressed with a level of confidence of 95%. Included in this uncertainty are contributions from flow rate accuracy, repeatability, drift and leaks. The leakage is the dominant term.

The leak rate was not used to correct the results, but was included as an uncertainty. For the uncertainty calculation, the average value of leak rate determined in the 2018 audits was used. As leak rate is considered to have a rectangular distribution, its contribution to the standard uncertainty in sample volume was 6.2%.

4.2 MEASUREMENT OF ABSORPTION

The Aethalometer measurement does not depend on any absolute calibration of the detectors' response signals, but instead relies upon their ability to determine very small relative changes in optical transmission. Determining the zero noise of the system gives relevant information on the instrument's ability to measure small changes in optical transmission. Results from the HEPA filter zero noise tests show that the stability of the optical / electrical system was approximately $\pm 0.05 \mu\text{g.m}^{-3}$ for hourly means, compared to the network mean concentration of $0.97 \mu\text{g.m}^{-3}$. Converting this into a standard uncertainty represents an average contribution of 5.5 %.

4.3 CORRECTION FOR SPOT DARKENING

The Virkkula² model was used to correct the measured concentrations to account for the nonlinearity of Aethalometer Black Carbon and UV component measurements with filter loading. This effect and its correction introduces an uncertainty into the measurements. At most sites the correction can be seen to work well on the 15-minute data, in that there was minimal discontinuity when the spot location changes, and the associated uncertainty was considered to be small compared to other components. At sites where the concentration was changing quickly, such as Marylebone Road, this uncertainty in the 15-minute data can become significant although this was decreased when hourly mean concentrations were calculated.

The differences between using individual spot corrections versus seasonal or monthly corrections have been examined, with the conclusion that individual spot correction is the most suitable method.

The uncertainty due to the spot darkening cannot easily be directly determined and has not been included in the overall measurement uncertainty.

4.4 INDICATIVE OVERALL MEASUREMENT UNCERTAINTY

When the contributions from sample volume and optical /electrical stability were combined, the overall average measurement uncertainty for hourly Black Carbon concentrations was 17.7%, expressed with a level of confidence of 95%. The only source of uncertainty in the overall measurement uncertainty that reduces when producing longer term averages from the hourly data is the zero noise.

The overall measurement uncertainties for different averaging periods is given below, expressed with a level of confidence of 95%:

Hourly	17.7%
Monthly	13.9%
Yearly	13.9%

Indicative black carbon uncertainties year-on-year are consistent and compare well with the EU requirement of 25% for particulate mass concentration measurements.

This is an indicative measurement uncertainty for the Aethalometer method, using the conventional mass extinction coefficient $16.6 \text{ m}^2 \cdot \text{g}^{-1}$ and was calculated from the results of the 2018 audit data. The site-specific overall measurement uncertainty may differ from this value, and any effect from the spot darkening correction will be additional.

5.0 RESULTS

The concentration data for 2018 are presented in the following sections as time series graphs, summary graphs and tables of the annual mean concentration and data capture.

All of the Black Carbon and UV component data were corrected for spot darkening using the Virkkula method².

The hourly data set for Black Carbon and UV component concentrations can be downloaded from Defra's UK-AIR: Air Information Resource Web Pages, specifically the 'data selector tool' found at:

<http://uk-air.defra.gov.uk/>

5.1 TIME SERIES

The following sections present time series graphs of the Black Carbon and UV component concentrations.

5.1.1 Black Carbon

The following charts show the Black Carbon concentrations measured in 2018. The time resolution of the measurements is hourly. Data has been split into regions of the UK for presentation purposes. The maximum y-axis on these charts has been set to 25 $\mu\text{g.m}^{-3}$ to enable easy comparison between charts, except for Figure 8 which shows concentrations measured at rural locations.

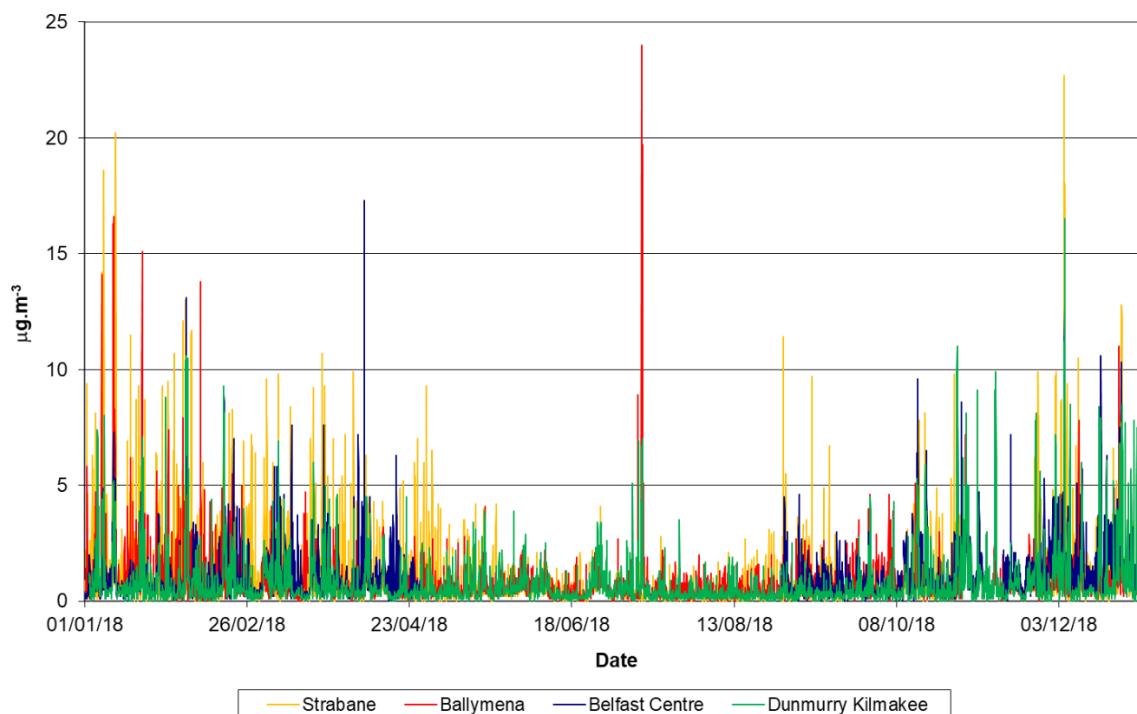


Figure 4 Black Carbon concentrations during 2018 in Northern Ireland

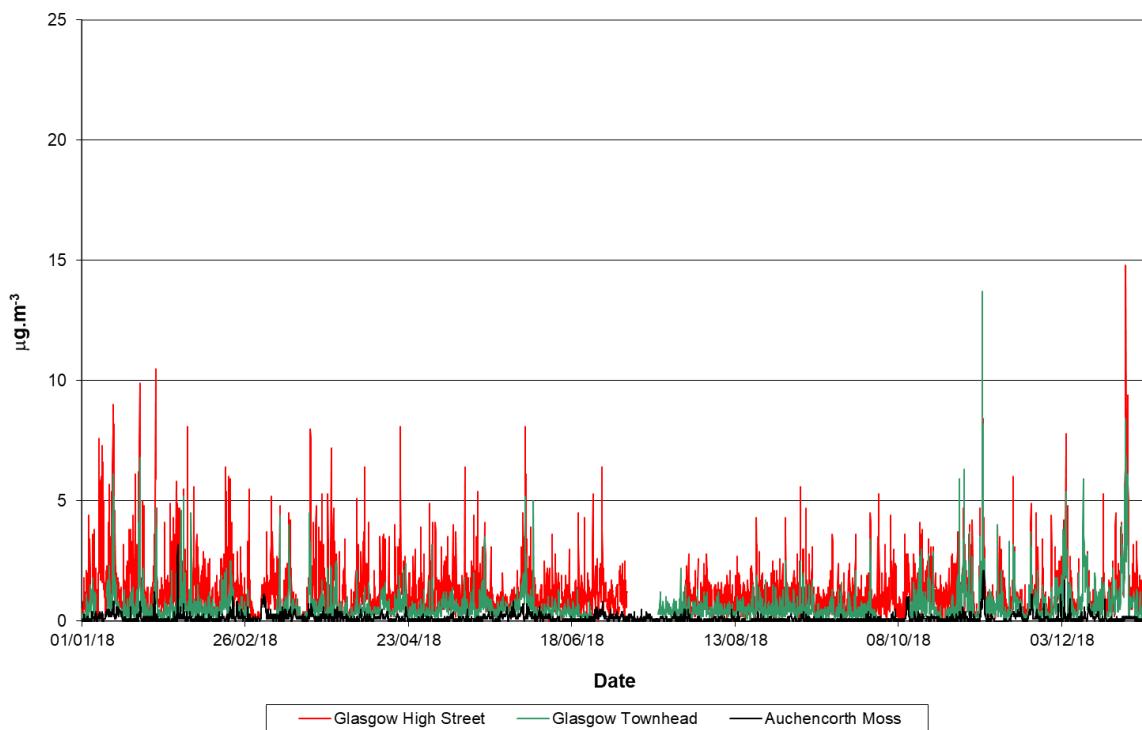


Figure 5 Black Carbon concentrations during 2018 in Scotland

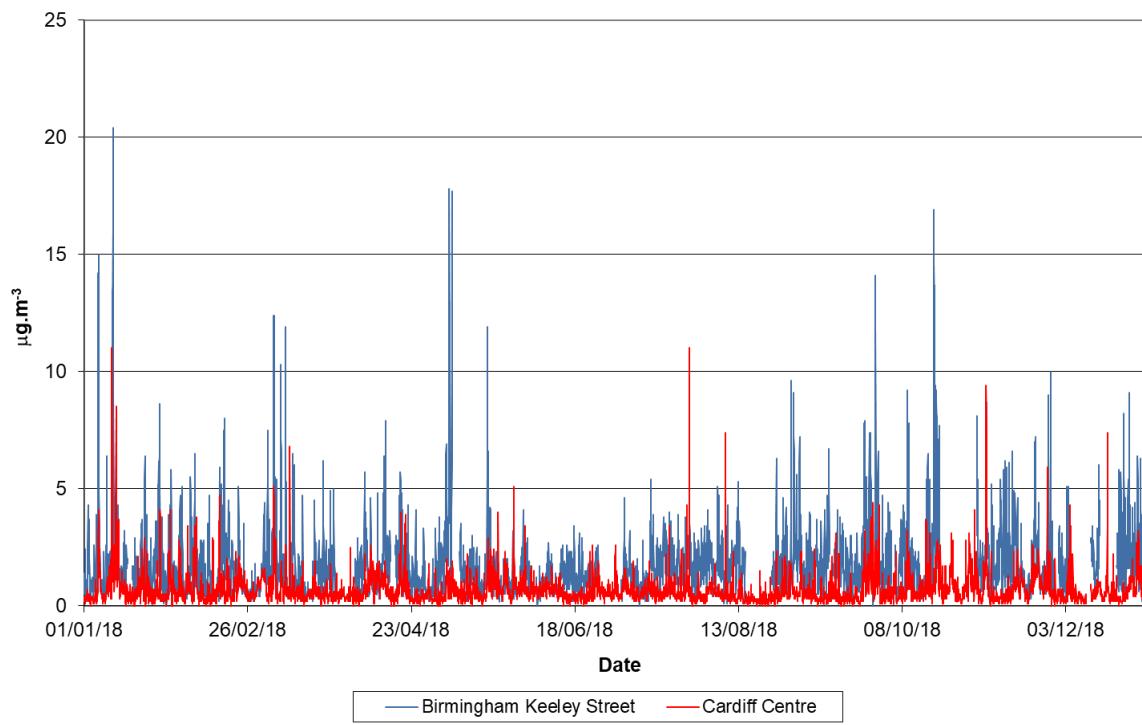


Figure 6 Black Carbon concentrations during 2018 in Wales and the Midlands

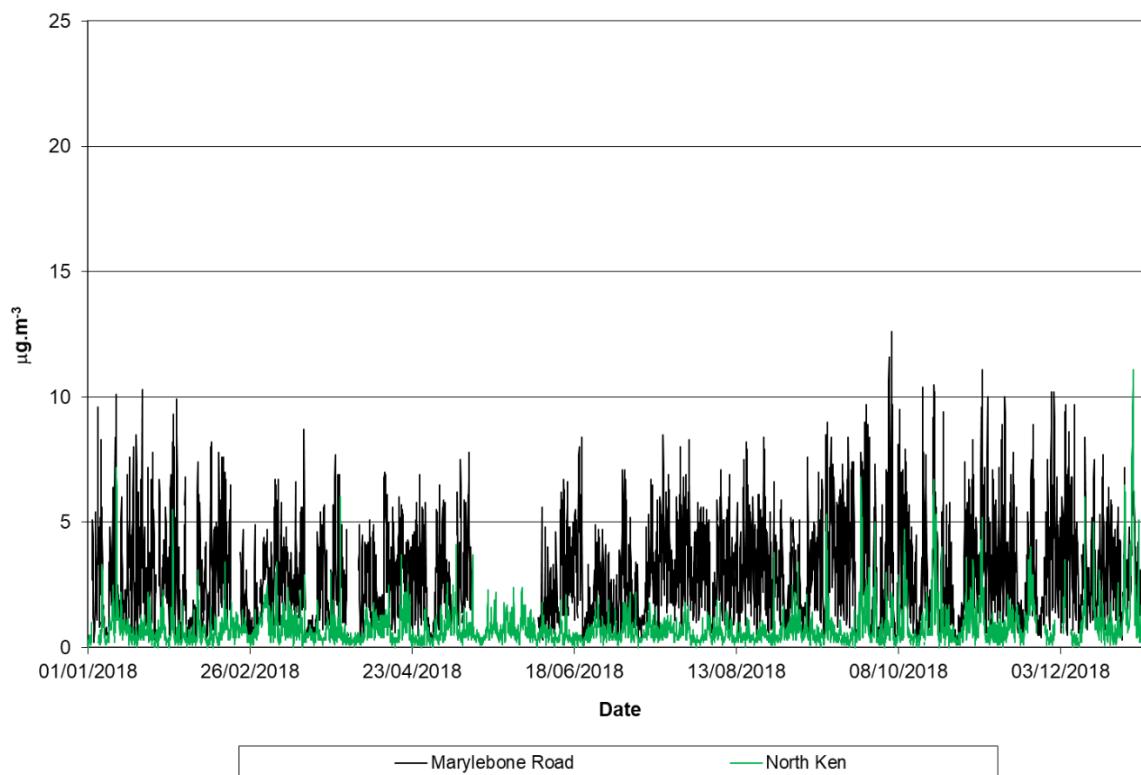


Figure 7 Black Carbon concentrations during 2018 in London

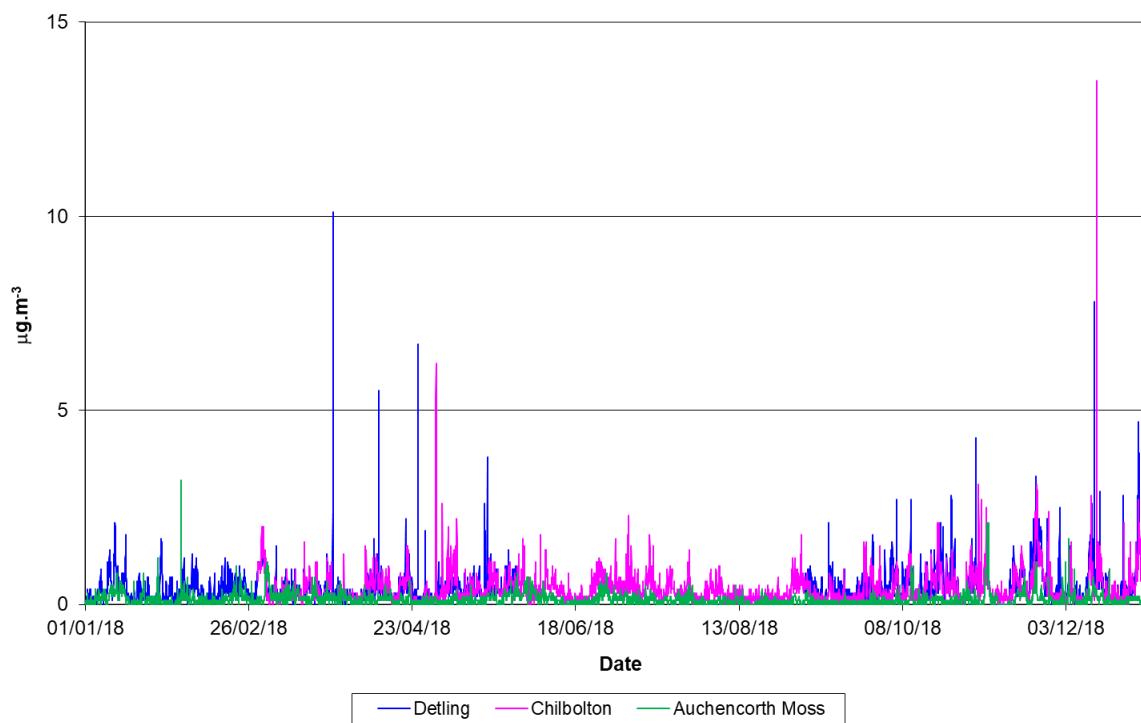


Figure 8 Black Carbon concentrations during 2018 at Rural Locations

As seen in previous years, Northern Irish sites generally measured increased concentrations during the colder months of January, February, March, October, November and December.

5.1.2 UV component

The following charts show the UV component concentrations measured in 2018. The time resolution of the measurements is hourly. Data has been split into regions of the UK for presentation purposes. The y-axis on the UV component time series graphs have not been fixed to the same value for every chart, because the UV component is much more dependent on local site-specific conditions.

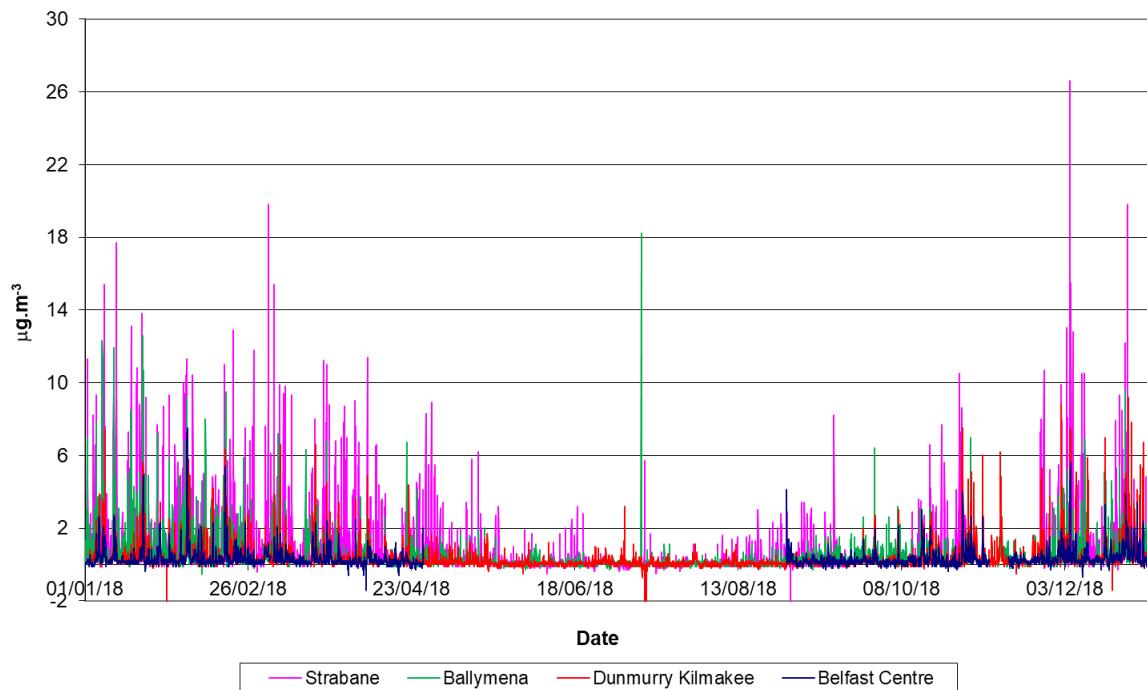


Figure 9 UV component concentrations during 2018 in Northern Ireland

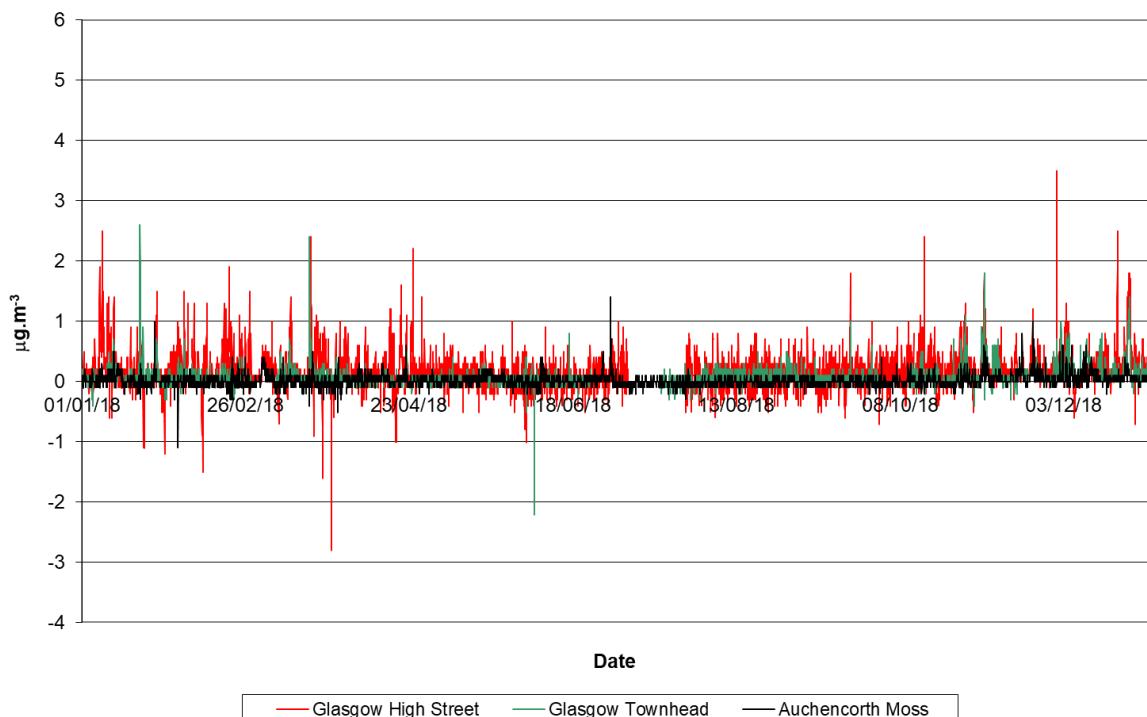


Figure 10 UV component concentrations during 2018 in Scotland

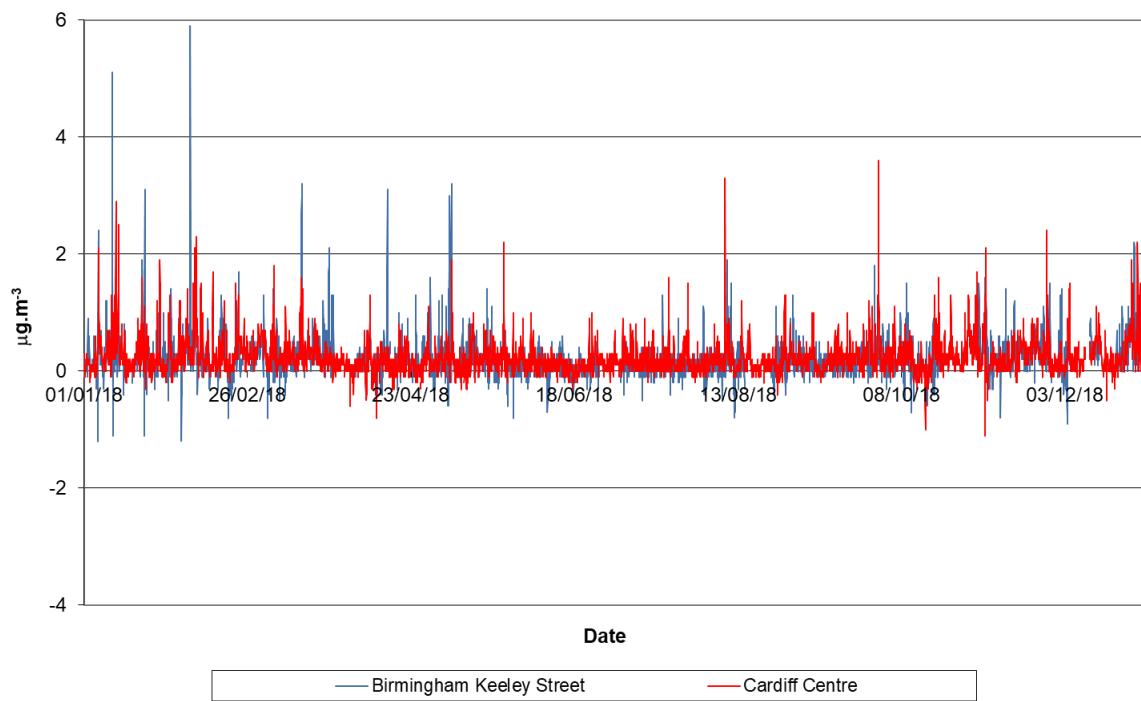


Figure 11 UV component concentrations during 2018 in Wales and the Midlands

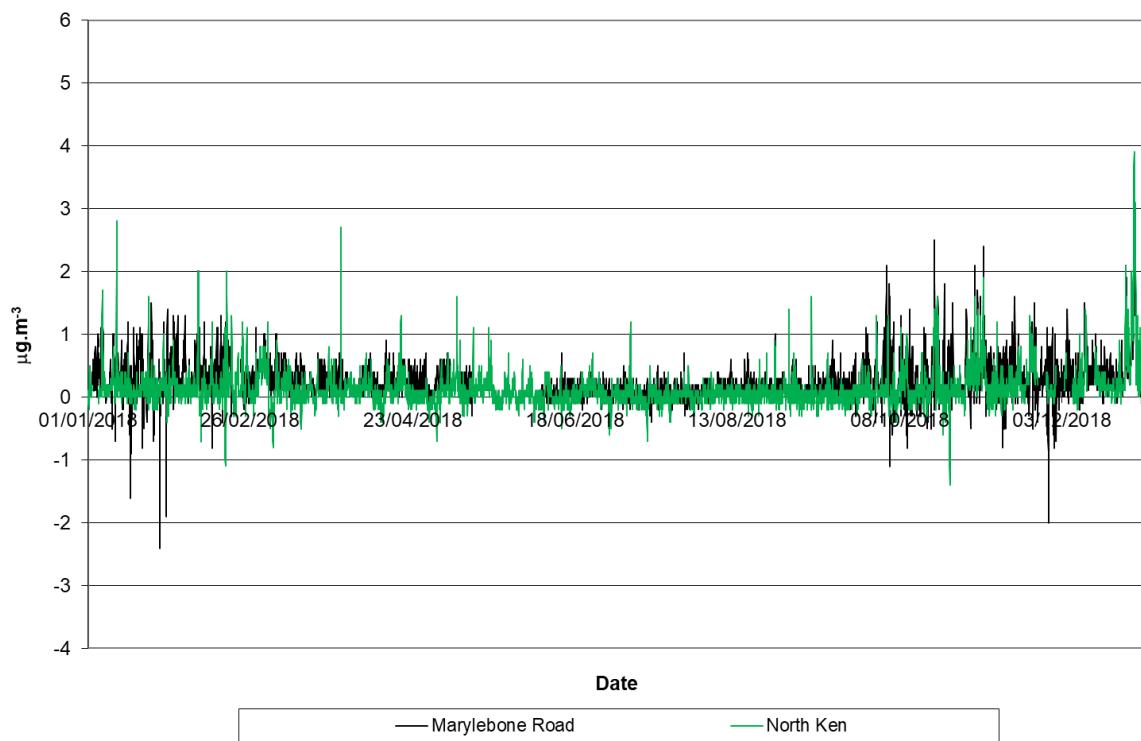


Figure 12 UV component concentrations during 2018 in London

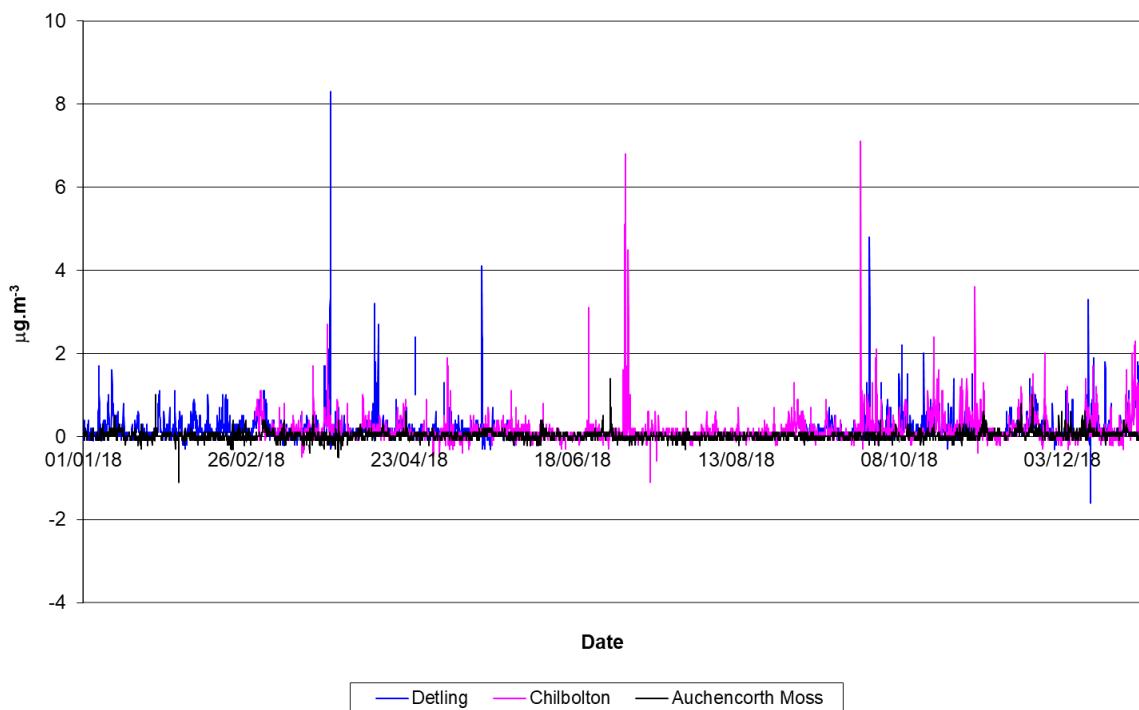


Figure 13 UV component concentrations during 2018 at Rural Locations

The Northern Irish sites measured increased UV component concentrations during the cold periods in January, February, March, October, November and December (Figure 9). Evidence from the UV component concentrations at Strabane in 2018 and previous years suggests that the heating seasons run from January to early June and from early September to the end of the year. However, elevated UV component concentrations are seen during July and August which suggests significant emissions of UV absorbing particulate mass are still present, either from solid fuel or wood burning. In Ballymena the UV component concentrations show a shorter heating season, finishing in early April and starting again in mid-October.

Monthly averages for 2017 and 2018 at these sites are shown in Figure 14. In general, data for both sites follow the same pattern (U-shape) in 2017 and 2018 with the lowest level of UV component concentrations in July.

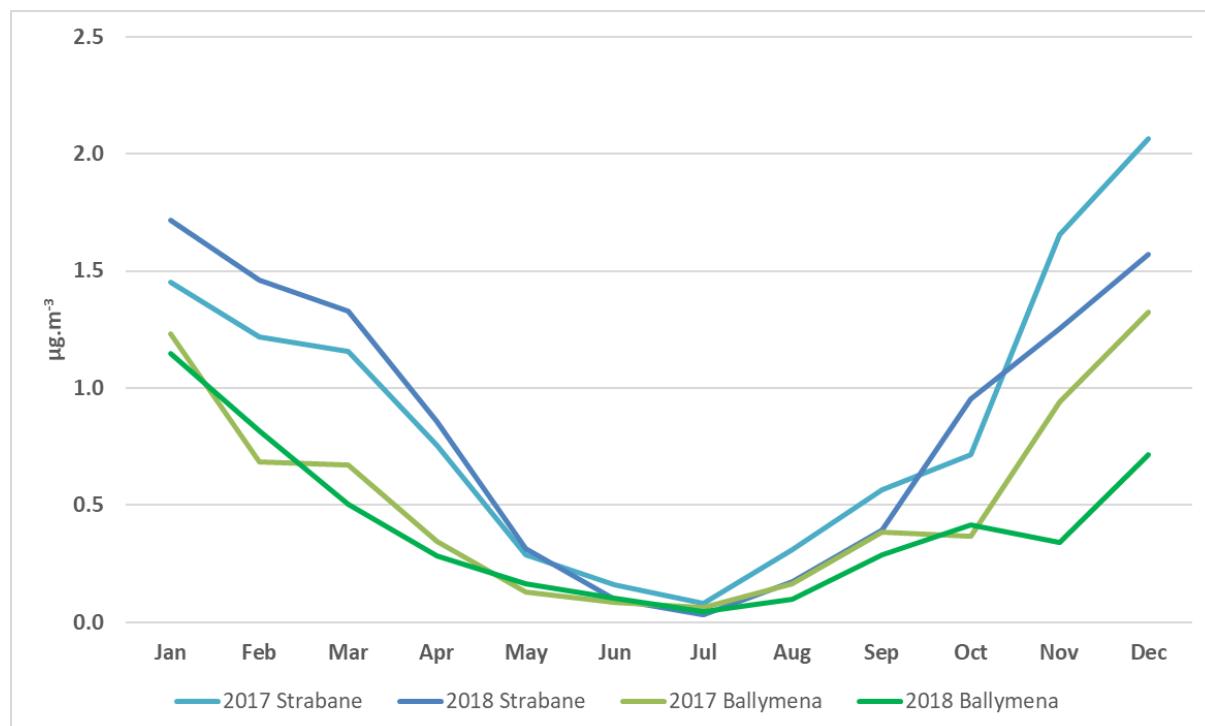


Figure 14 UV Component concentrations at Strabane and Ballymena, shown as monthly averages for 2017 and 2018.

The cause of the very short-term negative concentration spikes in the “UV component”, especially at roadside sites, is not clear. It may be due to the semi-volatile nature of the aromatic organic species that adsorb at the 370 nm wavelength. Combustion exhaust streams may contain filterable particles at high concentrations together with semi-volatile UV-active material that will be temporarily retained on the filter tape leading to a distinct increase in UV absorption. Over time these organic species evaporate from the tape and reduce the enhanced UV adsorption. If the effect of evaporation is greater than that of newly deposited material, negative UV component concentrations would be seen.

Another possible reason for positive and negative spikes in roadside data is the internal timing of the measurement process within the Aethalometer. The UV Channel reading is made around 20 seconds after the Black Carbon channel reading. If concentrations are changing rapidly, the subtraction of the Black Carbon concentration from the “UV” concentration could give misleading results.

Also, the UVPM calculation assumes an Ångström Coefficient of 1.0 for the wavelength-dependent absorption of freshly emitted Black Carbon from traffic sources. Fuller *et al*³ estimate the true value to be close to 0.96, which could cause negative UVPM when traffic emissions are dominant.

These three effects will be most prevalent at Marylebone Road due to the closeness of the inlet to the kerb, high traffic flow and predominantly diesel based exhaust signatures from HGVs, buses and taxis. A similar but smaller response was also found at Birmingham Keeley Street and Glasgow High Street, both of which have high traffic flows but less diesel based exhaust emissions than Marylebone Road.

3 FULLER, G. W. et. al. Contribution of wood burning to PM₁₀ in London. *Atmospheric Environment*, 2014, **87**, 87-94.

5.1.3 Pollution events captured by the network

Some specific air pollution events that occurred in 2018 were also seen in the data. In January, February and March, BC concentrations were elevated by cold, calm and settled conditions. One example from Northern Ireland is shown in Figure 15.

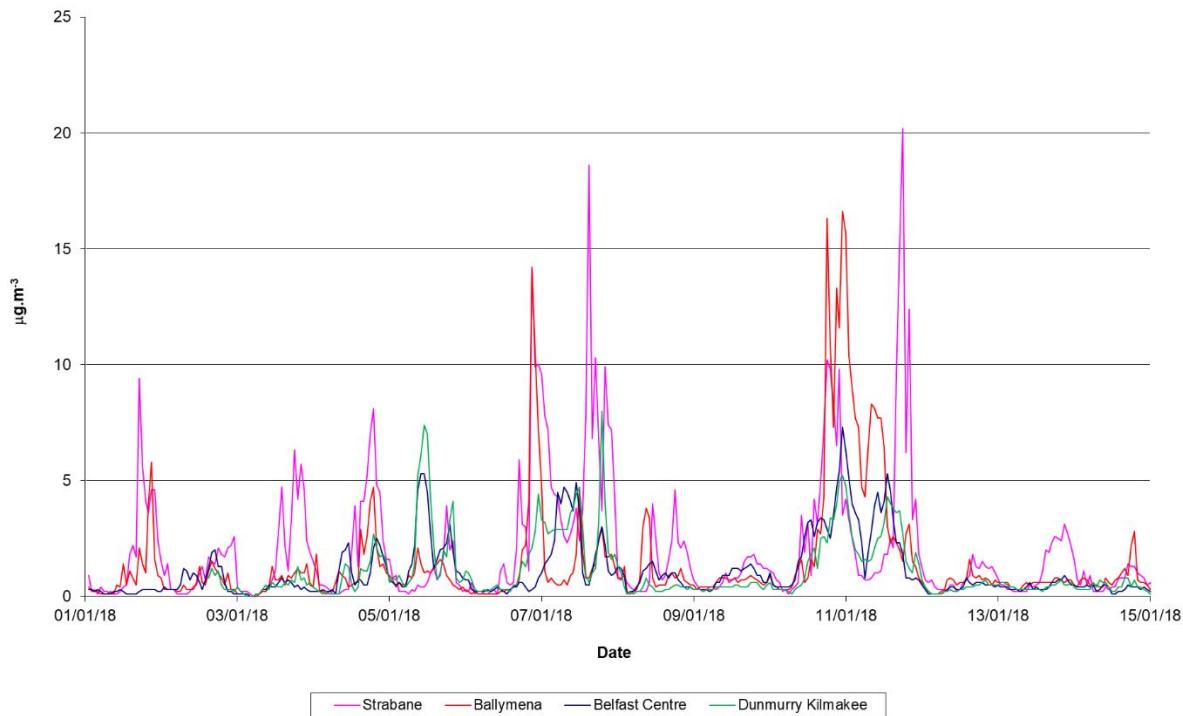


Figure 15 Black Carbon concentrations in Northern Ireland in January 2018.

This events were also seen in the UV component data, which is an indicator for wood burning. This suggests a significant contribution from domestic heating (Figure 16).

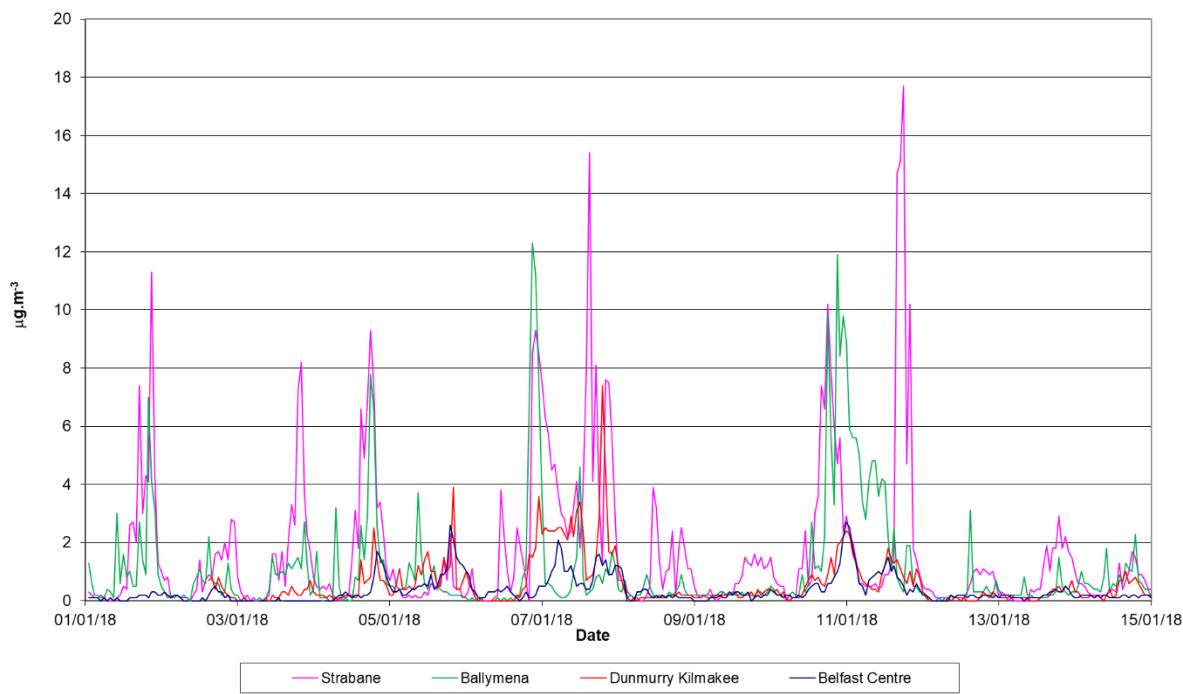


Figure 16 UV component concentrations in Northern Ireland in January 2018.

A similar effect on a smaller scale was observed between Christmas and New Year. At North Kensington and Detling both the BC and UV component concentrations recorded during this period were significantly higher than usually measured (Figure 17 and Figure 18).

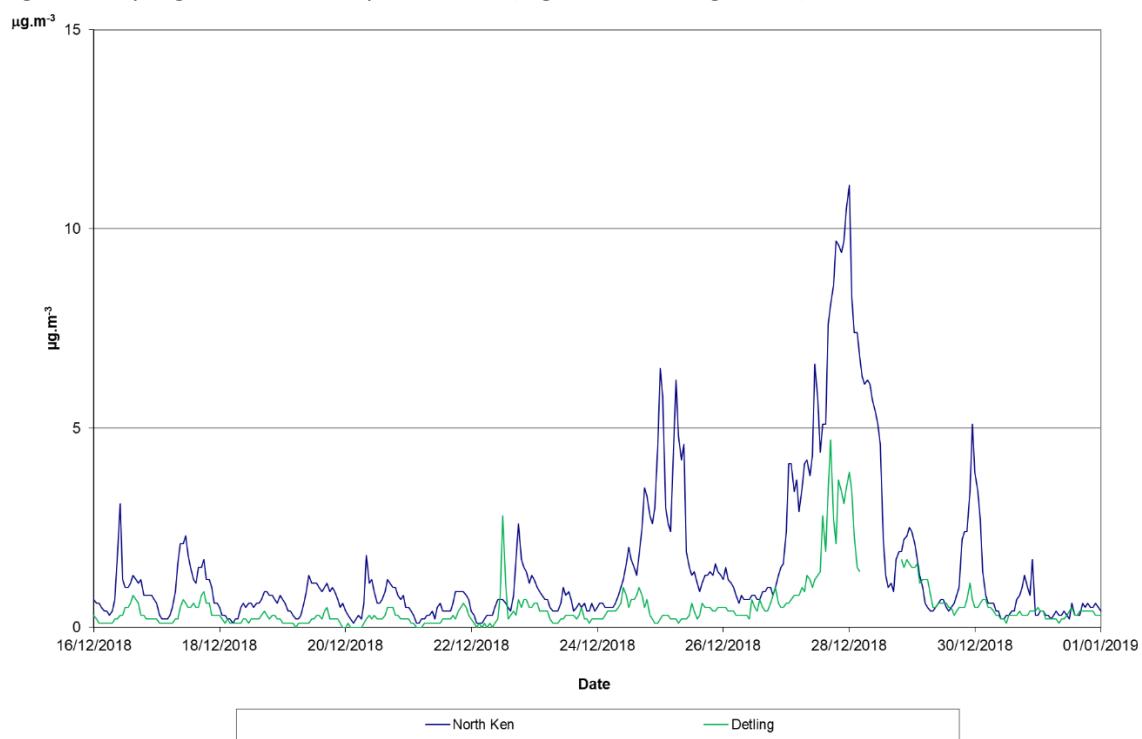


Figure 17 Black Carbon concentrations at North Kensington and Detling sites from 16 Dec 2018 to 01-Jan 2019.

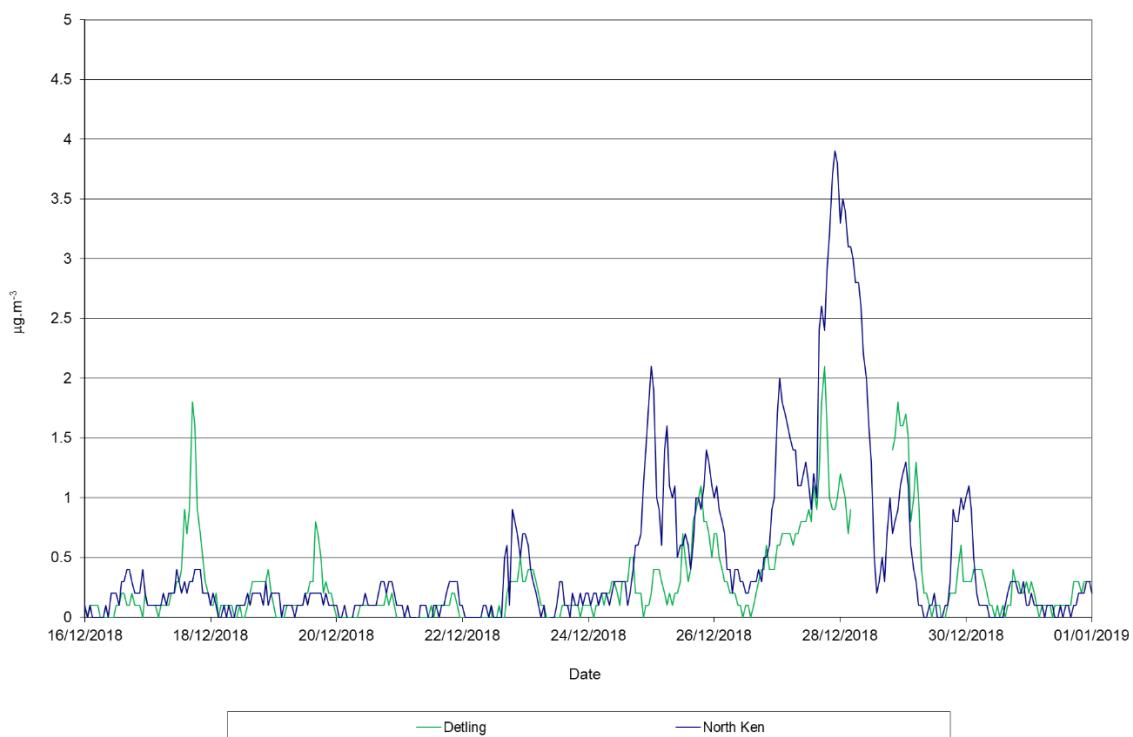


Figure 18 UV component concentrations at North Kensington and Detling sites from 16 Dec 2018 to 01 Jan 2019.

Bonfire night on 5th November did not have a widespread effect on the network, but some evidence for a rise in concentrations was seen at North Kensington (Figure 19). In general, concentrations at most of the stations within the Network were not as high as in previous years during this period.

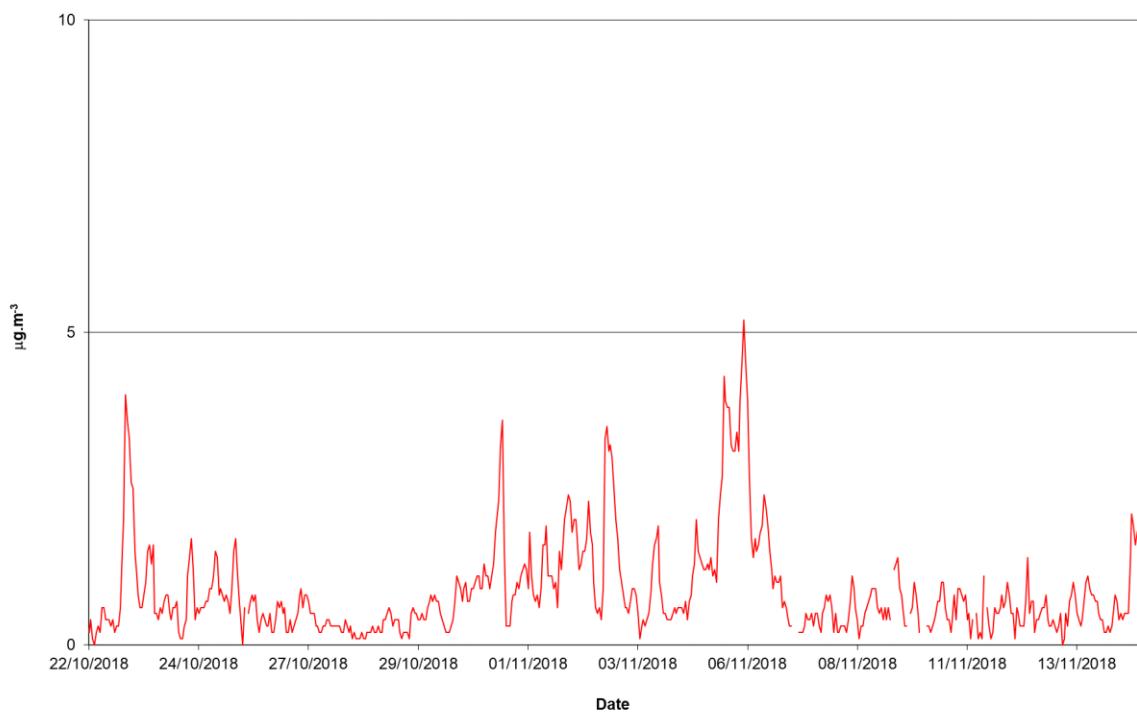


Figure 19 Black Carbon concentrations at North Kensington from 22 Oct to 14 Nov, with the reported pollution peak due to contribution from bonefires.

Some one-off, localised events were captured in the data such as the 12th July Bank Holiday in Northern Ireland. Bonfires associated with this, which took place on Thursday July 12th, were clearly seen at the Ballymena and Dunmurry Kilmakee sites (Figure 20).

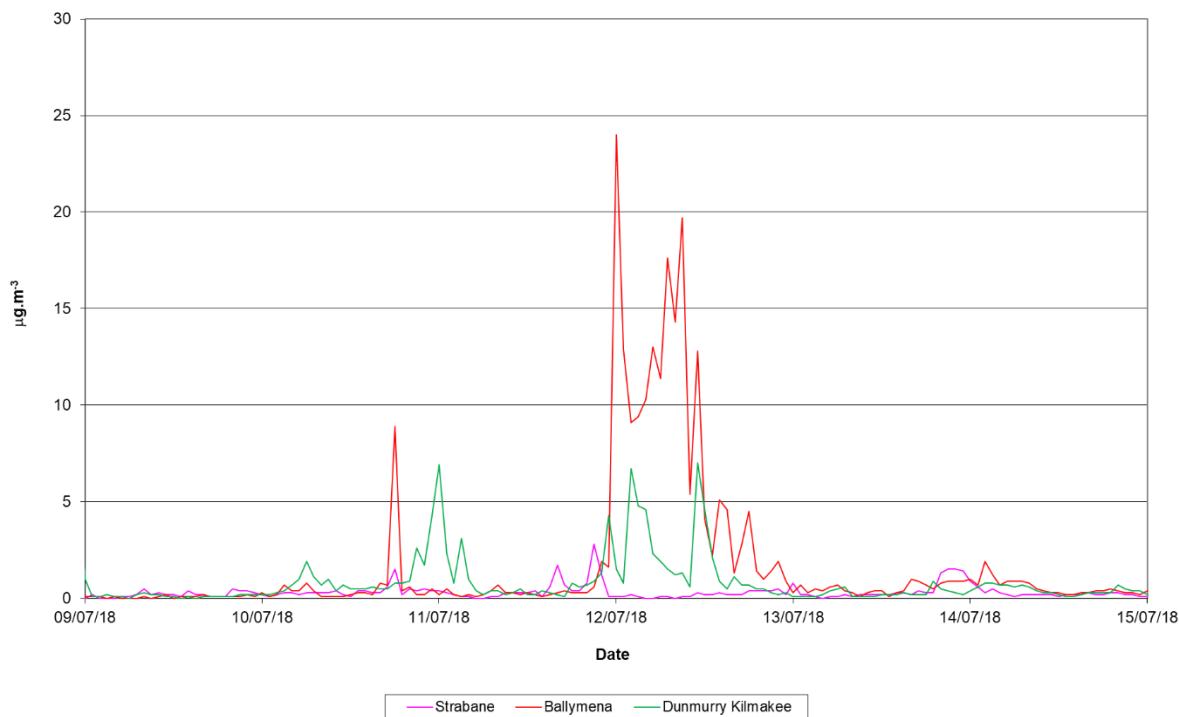


Figure 20 Black Carbon concentrations in Northern Ireland from 9 Jul to 15 Jul highlighting the contribution from bonfires.

There is no data available from the Belfast site for this period due to a disconnected sampling line which resulted in cabin air being sampled instead of the ambient air.

5.2 AVERAGES AND DATA CAPTURE

The following sections present the annual average Black Carbon and UV component concentrations along with the data capture statistics.

5.2.1 Black Carbon

Table 6 gives the annual mean for each site for 2018.

Site	Mean concentration $\mu\text{g}\cdot\text{m}^{-3}$
Auchencorth Moss	0.1
Ballymena	0.8
Belfast Centre	1.2
Birmingham Ladywood	-
Birmingham Keeley Street	1.8
Cardiff Centre	0.7
Chilbolton	0.4
Detling	0.4
Dunmurry Kilmakee	0.7
Glasgow High Street	1.2
Glasgow Townhead	0.6
Marylebone Road	2.8
North Kensington	0.8
Strabane	1.0

Note: Birmingham Ladywood site was not operational in 2018.

Table 6 Annual Mean Black Carbon Concentrations for 2018

The annual mean concentrations are presented as a bar graph (Figure 21) to aid the comparison of sites:

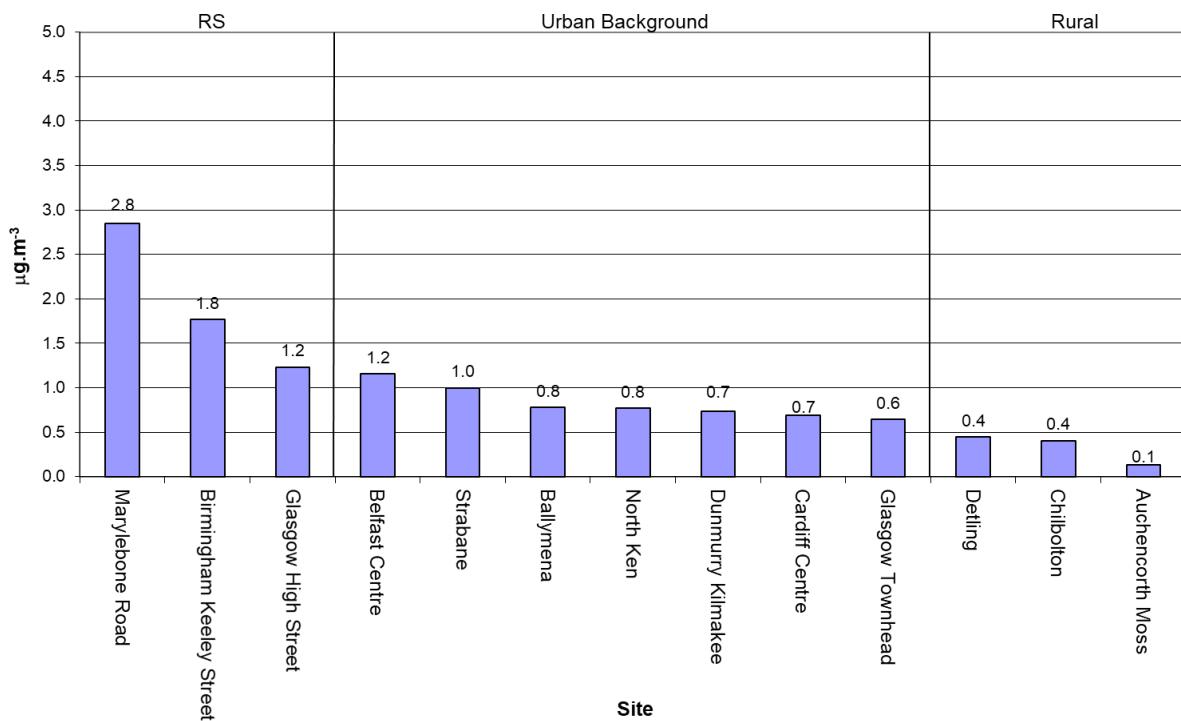


Figure 21 Annual Mean Black Carbon Concentrations for 2018

It should be noted that data from Belfast Centre site from 27th April until 29th August was not included due to a disconnected sampling line (causing the sampling of cabin air instead of ambient air). It was plugged back in during NPL's audit on 29th August, a day after the nearby clothing store caught fire. Thus, the yearly average concentration at Belfast Centre may not be properly representative.

Black Carbon concentration increments between rural, urban background and roadside sites for London and Glasgow have been analysed from periods when all the sites were producing valid data, and are shown in Table 7.

Conurbation	Increment, $\mu\text{g.m}^{-3}$	
	Urban	Roadside
London	0.4	2.0
Glasgow	0.5	0.6

Table 7 **Increments in Black Carbon concentrations between rural, background and roadside sites in 2018**

It can be seen that the urban increments for London and Glasgow were similar. The roadside increment for London was significantly larger than that for Glasgow. The site layouts are likely to be a significant contributing factor to this. At Marylebone Road the site is located about 1 m from traffic, whereas at Glasgow High Street the site is 5.5 m from traffic. Marylebone Road also has more of a street canyon effect, as the building facades are higher on average than the Glasgow site. However, the main difference is the much larger traffic flow and different vehicle profile of the Marylebone Road site compared with the Glasgow High Street roadside site. The Birmingham Ladywood site was not operational in 2018 and there is no data for this site to calculate urban and roadside increments for Birmingham. Nevertheless, Highways Agency traffic count data for 2018 are given for all three roads passing the monitoring sites as a comparison (Table 8).

Road (Count Point ID)	Motor cycles	Cars Taxis	Buses Coaches	All HGVs	All Motor Vehicles
Marylebone Road (27236)	3318	51985	2771	2605	72156
Birmingham Keeley Street (27736)	315	35993	84	2921	45991
Glasgow High Street (10821)	41	12449	166	406	15609
Ratio London to Birmingham	10.5	1.4	33.0	0.9	1.6
Ratio London to Glasgow	80.9	4.2	16.7	6.4	4.6
Ratio Birmingham to Glasgow	7.7	2.9	0.5	7.2	2.9

Table 8 **2018 Average daily traffic count data for Marylebone (London), Keeley Street (Birmingham) and Glasgow High Street Roadside sites**

The Marylebone Road roadside increment in Black Carbon concentration in 2018 was a factor of 3.3 higher than the Glasgow High Street roadside increment. This is somewhat lower than the ratio of all motor vehicles between the sites, a factor of 4.6. Although the increment is not as large as might be expected from the ratio of buses / coaches between the sites, this could reflect the cleaner fleet in London. Changes in emissions from London buses and taxis are discussed further in section 5.5.1.

Figure 22 shows how the urban and roadside increments in London have changed over the period 2012 to 2018. The average urban increment is roughly stable, with increases during the cold periods indicating the contribution from domestic heating. The roadside increment for London has clearly dropped steadily over the period. It should be noted that increment calculations are only possible for periods where parallel measurements are gathered from all London sites including two rural sites: Chilbolton and Detling. Both of the latter had issues with leaks in 2018 which caused the gaps in Figure 22.

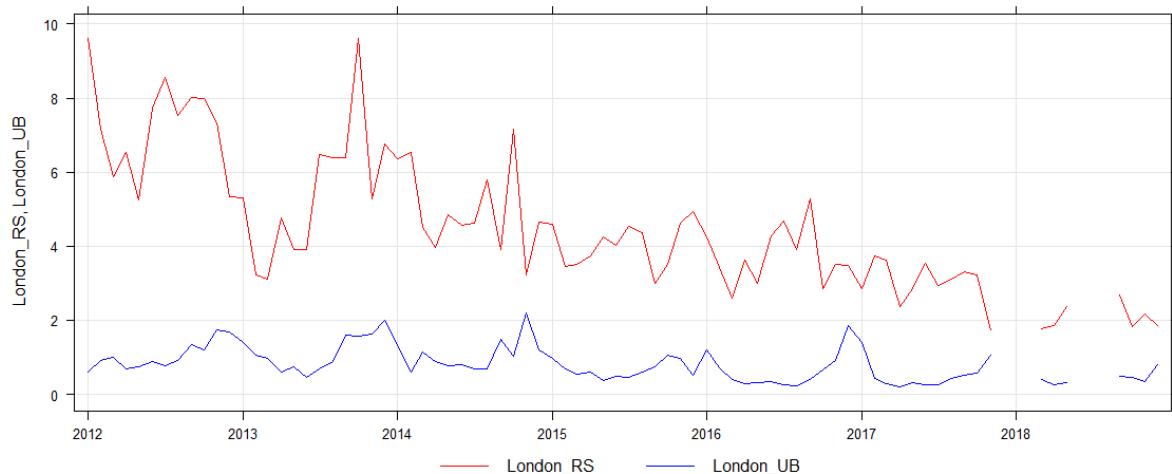
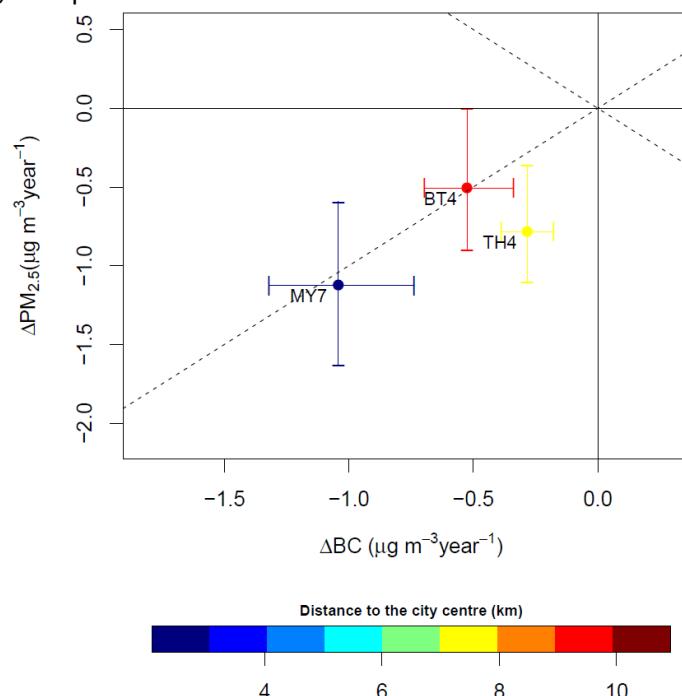


Figure 22 Urban and roadside increments in London for the period 2012 to 2018

The change of London roadside increment in Black Carbon was correlated with a reduction of the London roadside increment in $\text{PM}_{2.5}$ concentration⁴ from 2010-2014 as shown in Figure 23. The 1:1 correspondence indicates that the fall in $\text{PM}_{2.5}$ concentrations was entirely accounted for by the fall in Black Carbon, and that there are similar emission sources (road transport) for Black Carbon and $\text{PM}_{2.5}$ roadside increments, as expected.



Key: Blue: Marylebone Road
 Red and Yellow, other roadside sites in London operated outside of this Network

Figure 23 Relationship between trend in London roadside Black Carbon and $\text{PM}_{2.5}$ increments between 2010 and the end of 2014.

⁴ FONT, A. and FULLER, G. W. Did policies to abate atmospheric emissions from traffic have a positive effect in London? *Environmental Pollution*, 2016, **218**, 463-474.

5.2.2 UV component

Table 9 gives the annual average for each site for 2018.

Site	Mean concentration $\mu\text{g}\cdot\text{m}^{-3}$
Auchencorth Moss	0.0
Ballymena	0.4
Belfast Centre	0.3
Birmingham Ladywood	-
Birmingham Keeley Street	0.2
Cardiff Centre	0.2
Chilbolton	0.1
Detling	0.2
Dunmurry Kilmakee	0.3
Glasgow High Street	0.2
Glasgow Townhead	0.1
Marylebone Road	0.3
North Kensington	0.1
Strabane	0.8

Note: Birmingham Tyburn Ladywood site was not operational in 2018.

Table 9 Annual Mean UV component Concentrations for 2018

The annual mean concentrations are presented as a bar graph (Figure 24) to aid the comparison of sites:

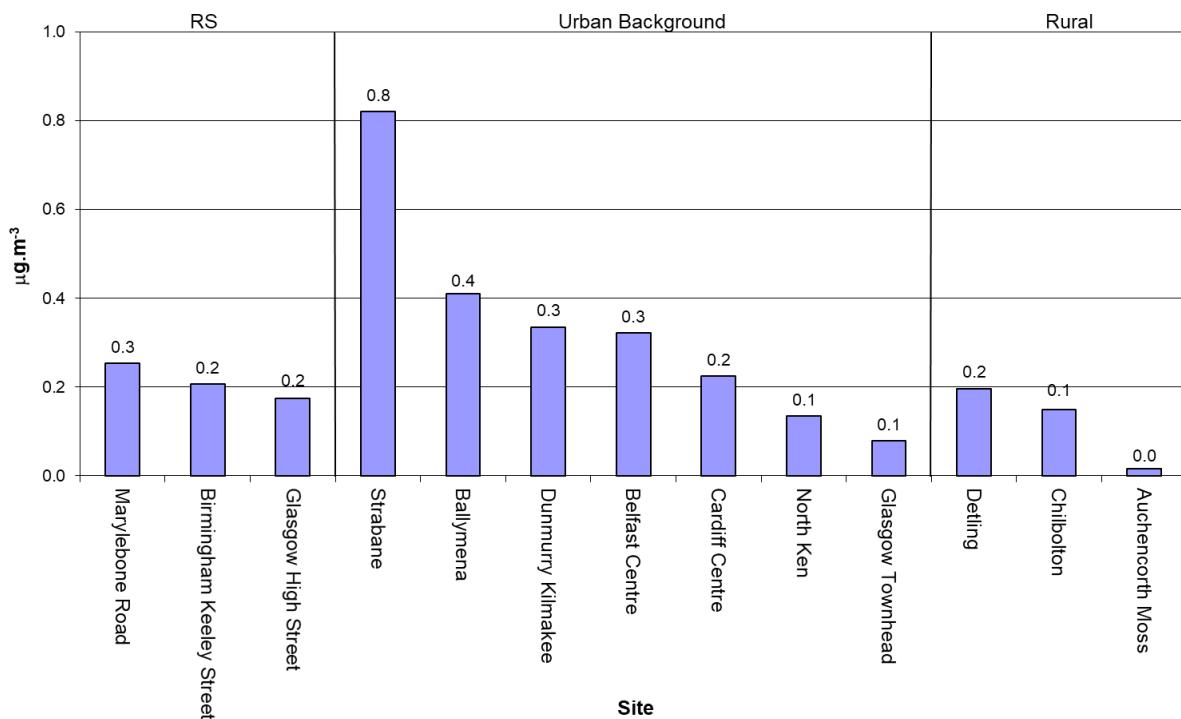


Figure 24 Annual Mean UV component Concentrations for 2018

UV component concentration increments between rural, urban background / centre and roadside sites for London and Glasgow have been analysed from periods when all the sites produced valid data, and are shown in Table 10.

Conurbation	Increment, $\mu\text{g.m}^{-3}$	
	Urban	Roadside
London	0.0	0.1
Glasgow	0.1	0.1

Table 10 Increments in UV component concentrations between rural, background and roadside sites in 2018

There was no significant difference in increments between 2017 and 2018.

It can be seen that the urban and roadside increments at all sites were small, indicating that domestic emissions in the two conurbations were small, and that road traffic was not a significant source for the UV component.

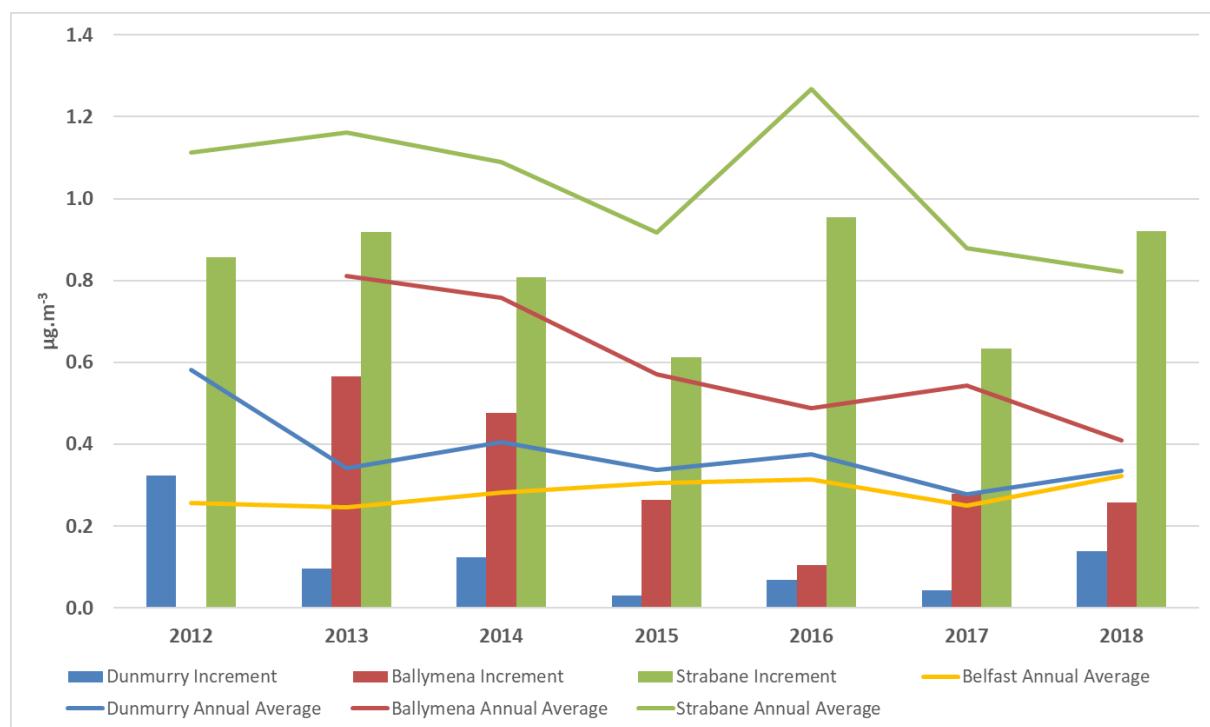
Using the same method, the urban increment in UV component concentration in Northern Ireland has been calculated relative to Belfast where gas heating has largely displaced oil and coal since 2000. The results are shown in Table 11.

Site	Increment compared to Belfast, $\mu\text{g.m}^{-3}$	Increment compared to Belfast, %
Dunmurry	0.1	43
Ballymena	0.3	80
Strabane	0.9	286

Table 11 Increment in UV component concentration in Northern Ireland

The increments at Dunmurry, Ballymena and Strabane are in line with a history of solid fuel usage for secondary heating in Dunmurry, and a significant usage of non-smokeless fuel in Strabane.

Changes in the UV component increment in Northern Ireland over the last 7 years are summarised in Figure 25.

**Figure 25 Annual Mean UV component concentrations and increments compared to Belfast for 2012-2018**

5.2.3 Data Capture

Table 12 gives the data capture for each site for 2018, given as a percentage of the time for which the site was expected to be operational. The time coverage, that is the percentage of the complete calendar year for which data from each site is available, has also been given.

Site	Data Capture %	Time Coverage %
Auchencorth Moss	99	99
Ballymena	97	97
Belfast Centre	63	63
Birmingham Ladywood	-	-
Birmingham Keeley Street	83	83
Cardiff Centre	98	98
Chilbolton	82	82
Detling	70	70
Dunmurry Kilmakee	99	99
Glasgow High Street	90	90
Glasgow Townhead	87	87
Marylebone Road	89	89
North Ken	96	96
Strabane	86	86

Table 12 Data capture rates of the Aethalometers for 2018

The average data capture for the Network was 87.6%. The low data capture seen at Belfast was due to the leak in the sampling line resulting in the measurement of cabin air instead of ambient air. At the Detling site a significant leak was found during the NPL audit visit and the invalid data has been removed from the final results. It was also noticed that in 2018 there were more technical issues with the instruments, resulting in lower data capture than previous years (~95%). This might be attributed to the equipment being old and in continuous use during the last 10 years.

5.3 TEMPORAL VARIATIONS

The following section presents analysis of the 2018 Black Carbon and UV component concentrations with respect to the hour of the day. Charts of variations over the day of the week and the month in the year were made using the data from 2009 – 2018, to avoid bias introduced by a single year of measurements. Y-axes vary by site.

All results have been grouped by site classification. The site order for the Roadside sites is by decreasing Black Carbon concentration, while the site order for the Urban Background and Rural Background sites is by decreasing UV component concentration. The units on the y-axis are $\mu\text{g.m}^{-3}$ for Black Carbon and equivalent $\mu\text{g.m}^{-3}$ for the UV component.

The data has been plotted in local time (GMT/BST), as the pollution sources are primarily attributed to human activity and not solar-driven atmospheric chemistry sources.

The 2018 data are presented in Figures 26 to 28.

The 2009-2018 data are presented in Figures 29 to 30. These long term plots only include those sites which have been operating for the whole of this time period. The site at Chilbolton was seen to show significantly different concentrations from that at Harwell, so the latter site has been removed from the long term time series plots.

Chart Key

For all of the charts, the continuous central line is the mean value and the shaded area about this line represents the uncertainty in the mean y-value due to the spread of the results over that averaging period, expressed with a level of confidence of 95%. It is not the overall measurement uncertainty. The shaded area on the x-axis in Figures 26-28 is for display purposes only, to allow the uncertainty in the mean value to be seen more clearly.

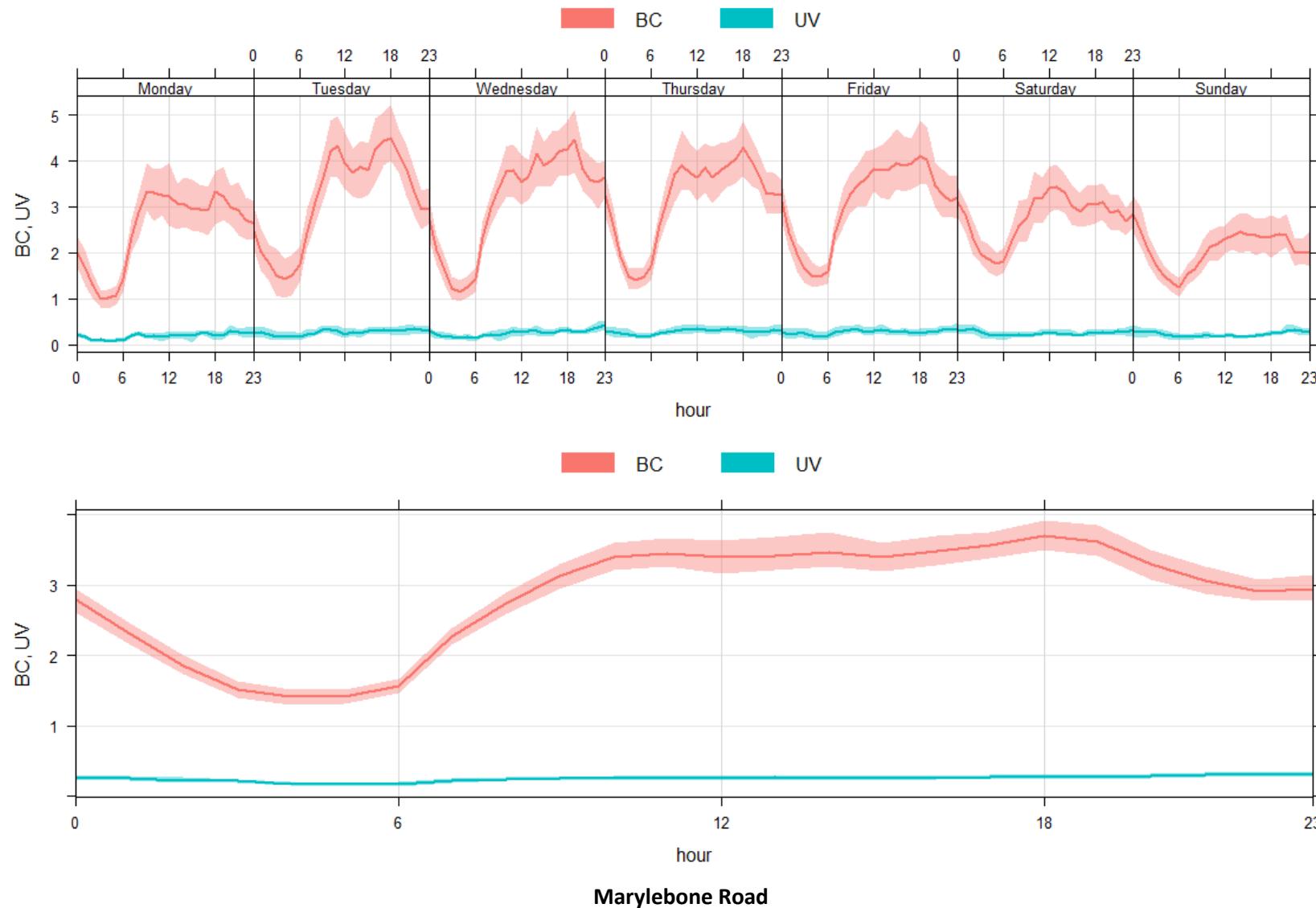
Acknowledgement

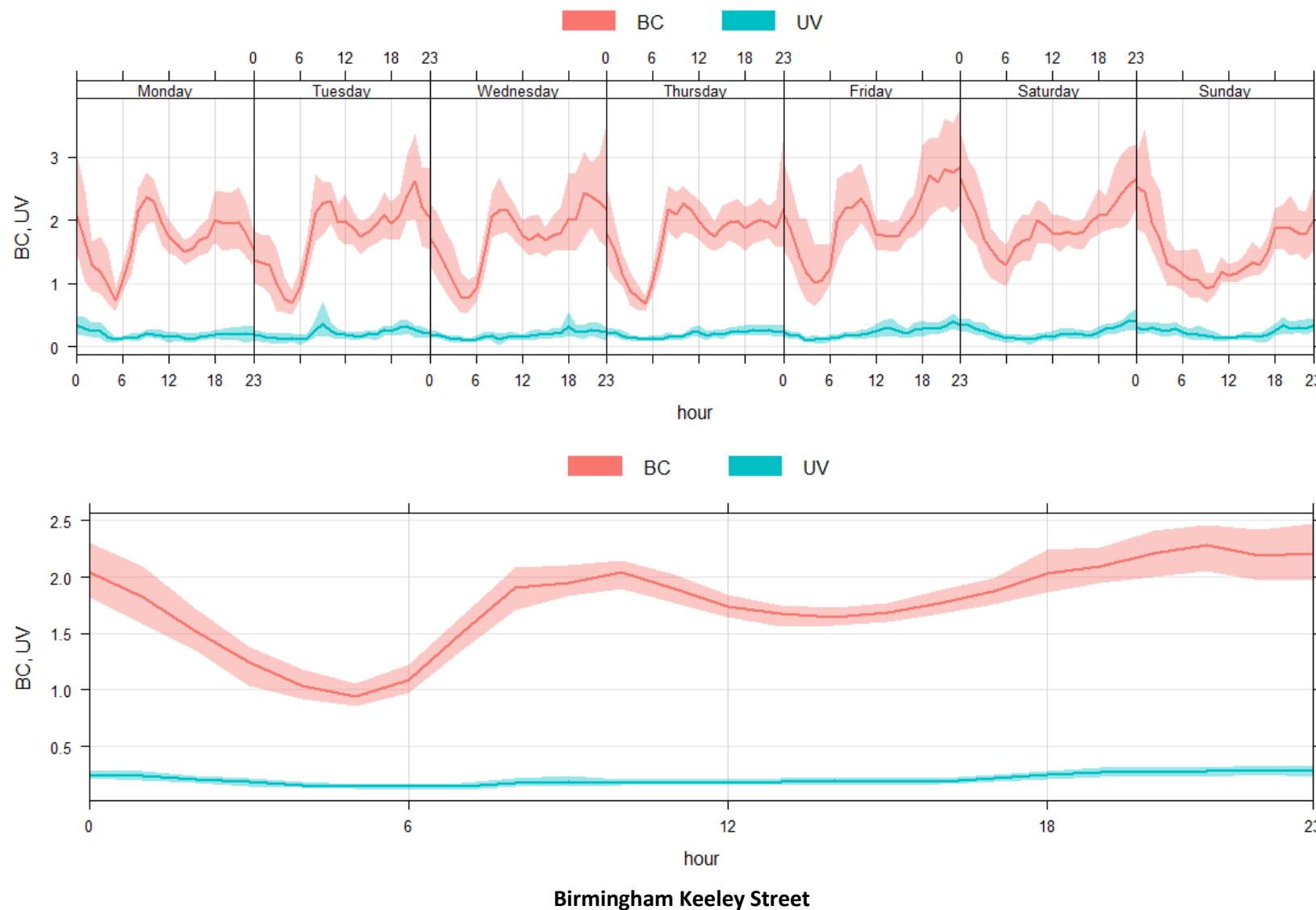
Figures 26 to 30 are generated using the Open-Air Tools run on the R software platform^{5,6}.

5 CARSLAW, D. C. and ROPKINS, K. OpenAir --- an R package for air quality data analysis. *Environmental Modelling & Software*, 2012, **27-28**, 52-61.

6 CARSLAW, D. C. and ROPKINS, K. OpenAir: Open-source tools for the analysis of air pollution data, R package version 3.6.1, 2019.

2018 Data





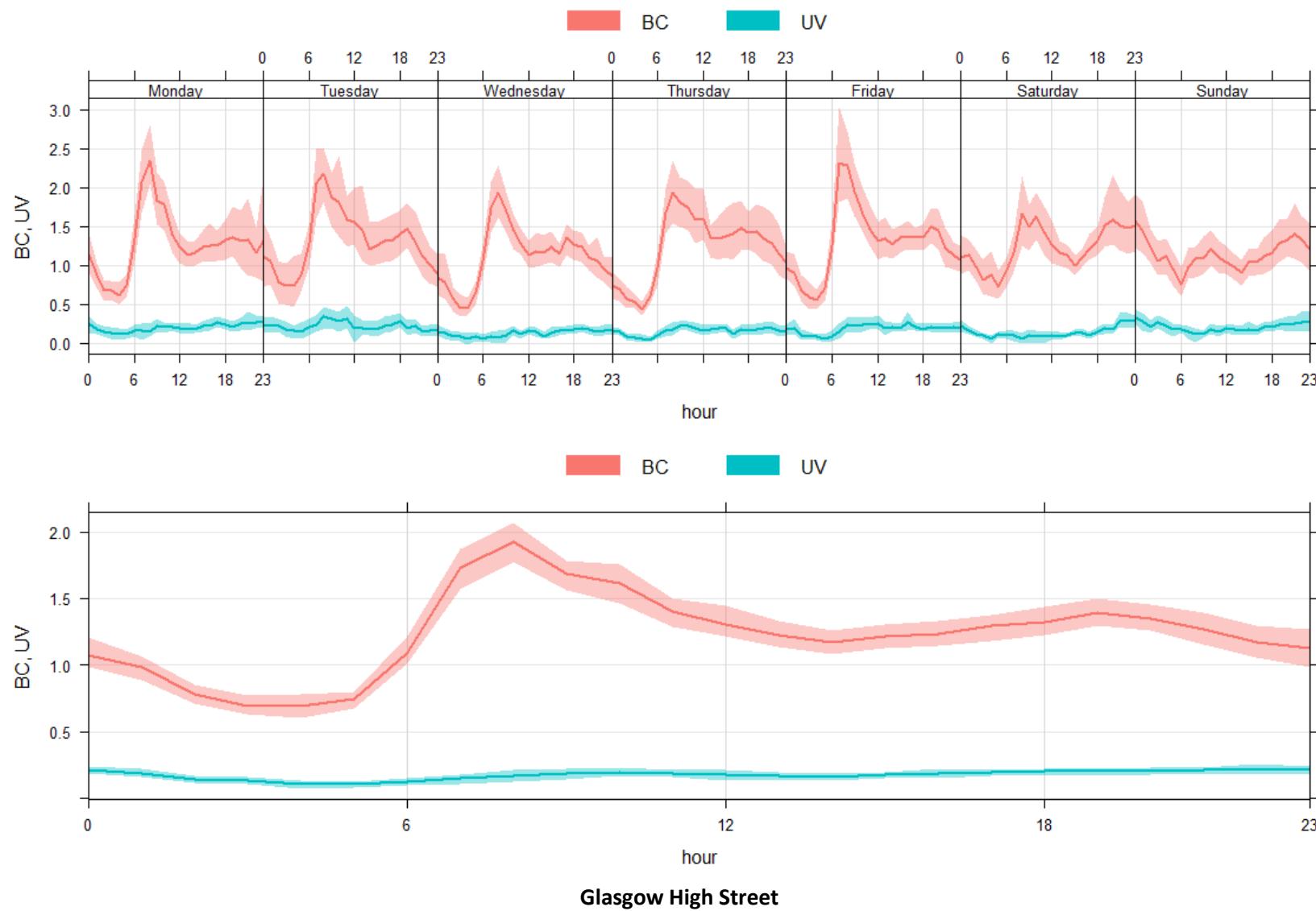
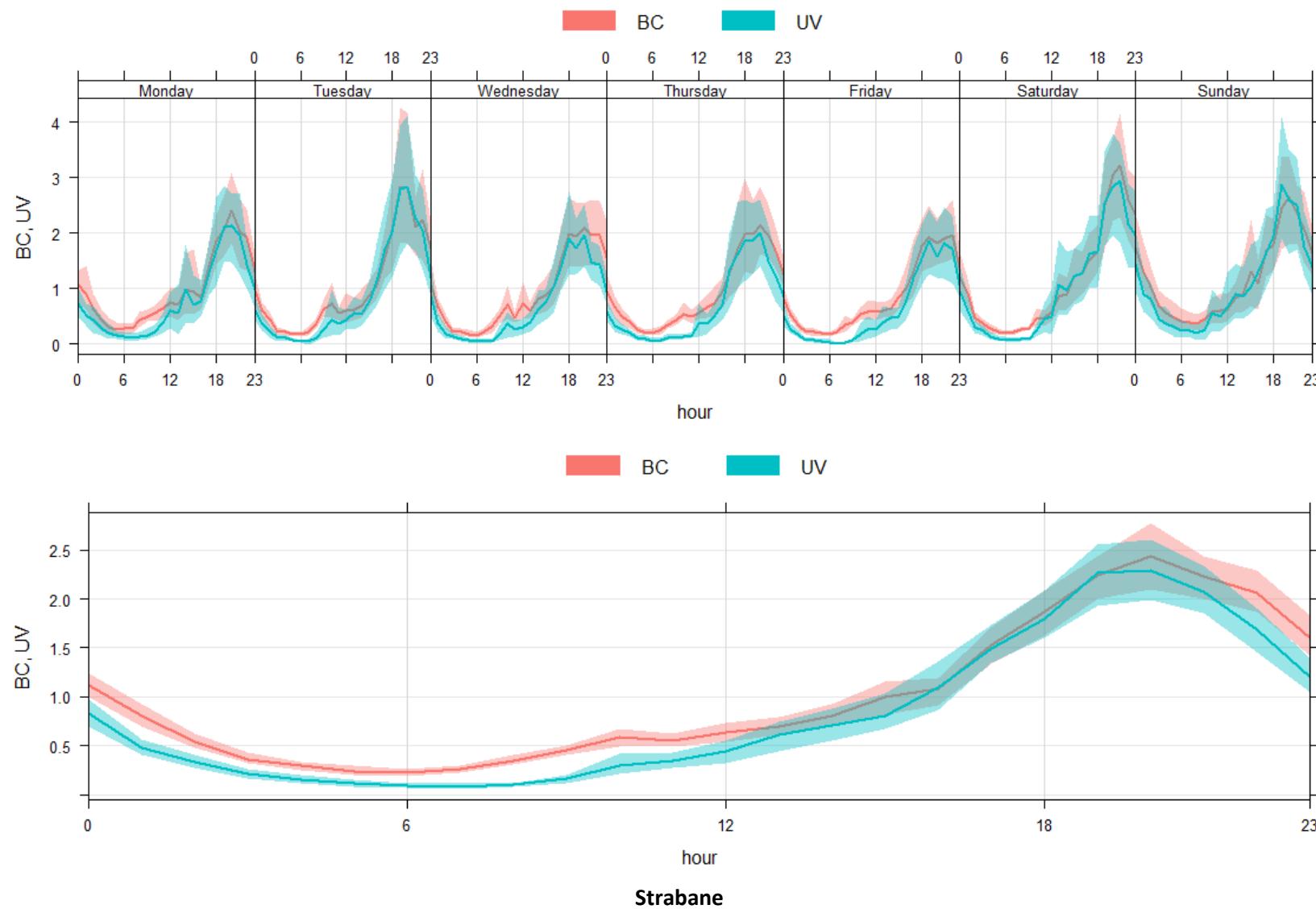
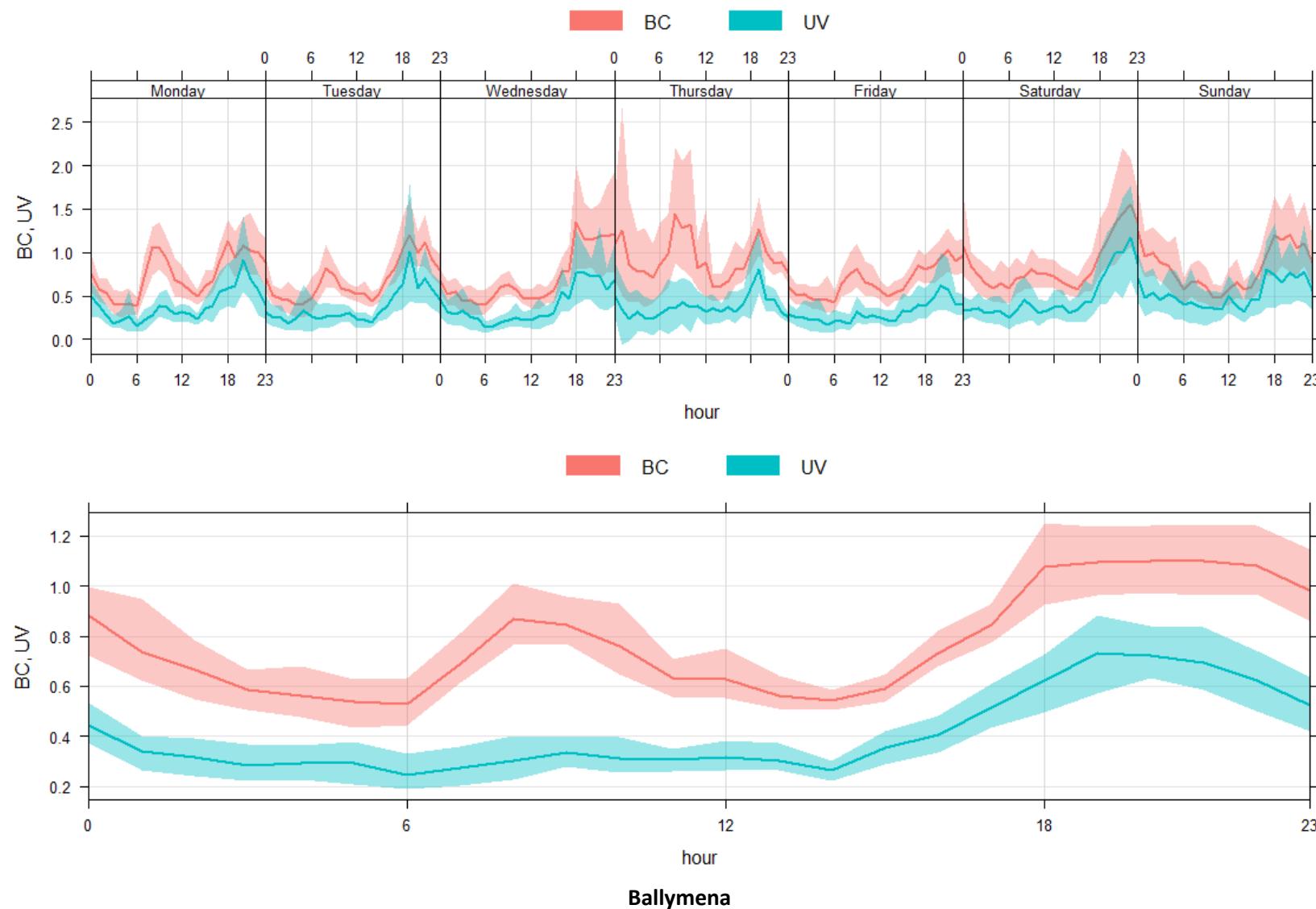
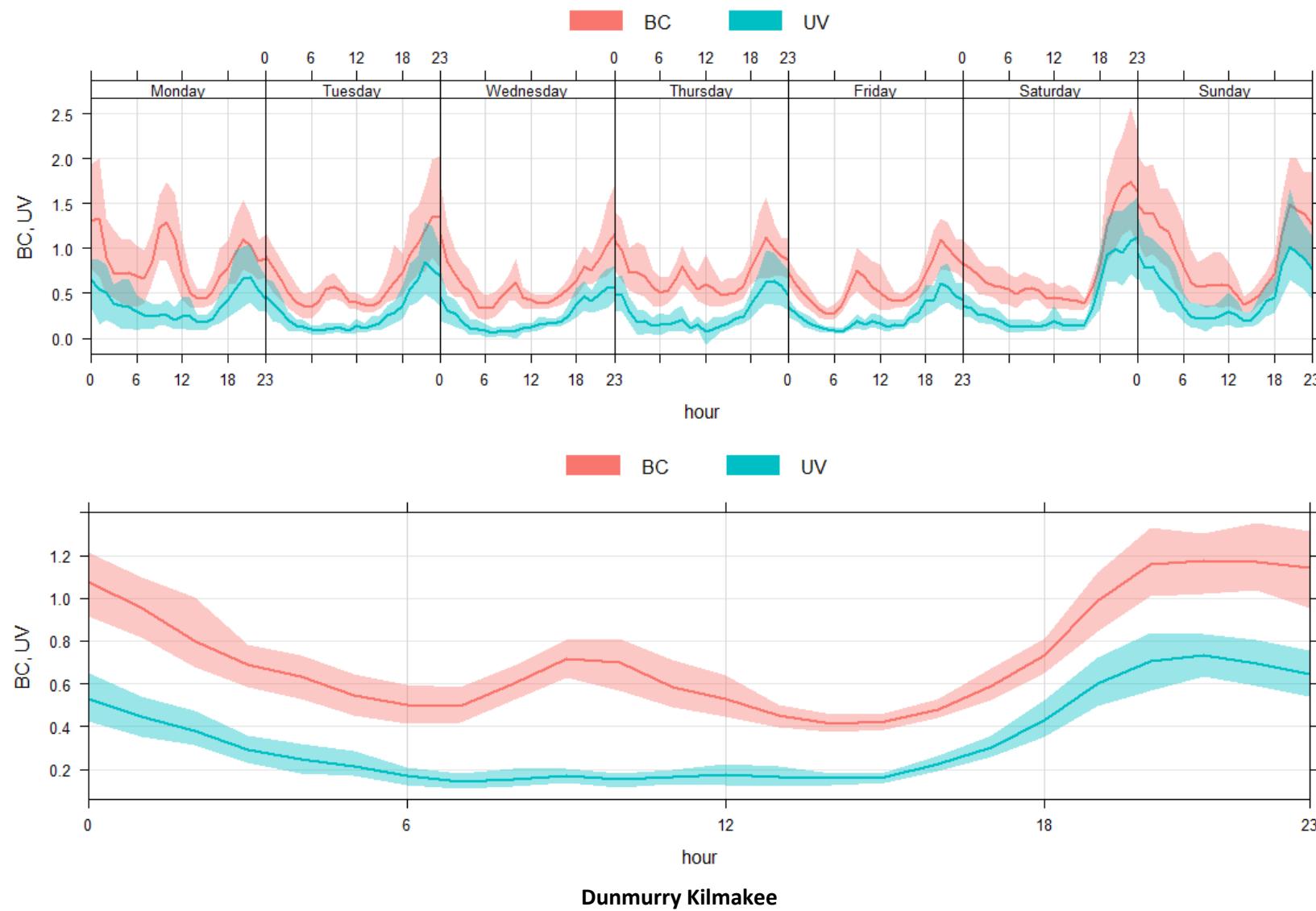
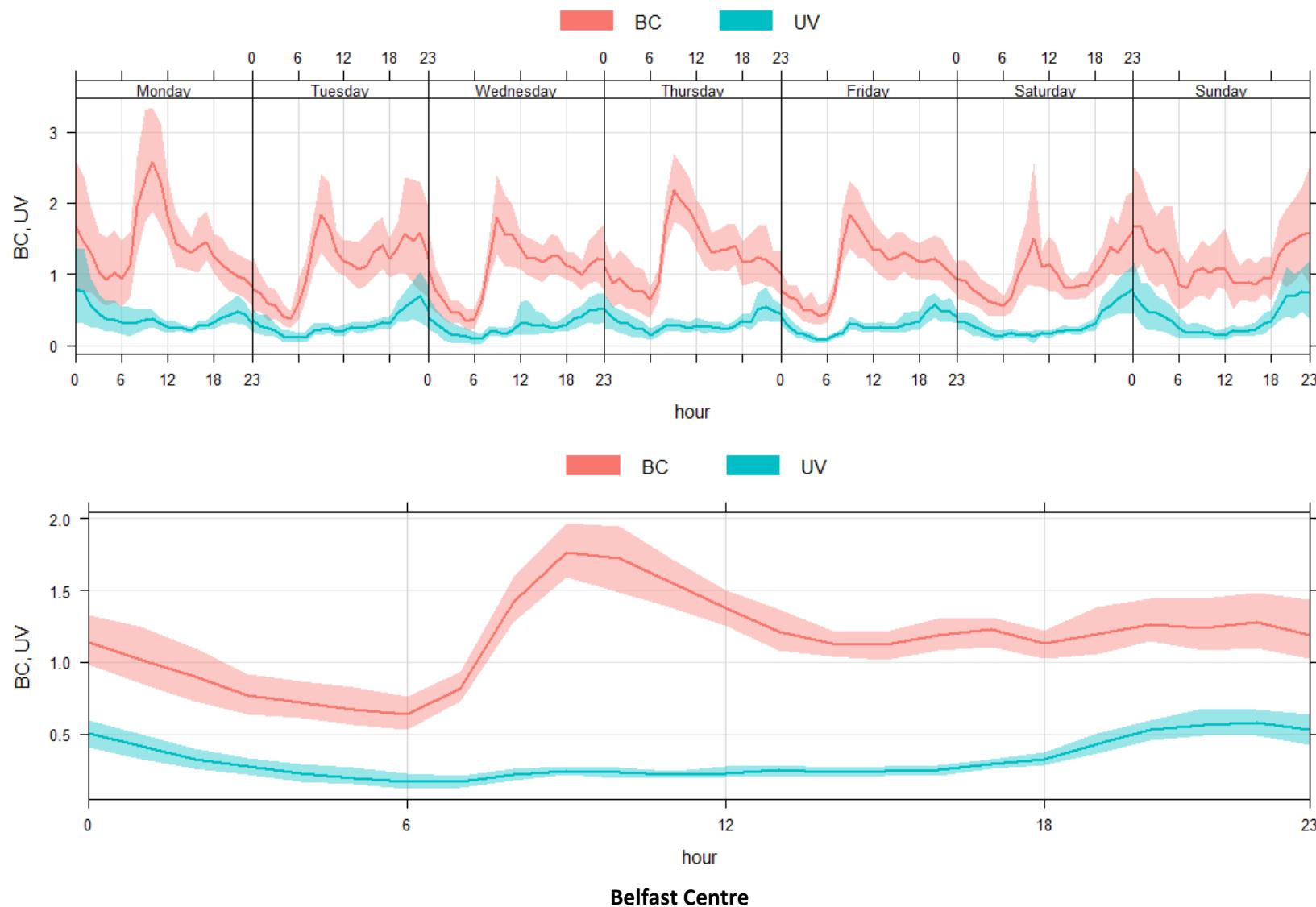


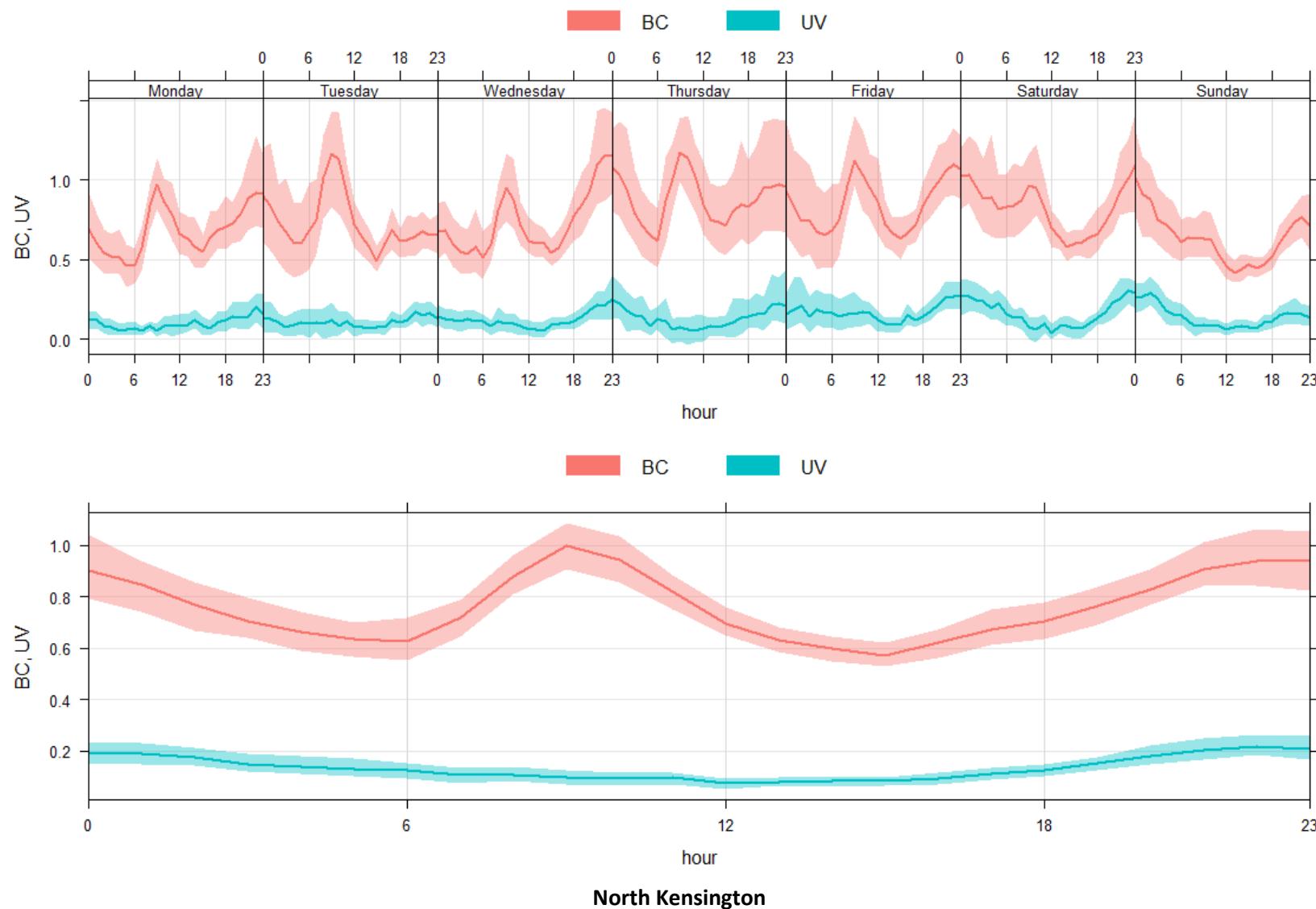
Figure 26 Roadside Sites

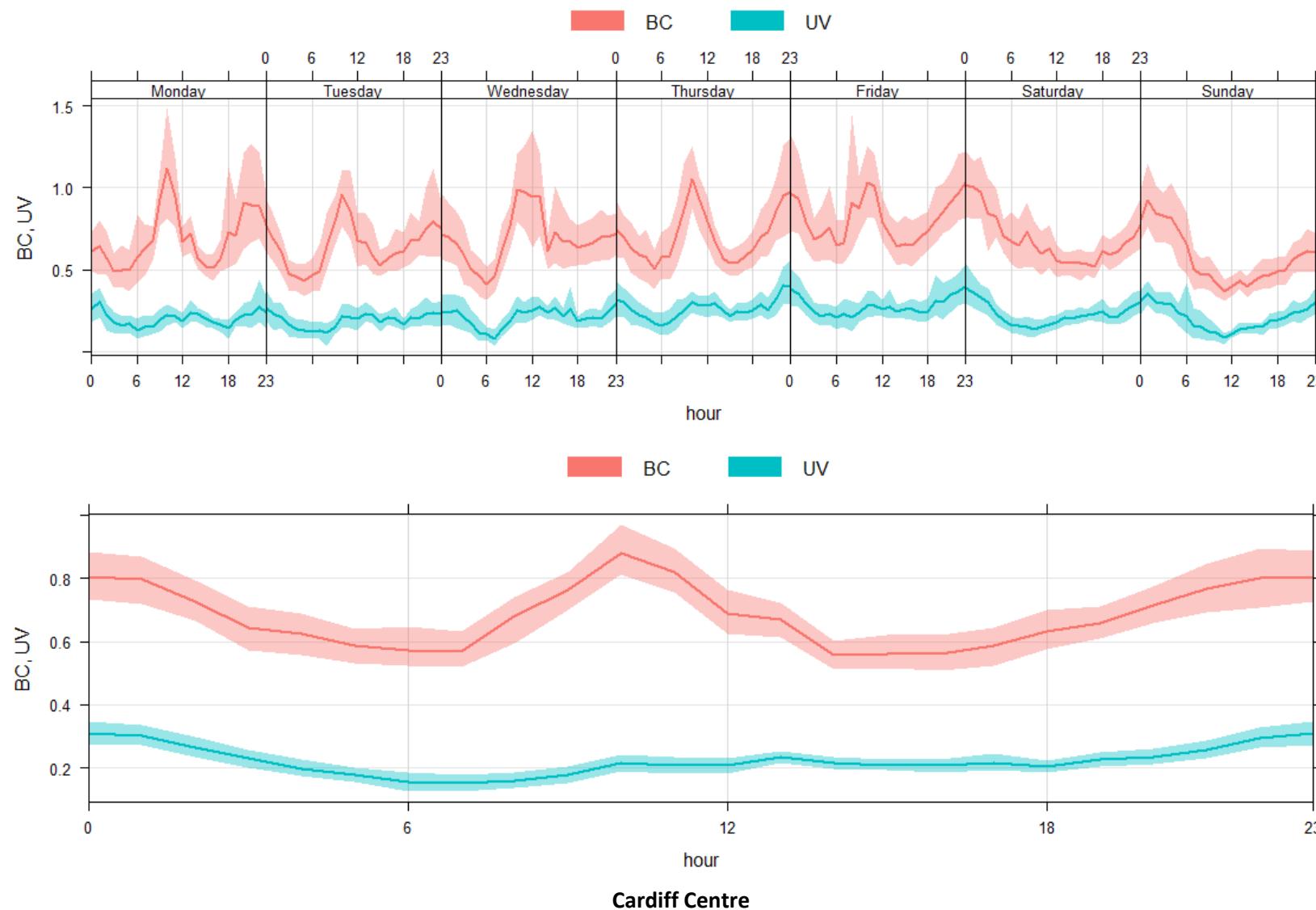












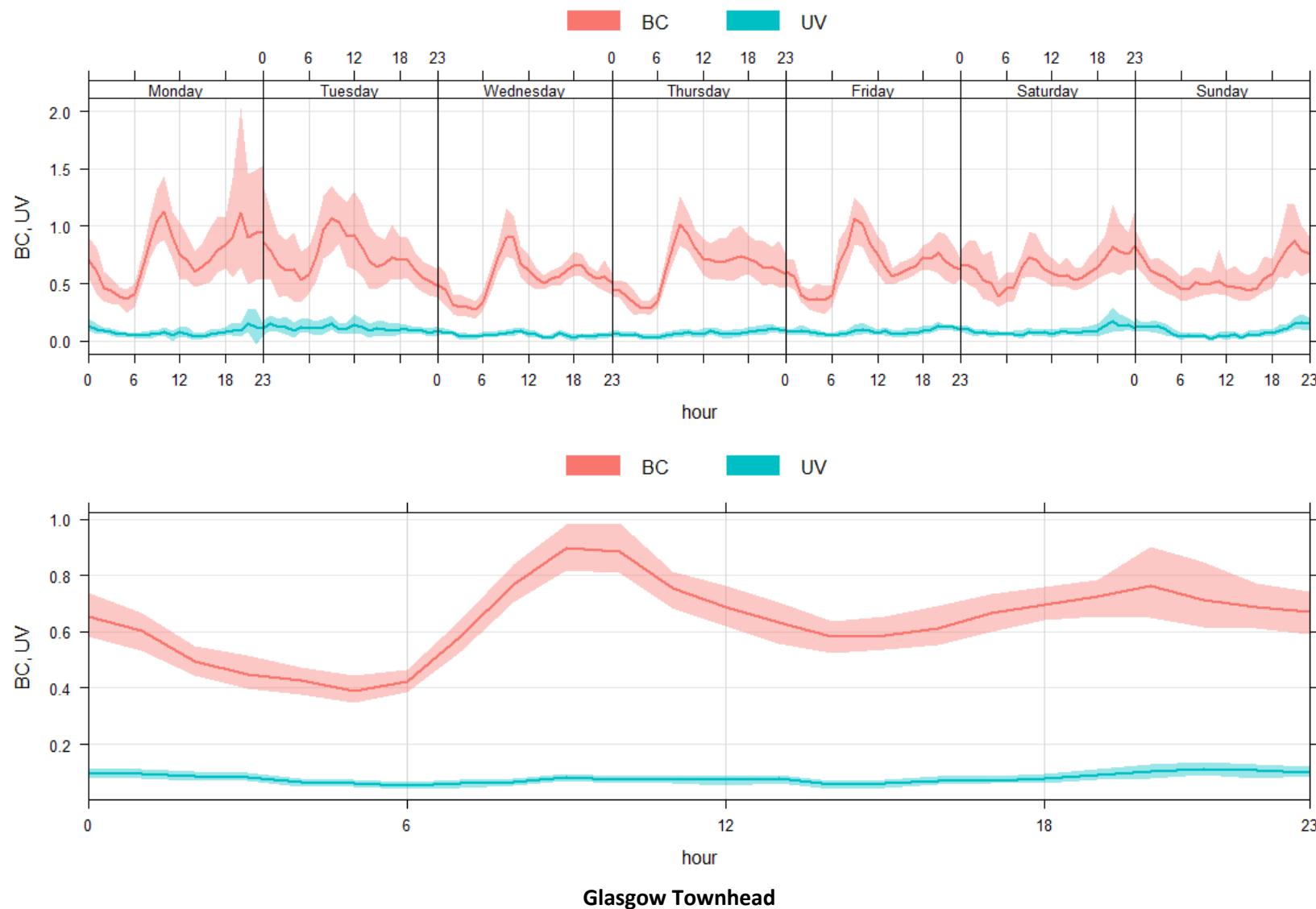
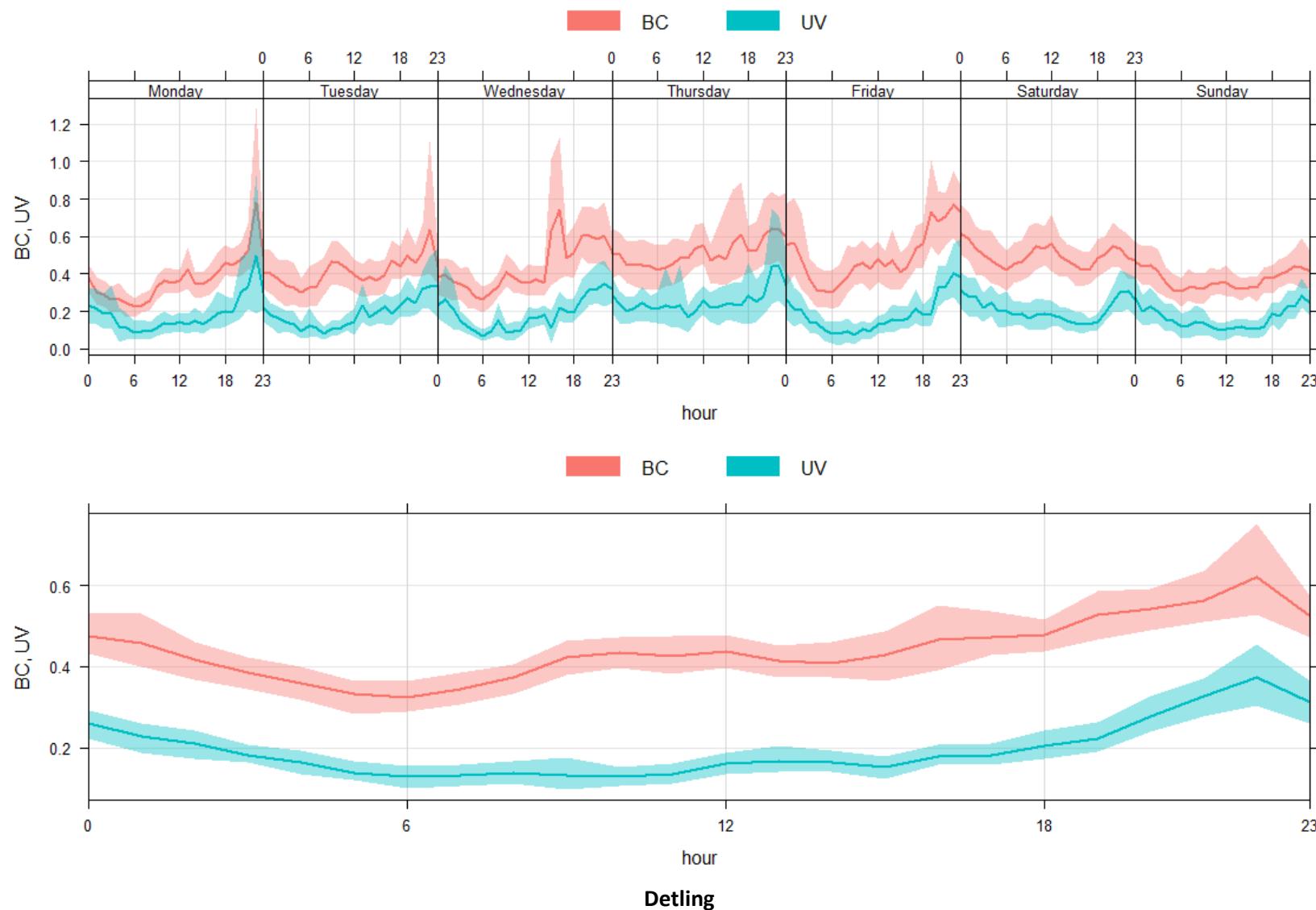
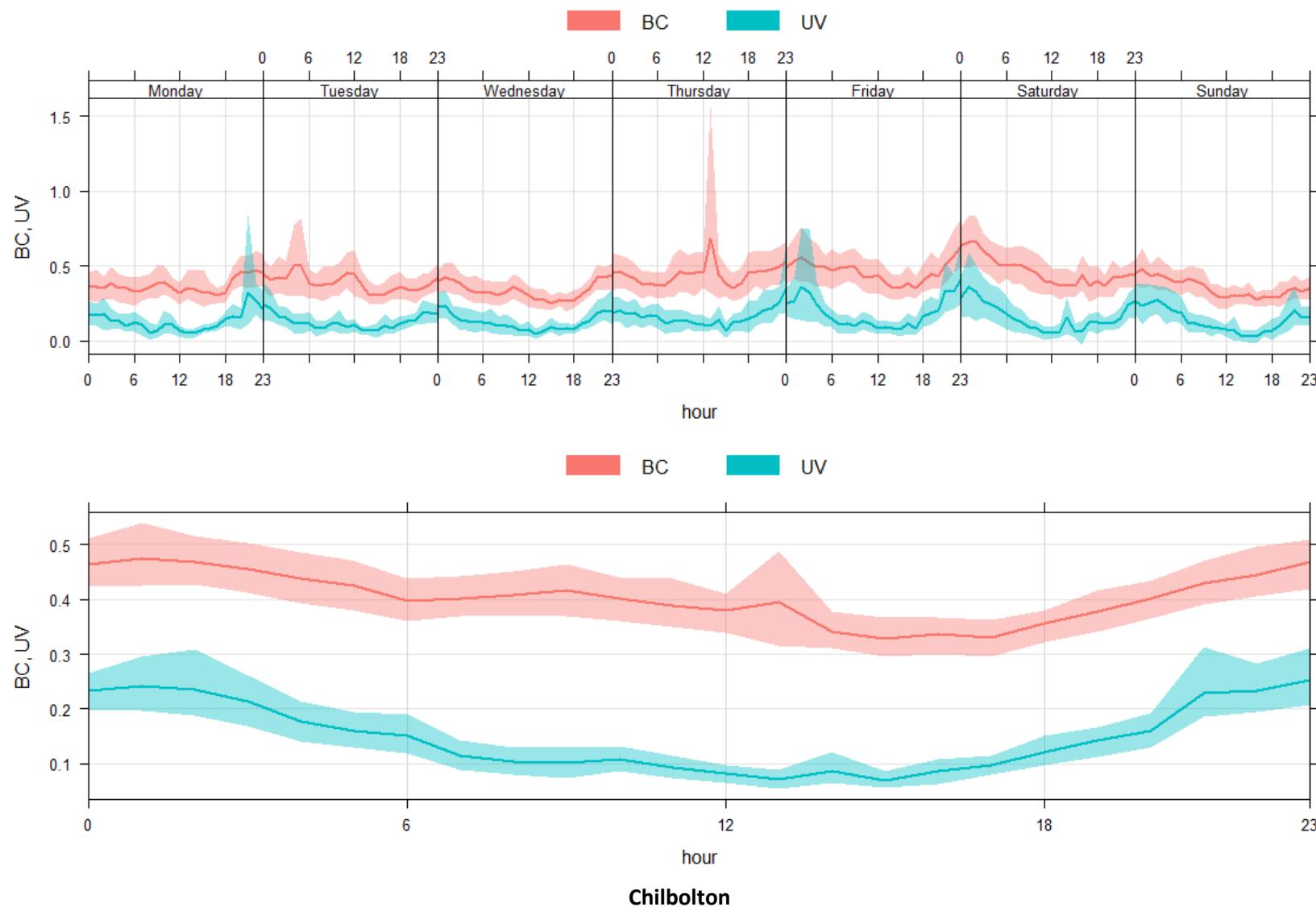


Figure 27 Urban Background Sites





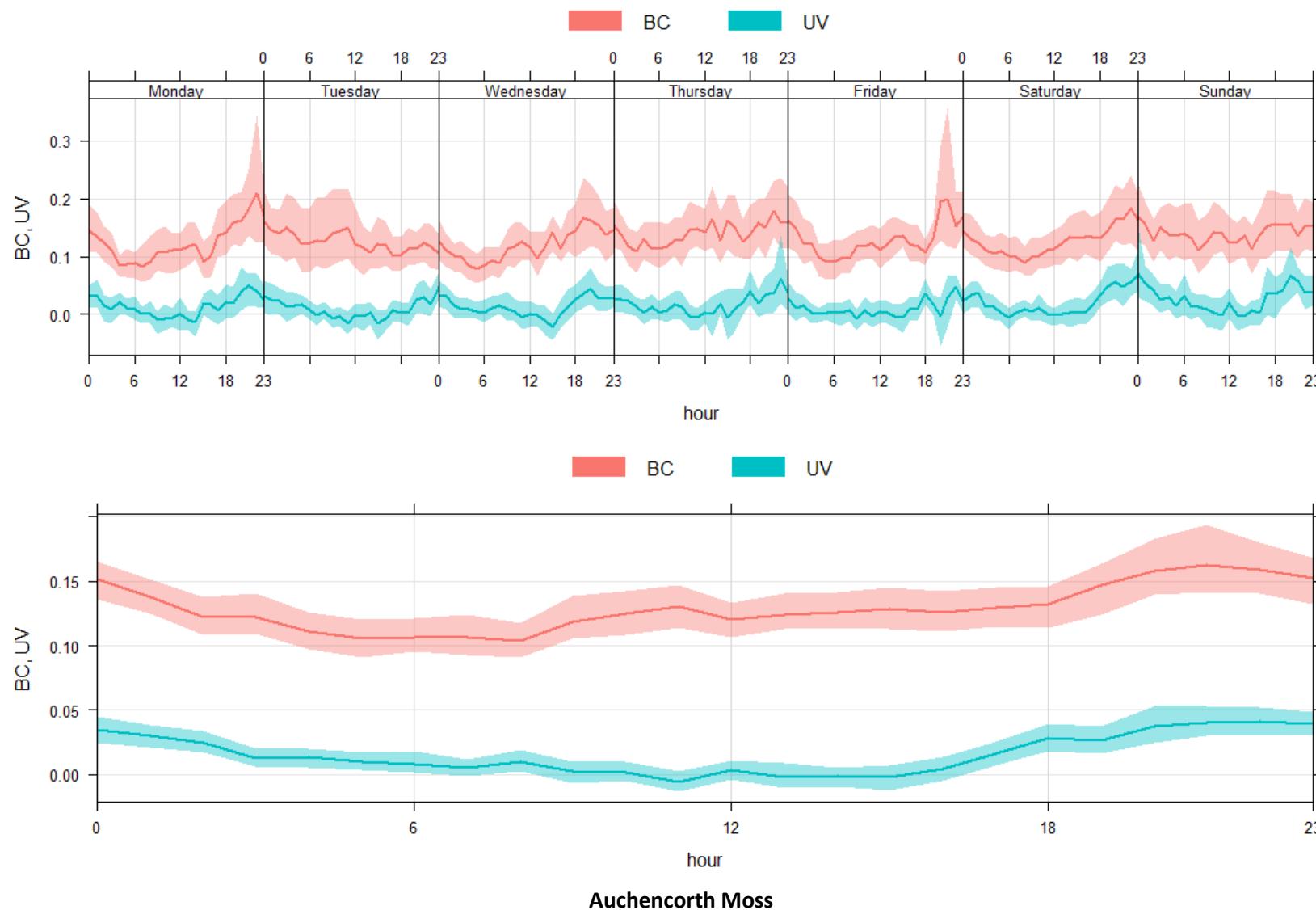


Figure 28 Rural Sites

2009 – 2018 Data

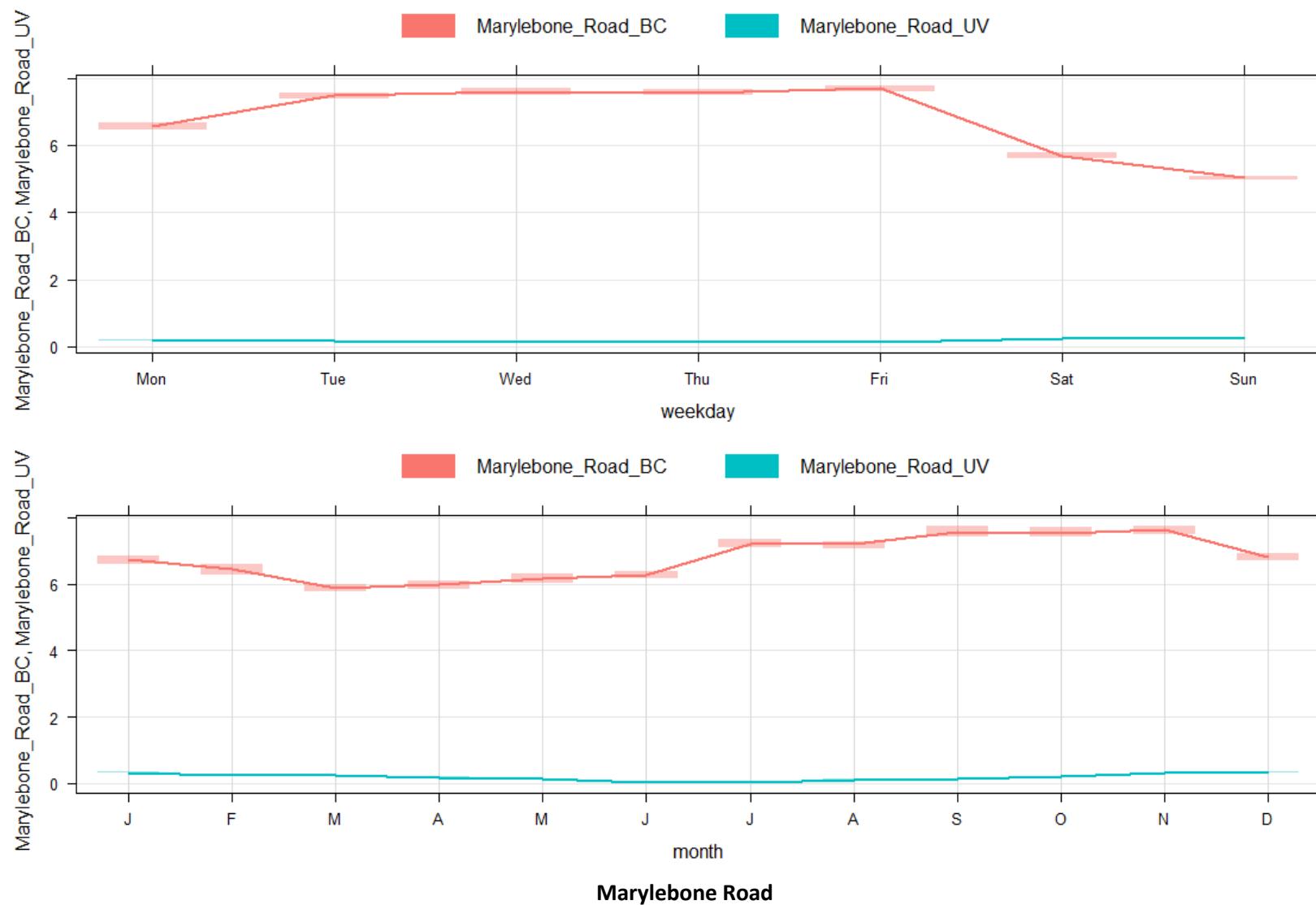
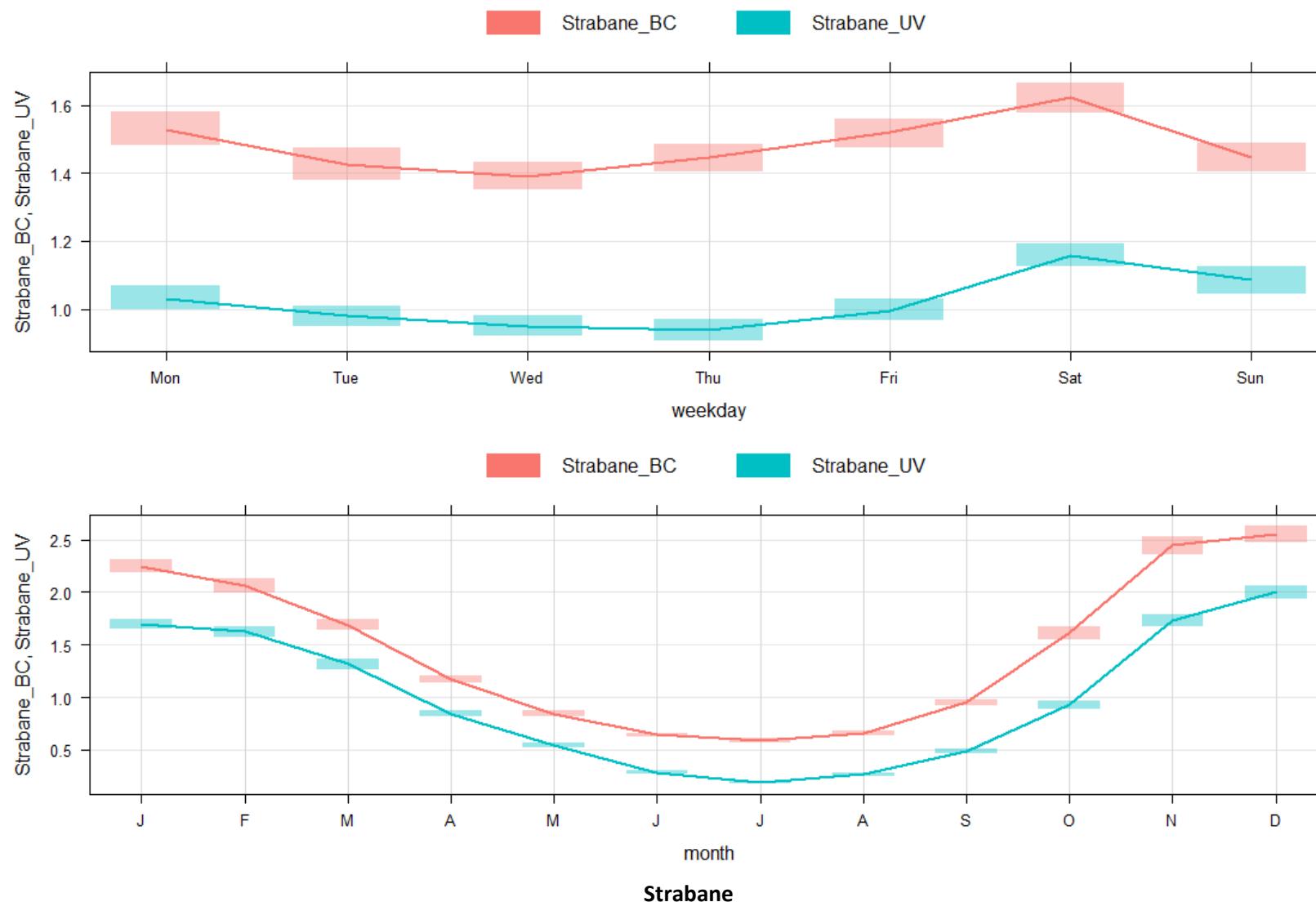
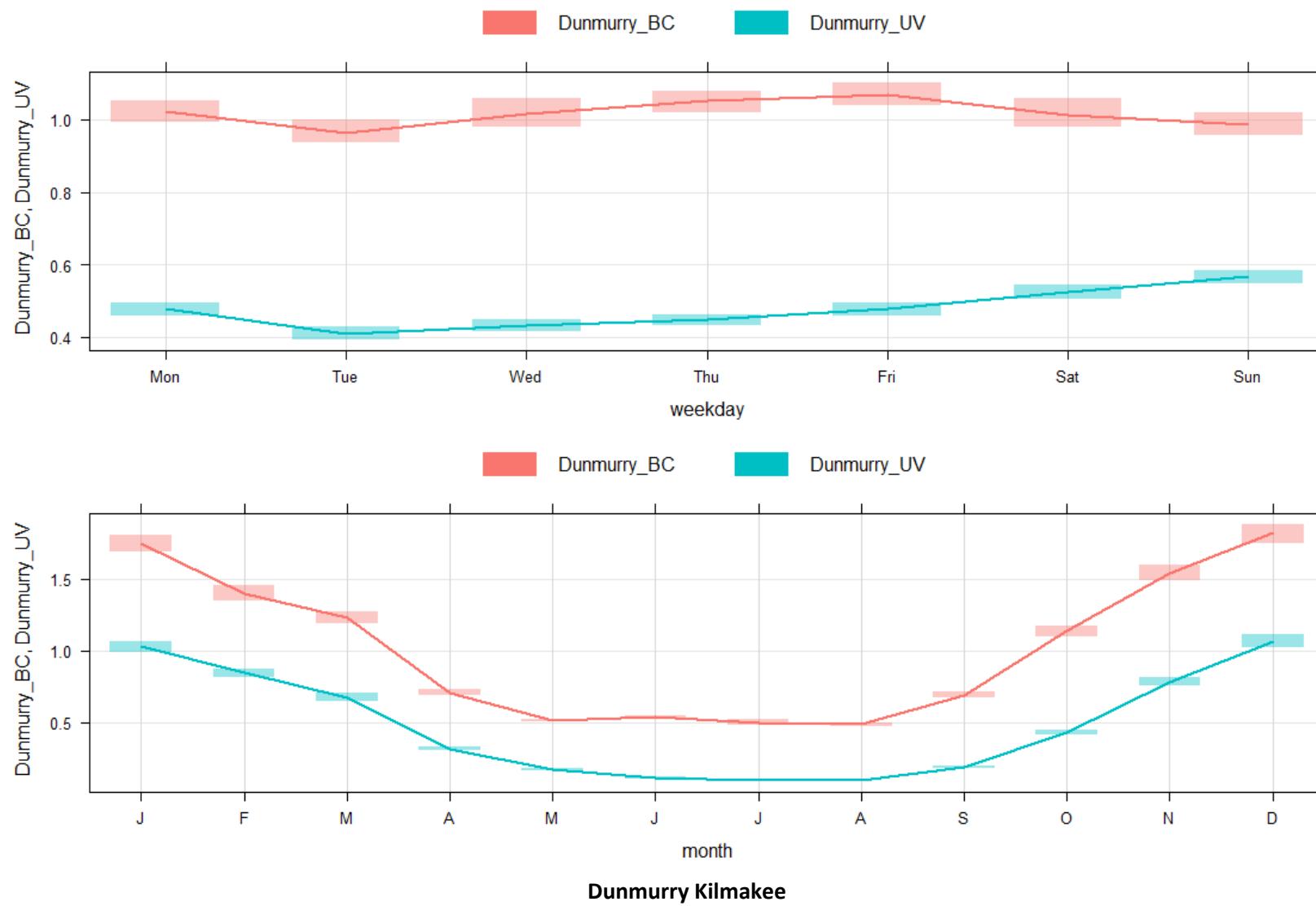
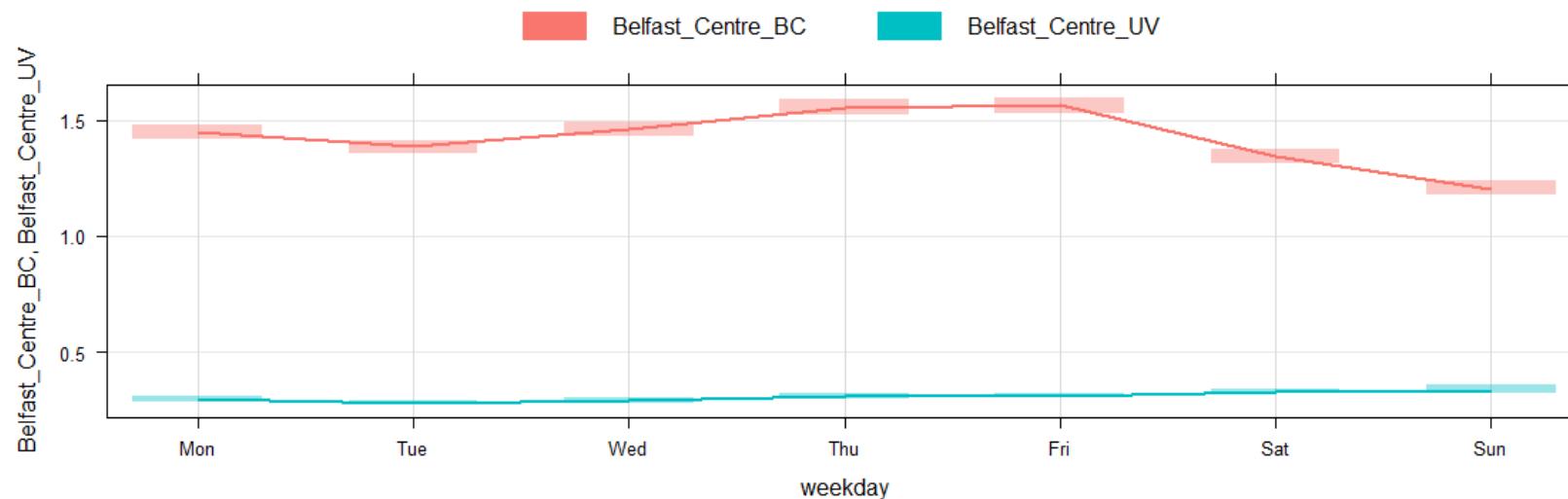
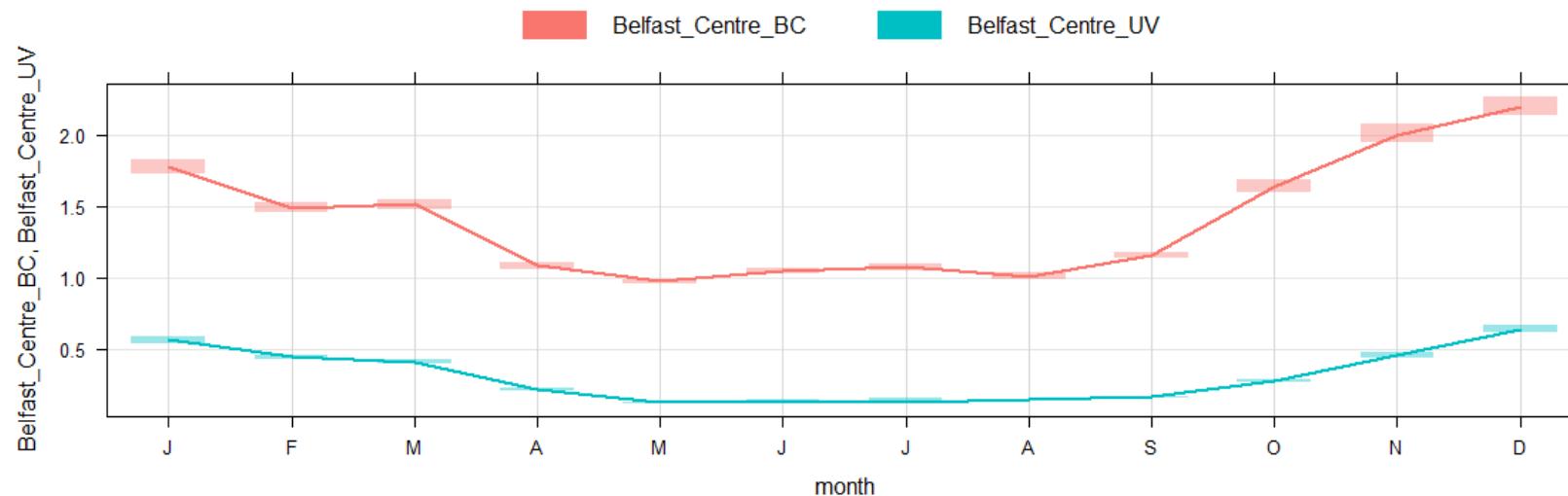


Figure 29 **Roadside Sites**







Belfast Centre

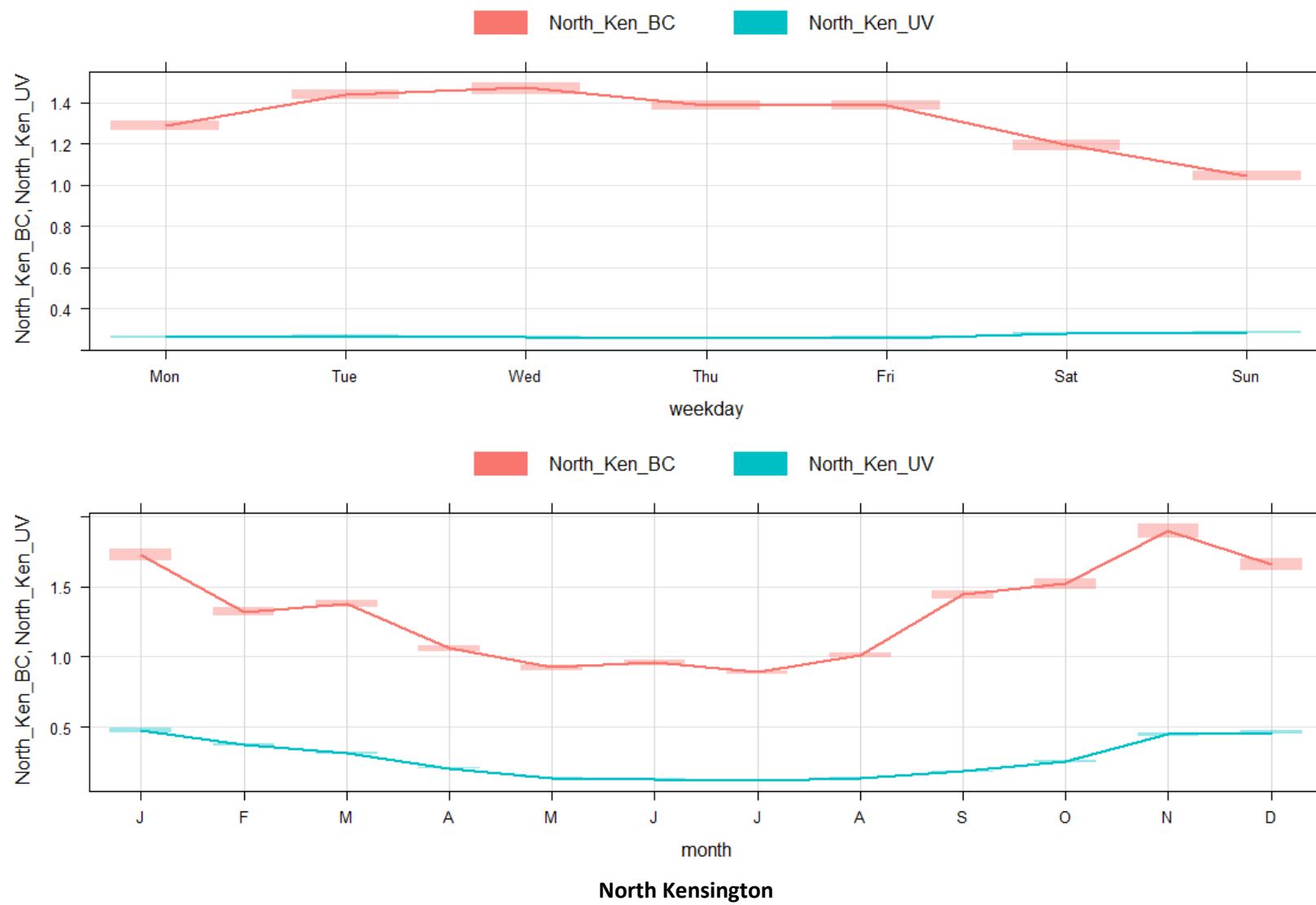


Figure 30

Urban Background Sites

Roadside sites

On weekdays the Black Carbon concentrations at the roadside sites followed the expected profile for traffic movements through the day, with raised concentrations in the morning and evening rush hours. This double peak can be seen at all the roadside sites but is particularly noticeable at Birmingham Keeley Street, where a dip in concentration during the day is followed by a significant evening rush hour peak. Glasgow High Street showed a smaller evening peak, and at Marylebone Road it is less prominent because the concentrations remained at a higher level throughout the middle part of the day.

In general the weekend days showed slightly lower and more constant concentrations, particularly at Marylebone Road. This corresponds to reduced traffic and a change in vehicle fleet over the weekend when there is a considerable drop in heavy goods vehicles passing this site. This weekend reduction in Black Carbon concentrations was less obvious than in previous years, however, suggesting that the falling annual average at this site is driven more by weekday concentrations. Weekends at Birmingham Keeley Street and Glasgow High Street still show some variability throughout the day but instead of the clear morning rush hour peak they rise to a high in the evening. This could be due to recreational journeys and return travel at the end of the weekend.

Seasonal variations at roadside sites are relatively small, as expected when traffic is the dominant source.

There was little UV component signature in any of the roadside sites.

Urban Background sites

Urban background sites can be split into two categories: those away from main roads with mixed influences from both road traffic and domestic sources; and those predominantly influenced by emissions from domestic heating.

Concentrations measured at Belfast Centre, Cardiff Centre, North Kensington and Glasgow Townhead showed a signature from traffic, seen as a peak in the morning rush hour with little corresponding increase in UV component concentrations. Peaks related to the evening rush hour were also seen, but these often also showed an increase in UV component concentrations. This, along with the fact there was less of a drop in concentrations later in the evening than seen at roadside sites, indicates a domestic emission source which is likely from secondary heating. This effect was particularly visible at Cardiff and North Kensington.

Black Carbon concentrations were generally lower at weekends than on weekdays at these sites and showed less influence from rush hour traffic. The morning peak seen on weekdays is either absent or reduced, and concentrations peak in the evening and into the early hours. This is visible in both the Black Carbon and UV Component, suggesting contributions from both evening leisure journeys and secondary heating.

Both Black Carbon and UV component concentrations showed some seasonal dependence, with a decrease in concentration over the summer months and an increase in concentration in the winter months. If it is assumed that the emissions from road transport were relatively consistent through the year then normalising the concentrations should reveal any monthly variability in either the Black Carbon or UV component emissions. In this process each concentration is divided by the annual mean of that component, allowing patterns to be compared even when results are on very different scales. This can be seen in Figure 31, using Belfast Centre as an example.

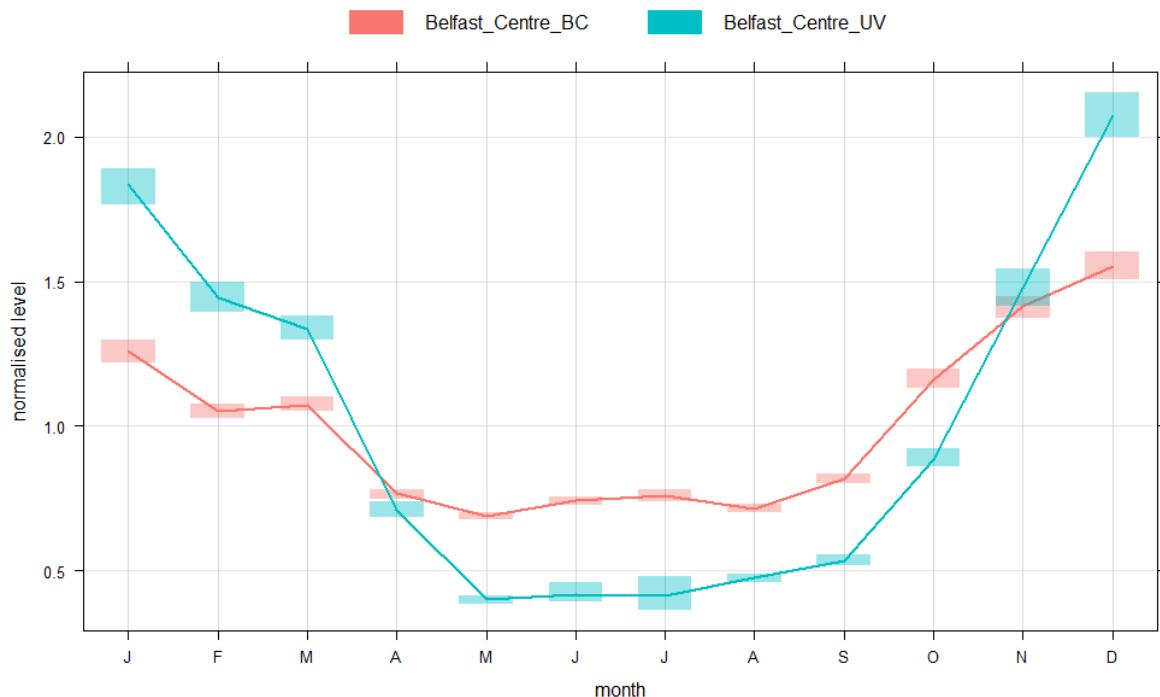


Figure 31 Normalised monthly variability at Belfast for the period 2009 - 2018

UV component and Black Carbon concentrations at the Northern Irish sites of Strabane, Ballymena and Dunmurry Kilmakee followed similar hourly, daily and seasonal trends. Concentrations at these sites were dominated by emissions from domestic heating. The highest levels were seen at Strabane, which is not on the natural gas supply and where domestic heating mainly comes from oil. Strabane is in a smokeless zone, however it appears that solid fuel burning may be occurring in residential areas. Due to the large emission factors of PAHs from smoky coal⁷ compared to oil and gas, it does not take many houses burning this coal to have a big influence on ambient concentrations. There is little evidence of traffic emissions during the rush hour periods.

Ballymena and Dunmurry are on the natural gas supply and this is the predominant source of domestic heating, however coal is often used as secondary heating in the evenings. Due to the difference in emission factor discussed above this can have a significant effect on ambient concentrations. At these sites the morning rush hour is visible in the Black Carbon concentrations, but in the evening the traffic signal is masked by domestic emissions which were also seen in the UV component. The highest concentrations of both Black Carbon and UV component were seen on the weekend evenings which indicates secondary heating, as opposed to Strabane where there was little variability between days of the week. Figure 32 gives the normalised monthly variability and Figure 33 gives the hourly variability.

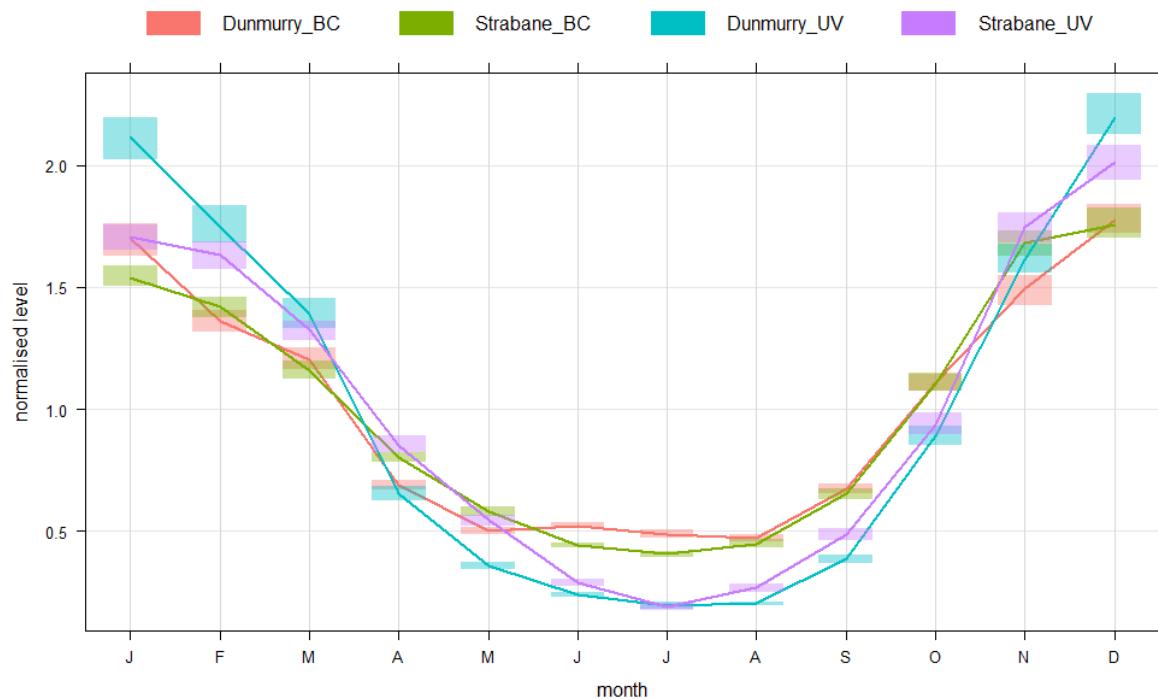
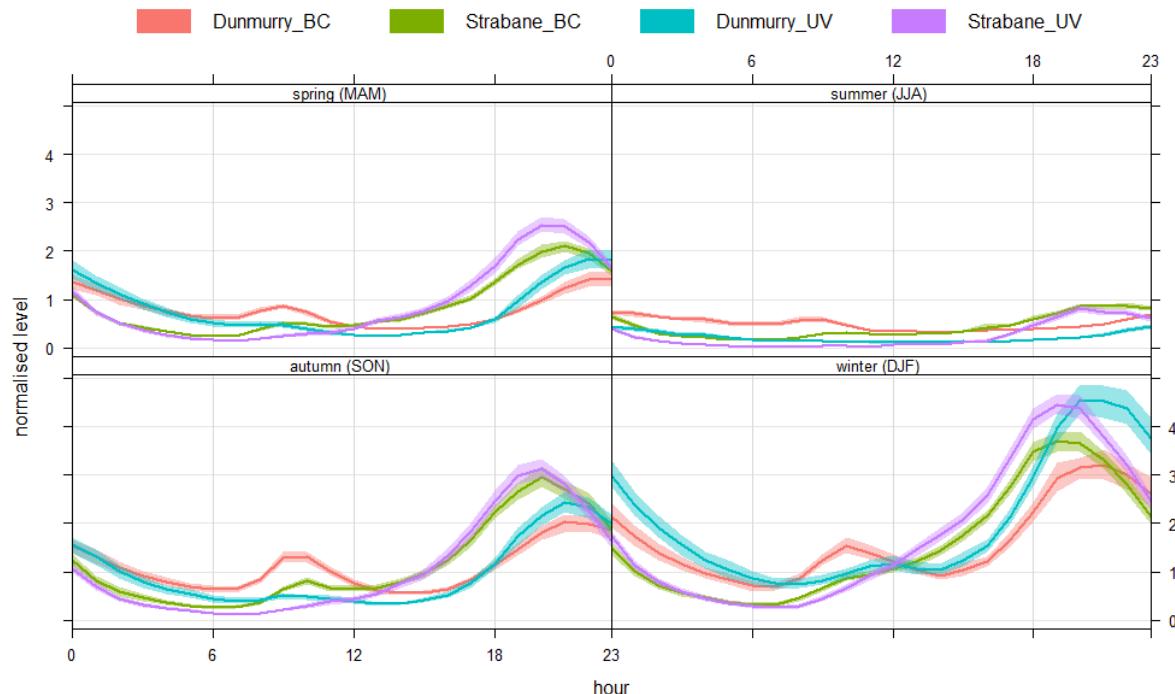


Figure 32 Normalised monthly variability at Strabane and Dunmurry for the period 2009 - 2018



Note: In the above charts the shaded area on the y-axis represents the uncertainty in the mean y-value due to the spread of results, expressed with a level of confidence of 95%.

Figure 33 Seasonal Black Carbon and UV component concentrations measured at Strabane and Dunmurry for the period 2009 – 2018

It can be seen that the evening concentrations of both Black Carbon and UV component peaked an hour earlier in Strabane than they did in Dunmurry. Also there was still a signature of domestic emission during summer in Strabane that was not present at Dunmurry.

Rural sites

The rural background site concentrations were lower than the other site classifications and without visible morning and evening rush hour peaks. The rise in concentrations in the evening were later than would be expected for a traffic signal and were also seen in the UV component suggesting a domestic heating source. All three rural sites showed lower and more stable concentrations at the weekends with a heating trend seen but no visible effects from traffic.

5.4 COMPARISONS WITH OTHER POLLUTANTS

Comparisons are possible between Elemental Carbon and Black Carbon concentrations at three sites, and between PAH and UV component concentrations at three (different) sites.

Comparisons were also made with particle mass concentration measurements where these instruments were collocated with the Aethalometer.

5.4.1 Elemental Carbon

Daily Elemental Carbon (EC) measurements are made at the North Kensington, Marylebone Road and Chilbolton sites by the Particle Concentration and Number Network⁸. Aethalometer concentrations (BC) at these sites have been averaged into daily measurements and plotted as scatter plots against the elemental carbon (EC) concentrations in Figures 34 to 37. The regression is calculated according to the Reduced Major Axis (RMA) method⁹, which is based on minimising the product of the x and y deviations between the data values and "fitted values" instead of the least squares method, which minimises the sum of the squared deviations between the dependent variable (y) and the "fitted values". RMA is better suited to air quality measurements as pollutant concentrations are often related to each other, so there is no real separation into dependent and independent variables.

⁸ TOMPKINS, J. et. al. Draft NPL REPORT, 2018 Annual Report for Airborne Particulate Concentrations and Numbers in the United Kingdom (phase 3), October 2019.

⁹ AYERS, G.P. Comment on regression analysis of air quality data. Technical Note, *Atmospheric Environment*, 2001, **35**, 2423-2425.

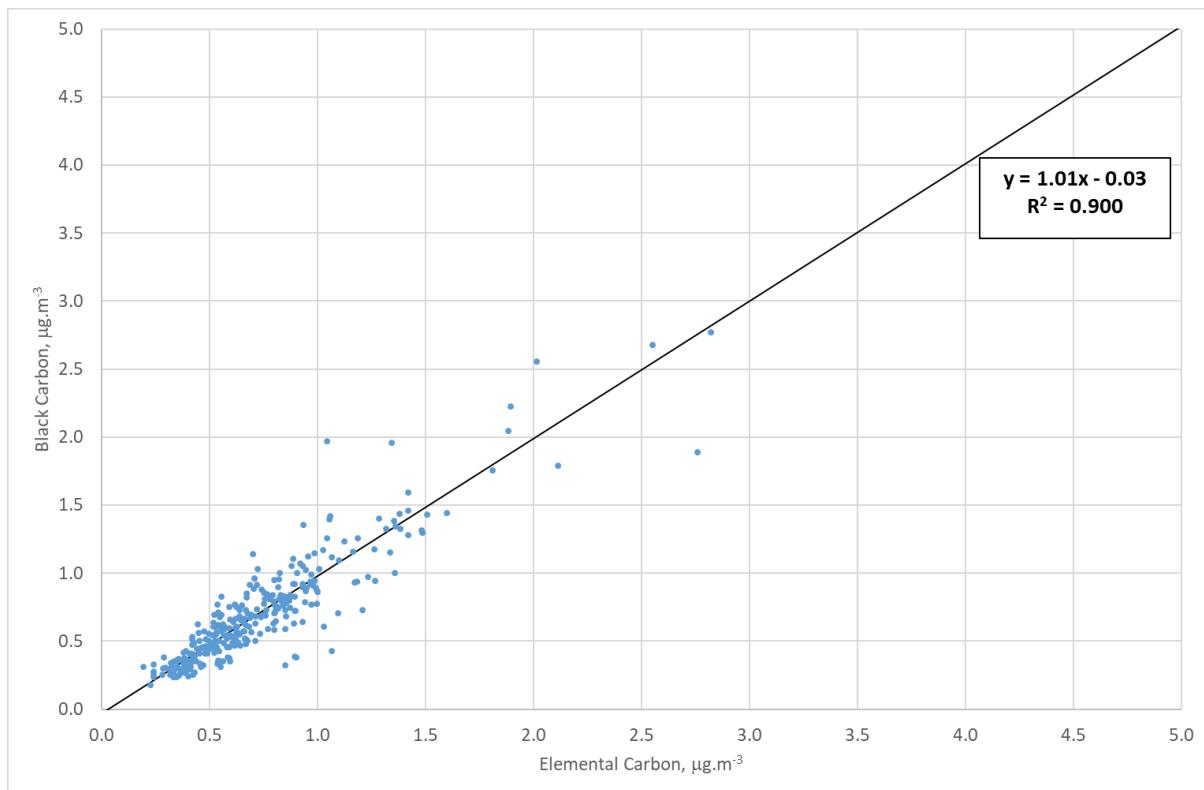


Figure 34 2018 EC and BC Measurements at North Kensington

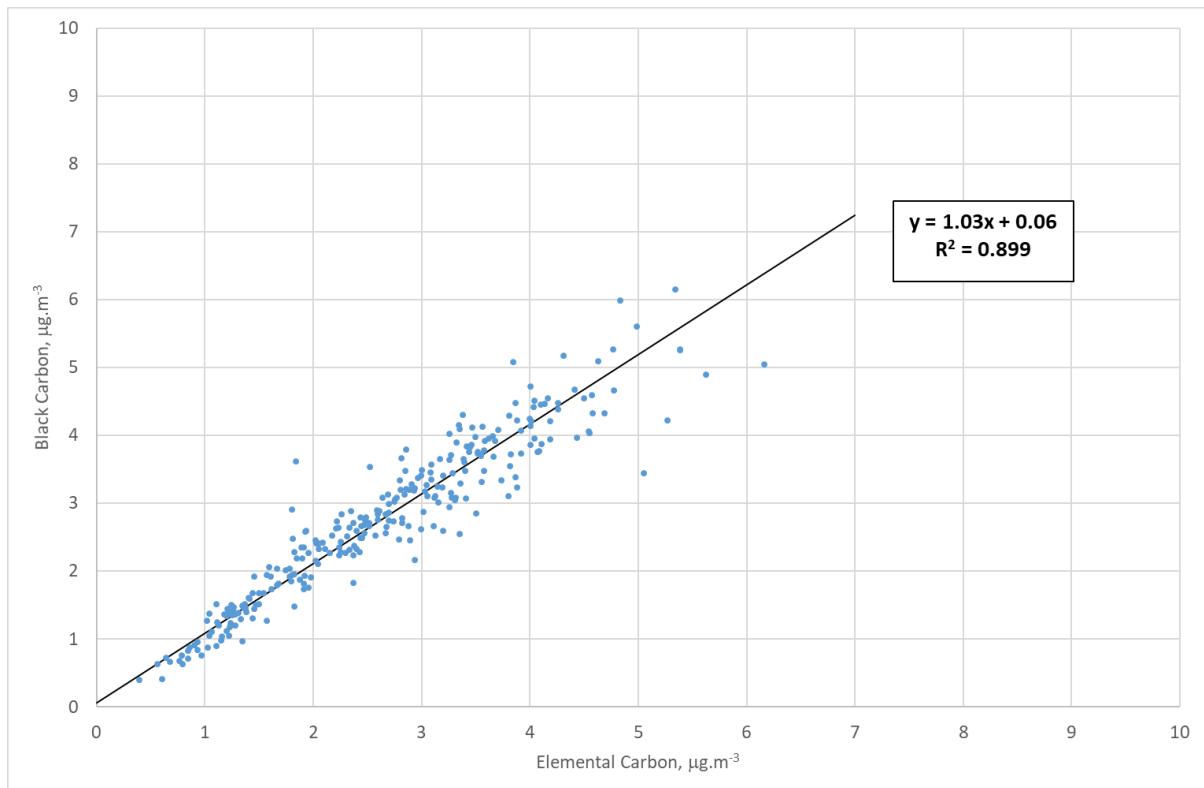


Figure 35 2018 EC and BC Measurements at Marylebone Road

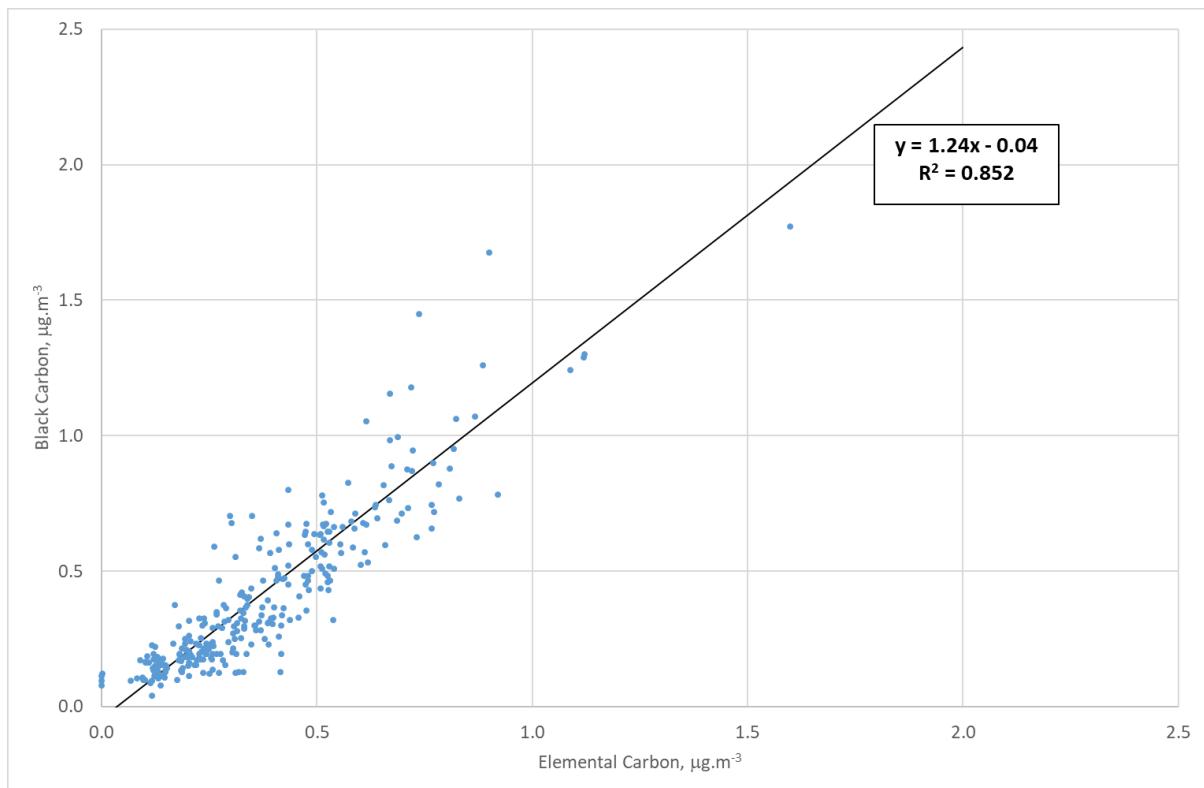
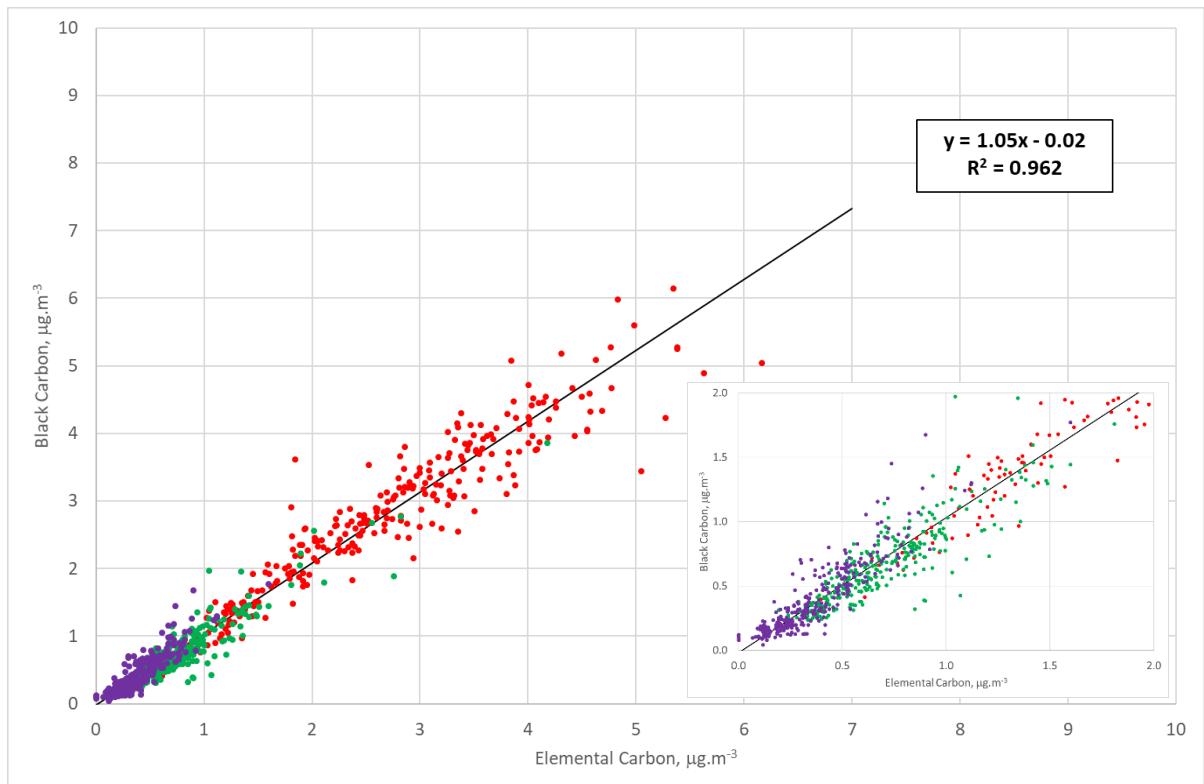


Figure 36 2018 EC and BC Measurements at Chilbolton



Key: Red = Marylebone Road, Green = North Kensington & Purple = Chilbolton

Figure 37 2018 EC and BC Measurements for all 3 sites. The inset graph shows points close to the origin.

It can be seen that there were good linear relationships ($R^2 > 0.85$) between the Elemental Carbon and Black Carbon concentrations measured at all three sites. The regression parameters between Black Carbon and Elemental Carbon were somewhat different between sites, although the differences in concentration ranges mean that the sites do not necessarily represent different populations of data, as shown in Figure 37. In all cases the intercept value was relatively small and at North Kensington and Marylebone Road was within a standard deviation of zero. This indicates that there was no significant zero offset between the two methods, which are based on entirely independent principles. The annual regression results (all RMA) are shown in Table 13.

Year	Harwell/ Chilbolton		North Kensington		Marylebone Road	
	Relationship	R ²	Relationship	R ²	Relationship	R ²
2009	N/A*	N/A	1.05 x + 0.20	0.858	1.36 x - 0.69	0.776
2010	1.32 x + 0.06	0.555	1.37 x - 0.32	0.734	1.28 x + 0.56	0.946
2011	1.52 x + 0.18	0.844	1.26 x + 0.07	0.810	1.50 x - 0.35	0.924
2012	1.84 x + 0.06	0.908	1.42 x + 0.17	0.906	1.43 x + 0.01	0.898
2013	1.74 x + 0.17	0.865	1.59 x + 0.33	0.871	1.47 x + 0.39	0.679
2014	2.02 x - 0.01	0.802	1.68 x - 0.00	0.872	1.32 x + 0.25	0.819
2015	1.67 x - 0.03	0.833	1.64 x - 0.17	0.893	1.23 x + 0.28	0.901
2016	1.31 x + 0.03	0.887	1.08 x + 0.03	0.958	1.25 x + 0.26	0.953
2017	0.92 x + 0.02	0.827	1.04 x - 0.01	0.939	1.15 x + 0.02	0.902
2018	1.24 x - 0.04	0.852	1.01 x - 0.03	0.900	1.03 x + 0.06	0.899

Note: There was not enough BC data collected at Harwell in 2009 to form a reliable relationship as the Aethalometer was only installed in November 2009. The rural data from 2016 onwards are from Chilbolton and so may not be directly comparable to previous years.

Table 13 Relationships between Black Carbon and Elemental Carbon over the period 2009 – 2018

It can be seen that the slopes, which show the measured amounts of Black Carbon relative to Elemental Carbon, had been roughly constant at Marylebone Road from 2009 to 2014 at 1.4 ± 0.1 . This dropped to around 1.2 in 2015, remained at a similar value in 2016 and then dropped again in 2017 and 2018, indicating a possible downwards trend. The parameters at Harwell and North Kensington have been more variable, as would be expected because of the lower concentrations.

5.4.2 Polycyclic Aromatic Hydrocarbons (PAH)

Monthly concentrations of benzo[a]pyrene are measured at Auchencorth Moss, Ballymena, Dunmurry Kilmakee, Glasgow Townhead, Chilbolton and Marylebone Road under the UK PAH Network¹⁰. The BaP and UV component have similar emission sources and a 2016 paper exploring the relationship between collocated Aethalometer UV component measurements and Defra PAH Network BaP measurements, by R Brown *et al*¹¹, determined the following quadratic relationships between the two pollutants.

$$BaP = a \cdot UV^2 + b \cdot UV + c \quad \text{Equation 2}$$

Where

BaP = predicted BaP concentration in $\text{ng} \cdot \text{m}^{-3}$;

UV = measured UV component concentration in $\mu\text{g} \cdot \text{m}^{-3}$

Table 14 gives the coefficients a, b and c for the different site types, as given in the paper.

Site type	Class	a	b	c
Marylebone Road	MY	0.000	0.947	0.076
Northern Ireland	NI	0.285	0.934	0.000
Rural	R	0.902	0.293	0.000
Urban and Roadside	UR	2.369	0.107	0.000
All sites		0.343	0.827	0.001

Table 14 Coefficients for predicting BaP concentrations from measured UV component concentrations

Using this relationship, Table 15 shows the measured and predicted annual 2018 BaP concentration at each Aethalometer site based on the measured annual UV component concentration.

10 <https://uk-air.defra.gov.uk>

11 BROWN, R., BUTTERFIELD, D., GODDARD, S., HUSSAIN, D., QUINCEY, P. and FULLER, G. Wavelength dependent light absorption as a cost effective, real-time surrogate for ambient concentrations of polycyclic aromatic hydrocarbons. *Atmospheric Environment*, 2016, **127**, 125-132.

Site	Class	UV component, $\mu\text{g.m}^{-3}$	Predicted BaP, ng.m^{-3}	Measured BaP
			site specific	ng.m^{-3}
Auchencorth Moss	R	0.02	0.0	0.019
Ballymena	NI	0.41	0.4	0.37
Belfast Centre	NI	0.32	0.3	
Birmingham Ladywood	UR	-	-	-
Birmingham Keeley Street	UR	0.21	0.1	
Cardiff Centre	UR	0.22	0.1	
Detling	R	0.20	0.1	
Dunmurry Kilmakee	NI	0.34	0.3	0.26
Glasgow High Street	UR	0.17	0.1	
Glasgow Townhead	UR	0.08	0.0	0.065
Chilbolton	R	0.15	0.1	0.061
Marylebone Road	MY	0.25	0.3	0.12
North Ken	UR	0.13	0.1	
Strabane	NI	0.82	1.0	

Note: Birmingham Ladywood site was not operational in 2018.

Table 15 Predicted 2018 annual BaP concentration based on measured UV component concentrations

It can be seen that the predicted BaP concentration is within 0.2 ng.m^{-3} of the measured value at all of the sites where BaP is measured. As the 2018 Black Carbon and BaP data were not included in the dataset to calculate the a, b & c coefficients in Equation 2, it shows the good predictive quality of the relationship between UV component and BaP.

Using the relationship above for Northern Ireland sites, Table 16 gives the predicted BaP concentrations at Strabane for the last 7 years. This site has the highest measured UV components and is therefore the most likely of the sites to be close to the BaP target values. The 2016 data has been omitted because the incomplete data gives an average that is not representative.

Year	UV component concentration	Predicted BaP concentration ng.m^{-3}
2009	0.9	1.1
2010	1.3	1.7
2011	0.8	0.9
2012	1.1	1.4
2013	1.2	1.5
2014	1.1	1.4
2015	0.9	1.1
2016	-	-
2017	0.9	1.0
2018	0.8	1.0

Table 16 Predicted BaP concentrations from UV component concentration at Strabane for the period 2009 to 2018.

Six out of these nine years have had predicted BaP concentrations above the 1.0 ng.m^{-3} target value in the EC Directive 2004/107/EC¹² relating to ambient BaP concentrations. The average concentration over these 9 years is predicted to be 1.2 ng.m^{-3} . There will, however, be significant uncertainty attached to these values.

5.4.3 Particulate Mass Concentration

The annual average particulate mass concentration was compared with the Black Carbon concentration at collocated sites where automatic particulate mass instrumentation was installed. The results are shown in Table 17.

Site	BC $\mu\text{g.m}^{-3}$	TEOM FDMS PM_{10} $\mu\text{g.m}^{-3}$	TEOM FDMS $\text{PM}_{2.5}$ $\mu\text{g.m}^{-3}$	MetOne BAM PM_{10} $\mu\text{g.m}^{-3}$	Percent BC PM_{10} %	Percent BC $\text{PM}_{2.5}$ %
Marylebone Road	2.8	24	16		12	18
Birmingham Keeley Street	1.8	18	12		10	15
Glasgow High Street	1.2	14	7		9	17
North Kensington	0.8	14	9		6	9
Glasgow Townhead	0.6	11	7		5	9
Cardiff	0.7	17	10		4	7
Belfast Centre	1.2	16	10		8	12
Chilbolton	0.4	12	9		3	4
Auchencorth Moss	0.1	7	5		1	2
Detling*	0.4	20			2	
Strabane*	1.0			15	7	

Note: * Local Authority run site for PM; may not have identical QAQC procedures to AURN datasets.
Grey shaded cells indicate no measurements were made.

Table 17 Comparison of Annual Black Carbon and Particulate Mass Concentrations

It can be seen that the PM_{10} and $\text{PM}_{2.5}$ mass concentration measured at Marylebone Road, Birmingham Keeley Street and Glasgow High Street sites had a much higher percentage of Black Carbon than the other sites. Black Carbon therefore represented a large proportion of the total particulate mass at sites influenced by road traffic emissions. It is noticeable, however, that these levels are much lower in comparison to the results from 2017. The most significant decrease has been observed at Marylebone Road site which suggest that any substantial reduction in Black Carbon emissions from road traffic lead to a measurable reduction in PM_{10} and $\text{PM}_{2.5}$ mass concentrations as found by Font⁴ *et al* (Figure 23). At the Belfast Centre site, the Black Carbon annual average concentration is higher compared to 2017, but it is very likely due to lack of data from the late spring and summer period, where usually the BC concentrations are much lower.

At the rural background sites Black Carbon made up 4% or less of the PM_{10} mass.

12 DIRECTIVE 2004/107/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL, relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air, 15 December 2004.

5.5 TRENDS

Trends in Black Carbon and UV component concentrations are given below.

5.5.1 Trends by Site

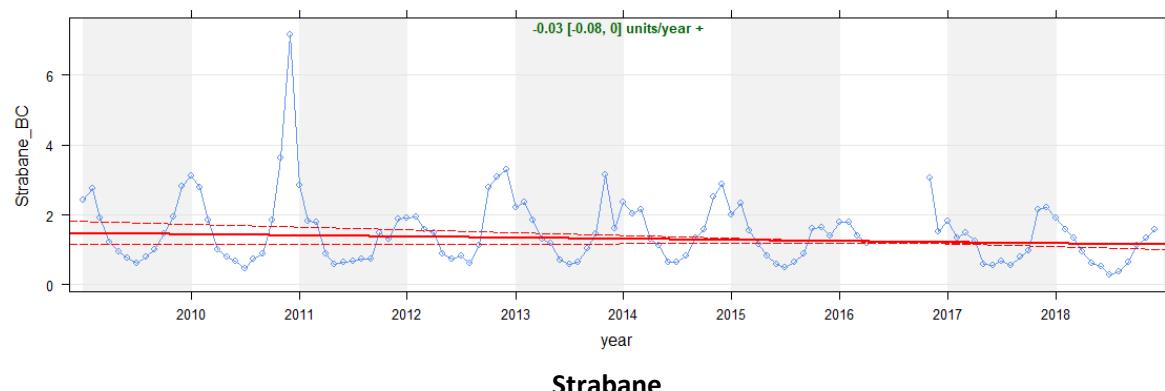
Figures 38 to 39 show the trend in Black Carbon and UV component concentrations from the long-running sites in the Network, as monthly averages over the full calendar years 2009 to 2018. 2016 data has been omitted for Strabane because measurements are not available for the whole year, resulting in a skewed dataset. The Theil-Sen method in OpenAir^{5,6} was used to calculate the regression parameters including slope and uncertainty in the slope.

The Theil-Sen method chooses the median slope among all lines through pairs of two-dimensional sample points. The Theil-Sen estimator tends to yield accurate confidence intervals even with non-normal data and heteroscedasticity (non-constant error variance). It is also resistant to outliers.

Bootstrap resampling provides the confidence interval for the regression slope. For these analyses the 2.5th and 97.5th percentile slopes are taken from all possible slopes.



Figure 38 Black Carbon trends measured at the roadside site, 2009 – 2018



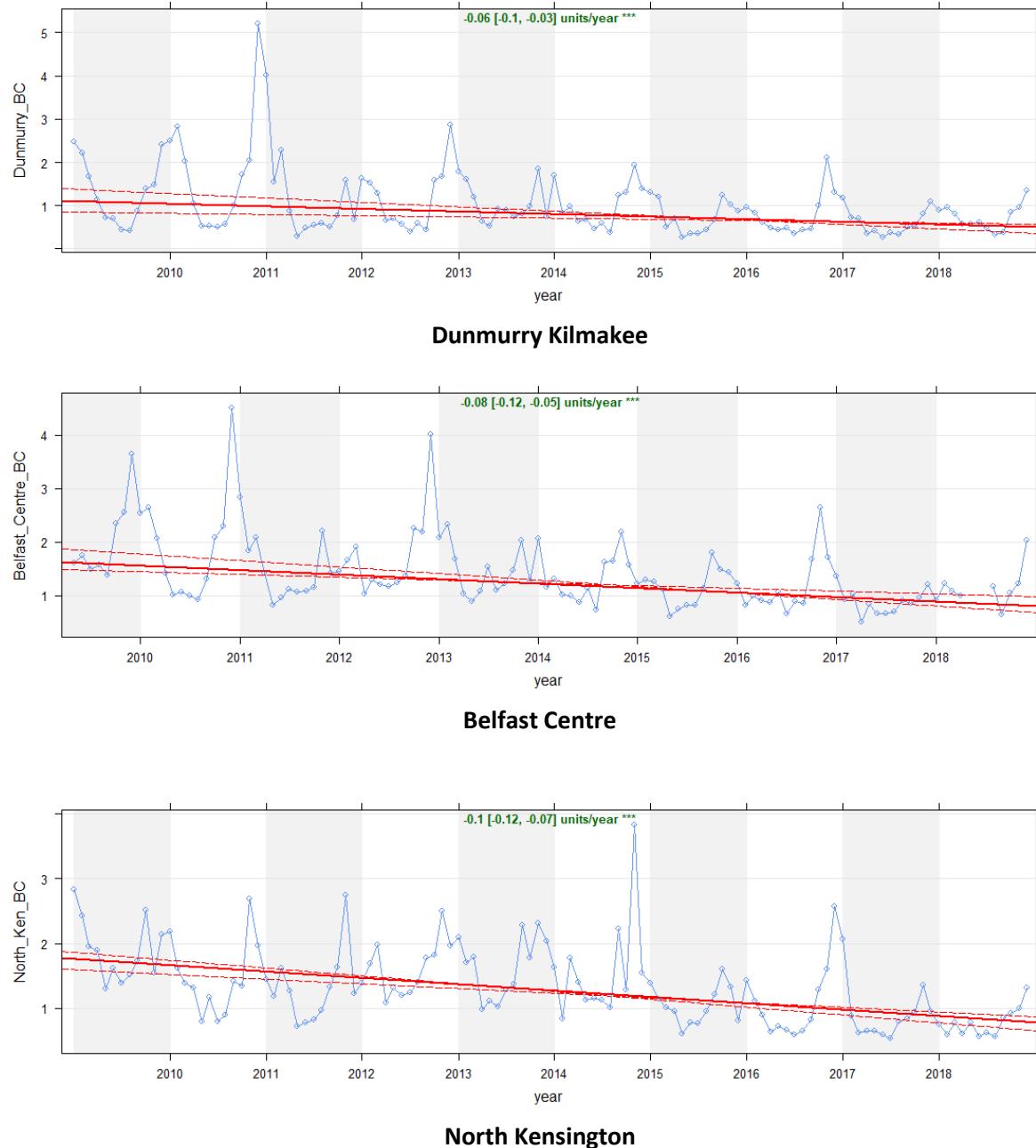


Figure 39 Black Carbon trends measured at urban background sites, 2009 – 2018

Table 18 summarises the trend estimates at each site.

Site	Slope $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Lower limit $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Upper limit $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Slope significant
Roadside				
Marylebone Road	-0.84	-0.92	-0.74	Y
Urban Background				
Strabane	-0.03	-0.08	0.0	N
Dunmurry	-0.06	-0.10	-0.03	Y
Belfast Centre	-0.08	-0.12	-0.05	Y
North Kensington	-0.10	-0.12	-0.07	Y

Table 18 Summary of Black Carbon trends

Over the period 2009 to 2018 all of the long-running sites in the network apart from Strabane have shown a significant downward trend in Black Carbon concentrations. The decrease at Marylebone Road is much larger than the other sites and Black Carbon concentrations have been falling consistently since 2011. The 2018 annual mean concentration was less than half that of 2011. The concentrations of Elemental Carbon made at Marylebone Road have followed a similar trend. Figure 40 shows the annual Black Carbon and Elemental Carbon concentrations along with the average daily traffic flow past the site¹³.

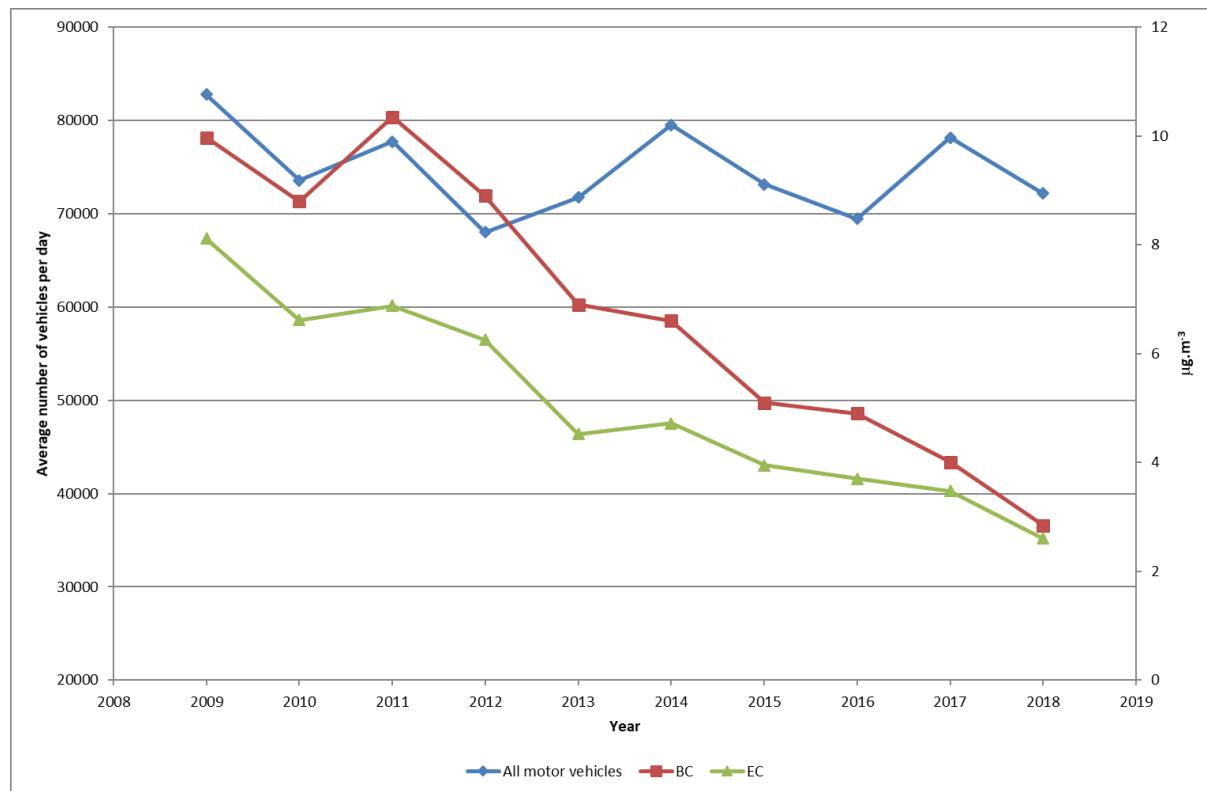


Figure 40 Annual Average Black Carbon, Elemental Carbon and Motor Vehicles per day at Marylebone Road for the period 2009 – 2018

13 <http://www.dft.gov.uk/traffic-counts>

It can be seen that the changes in Black Carbon and Elemental Carbon concentrations followed changes in the total traffic flow for the years 2009 to 2012 but since 2013 they have decreased while traffic flows have been effectively constant. This would indicate that Black Carbon emissions per vehicle have decreased over the last 5 years. The drop in emissions per vehicle type may be linked to the increased proportion of low emission buses (hybrid, electric and fuel cell / hybrid) in the London bus fleet¹⁴. Table 19 shows the composition of the London bus fleet over the period 2010 to 2018. The bottom row of the table shows the percentage of low emission buses, which is a combination of the hybrid, electric and fuel cell / hybrid bus numbers. In addition, all of London's Euro II and III diesel buses were retro-fitted with engine exhaust particulate filters by the end of 2011, which would have also reduced Black Carbon emissions.

Also, in 2012 the vehicle types affected by the London Low Emission Zone (LEZ) were increased to include large vans, minibuses and other specialist diesel vehicles. Vehicles entering the LEZ must be Euro III or higher to be compliant with the requirements. In addition, the requirements for lorries, buses, coaches, licensed private hire and specialist heavy vehicles changed from Euro III to Euro IV. These changes may have also reduced Black Carbon emissions from road transport.

Bus Type	Drive train type	Number of buses									
		2010	2011	2012	2013	2014	2015	2016	2017	2018	
New Routemaster	Hybrid	0	0	5	8	168	432	736	953	1,000	
Routemaster	Diesel	18	18	19	20	19	19	10	10	10	
Artic	Diesel	320	260	0	0	0	0	0	0	0	
Single deck	Diesel	2,676	2,670	2,661	2,608	2,606	2,662	2,617	2,612	2,587	
	Fuel Cell/Hybrid	0	5	5	5	8	8	8	8	10	
	Hybrid	27	27	33	28	23	23	18	18	13	
	Electric	0	0	0	0	2	8	17	66	91	
Double deck	Diesel	5,554	5,487	5,787	5,696	5,296	5,026	4,794	4,380	3,453	
	Hybrid	29	79	233	352	643	799	981	1,564	2,227	
	Electric	0	0	0	0	0	0	5	5	5	
TOTAL		8,624	8,546	8,743	8,717	8,765	8,977	9,186	9,616	9,396	
% low emission		0.65	1.30	3.16	4.51	9.63	14.15	19.21	27.18	35.61	

Table 19 Composition of London bus fleet, 2010 to 2018^[13]

Figure 41 is Figure 40 replotted with the number of motor vehicles per day passing the Marylebone Road monitoring site replaced by [100 – percentage of low emission buses in the London bus fleet].

14 Number of Buses by Type of Bus in London, tfl-buses-type.xls, London Datastore, <https://data.london.gov.uk>

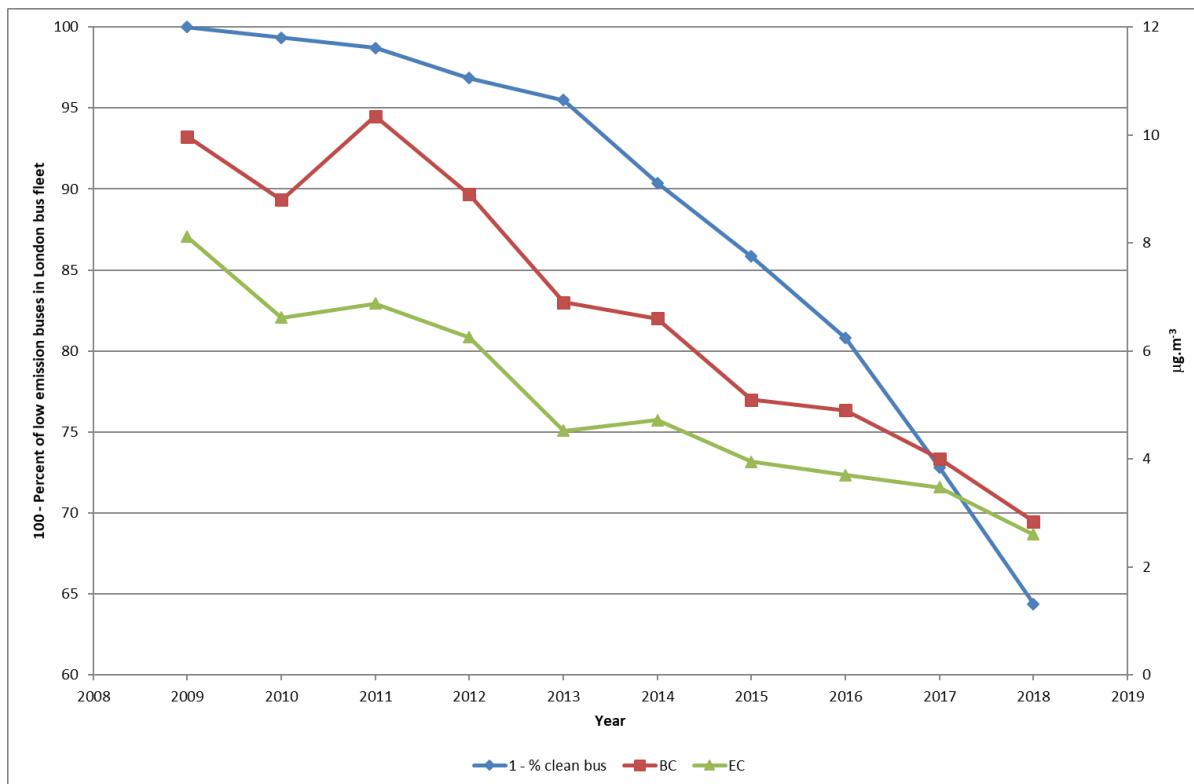


Figure 41 Annual Average Black Carbon, Elemental Carbon and 100 – percentage of low emission buses in the London bus fleet for the period 2009 – 2018

Assuming that the mix of buses passing the Marylebone Road site is representative of the whole London bus fleet, it is likely that the increase in low emission buses, and changes to the LEZ from 2012, have led to reduced Black Carbon and Elemental Carbon concentrations. The fall from 2012 to 2015 appears to mirror the clean buses, but as the number of these continues to rise sharply, the BC and EC concentrations have fallen less sharply. This suggests that there is another factor preventing concentrations continuing to fall and further increase in clean buses will not have a significant effect.

Figures 42 to 43 show the UV component trends.

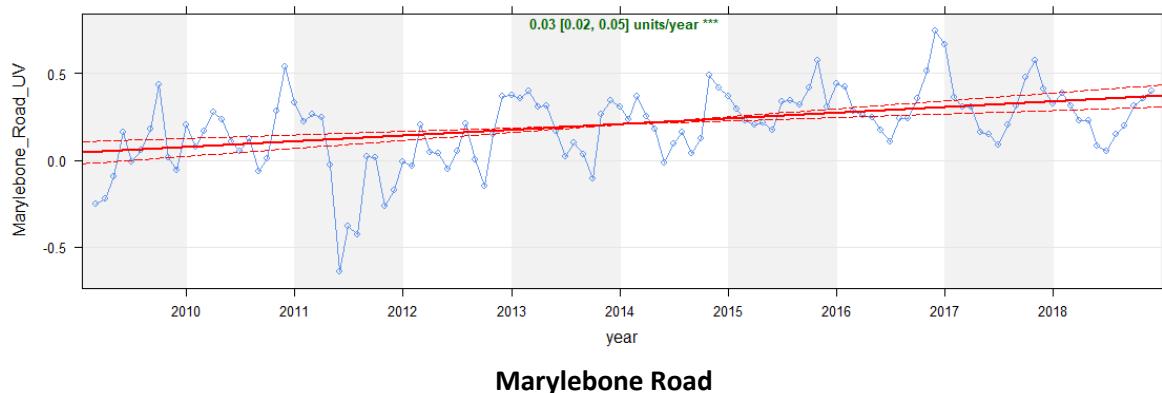


Figure 42 UV component concentrations measured at roadside sites, 2009 – 2018

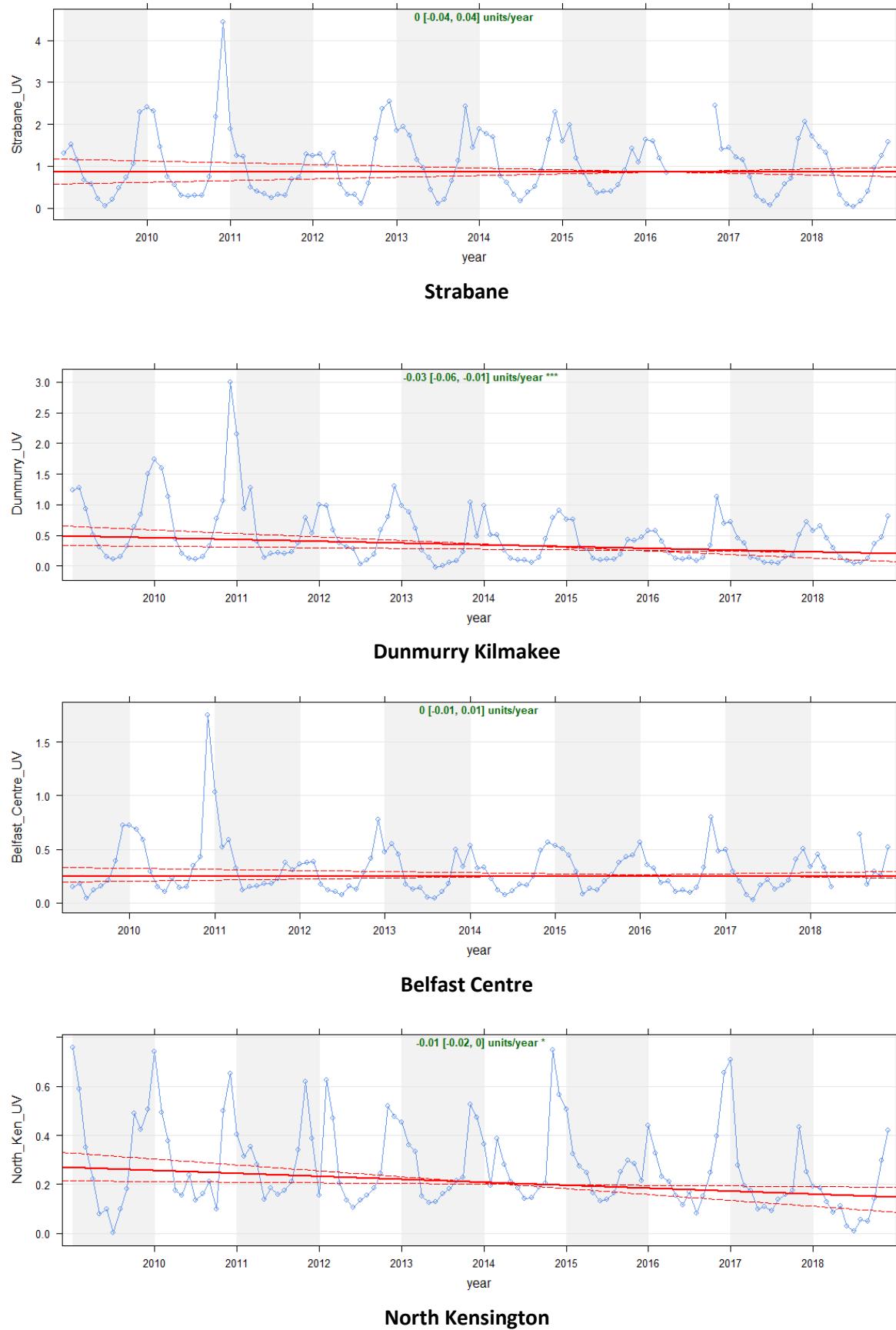


Figure 43 UV component concentrations measured at urban background sites, 2009 – 2018

Table 20 summarises the slopes at each site.

Site	Slope $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Lower limit $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Upper limit $\mu\text{g.m}^{-3}.\text{year}^{-1}$	Slope significant
Roadside				
Marylebone Road	0.03	0.02	0.05	Y
Urban Background				
Strabane	0.00	-0.04	0.04	N
Dunmurry	-0.03	-0.06	-0.01	Y
Belfast Centre	0.00	-0.01	0.01	N
North Kensington	-0.01	-0.02	0.00	N

Table 20 Summary of UV component trends

The Marylebone Road UV component concentration showed a significant upward trend over the period 2009 to 2018, this was probably due to the reduced Black Carbon concentrations over the last five years. The Aethalometer measures the UV component by the difference between the BC and UV channel. As Black Carbon has fallen from relatively high values, it is to be expected that the calculation of the small UV component will be affected. It is unlikely that the UV component emissions across London have risen in the last three years due to domestic fuel usage, as the Black Carbon concentrations at North Kensington, which were not dominated by traffic, have not shown an increase. This trend should be treated with caution due to the low concentrations involved.

There was also a significant downward trend in the Dunmurry data. This was partly driven by the warmer and wetter than usual 2015-2016 winter, but has continued despite next winters levels being more in line with previous years, suggesting that the falling concentrations are an overall trend.

To show how pollutant concentrations can depend strongly on the weather, the 2009-2018 UV component concentrations at Strabane, which were strongly affected by domestic solid fuel use, are plotted in Figure 44, along with average temperature for same period. Only the valid Strabane data from 2016 have been included and the gap is due to the instrument fault during that year. Temperature measurements from Armagh have been used as this is the nearest Met Office site with a long time series.

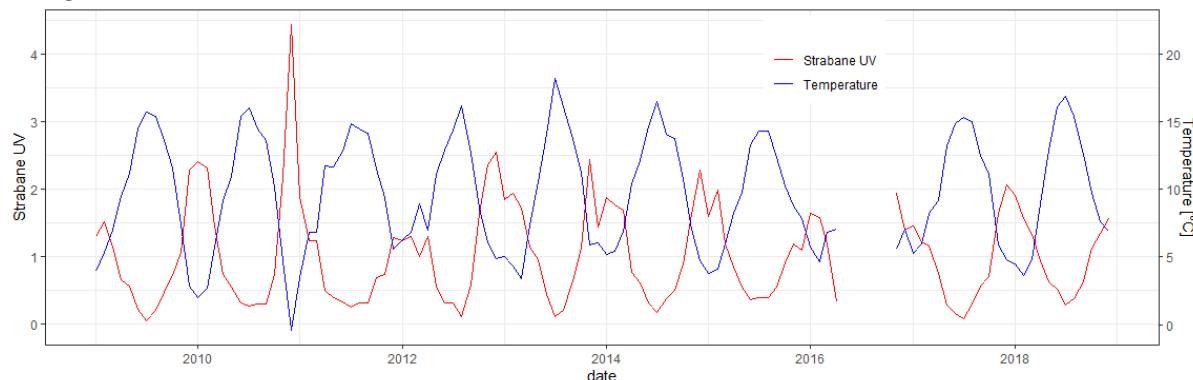


Figure 44 Strabane monthly UV component concentration and average ambient temperature for 2009 – 2018

It can be seen that the UV component concentration was inversely related to the average ambient temperature. This was due to the main source of UV component emissions being local domestic

heating in Strabane. This was evident in both the winter and the summer indicating that there were still solid fuel emissions in the summertime. The relationship is shown in Figure 45 as a scatter plot.

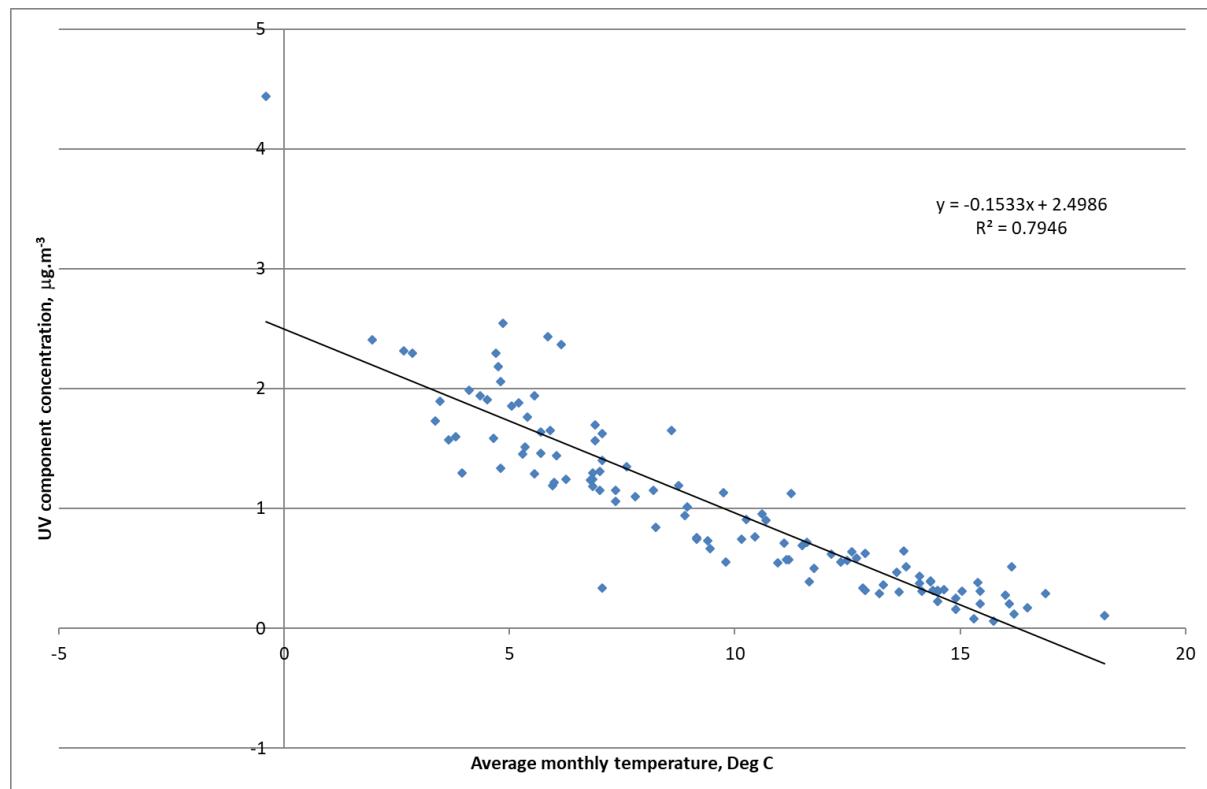


Figure 45 Scatter plot of monthly UV component concentration versus ambient temperature at Strabane over the period 2009 -2018

It can be seen that there was a clear linear relationship between increased UV component concentrations with a drop in ambient temperature, due to the increase in fuel usage in cold weather periods. There is an indication that the UV source became significant when average temperatures were below 15°C, linking the UV component to fuels used for domestic heating systems.

5.5.2 Trends over the Network as a Whole

Figure 46 and 47 show the Network annual mean and median concentrations for Black Carbon and UV component for the subset of sites that have been continuously running since 2009 (Belfast Centre, Birmingham Tyburn UB, Dunmurry, Harwell/Chilbolton, Marylebone Road, North Kensington and Strabane). The median concentration is shown to remove the influence of large changes in a single site (i.e. Marylebone Road) that would skew the overall result for the Network. Due to the inclusion of this median concentration, and as the data shown is an average of a large number of sites, data from all of these sites is included even if site moved or breakdowns meant they are not directly comparable year to year.

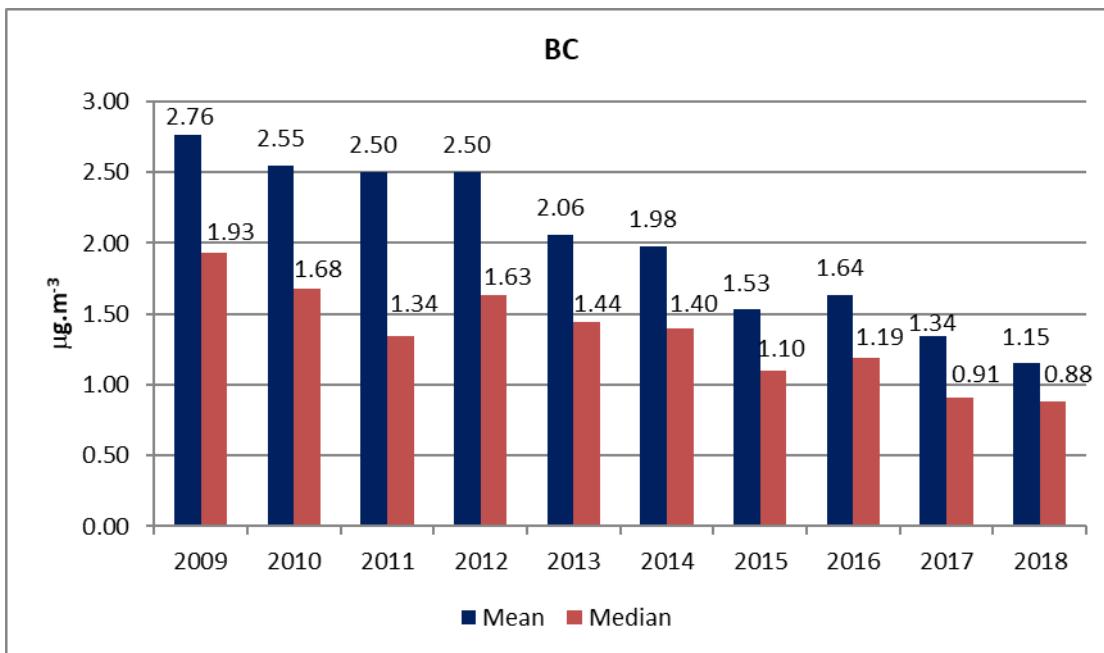


Figure 46 Annual average Black Carbon concentrations for long-term sites

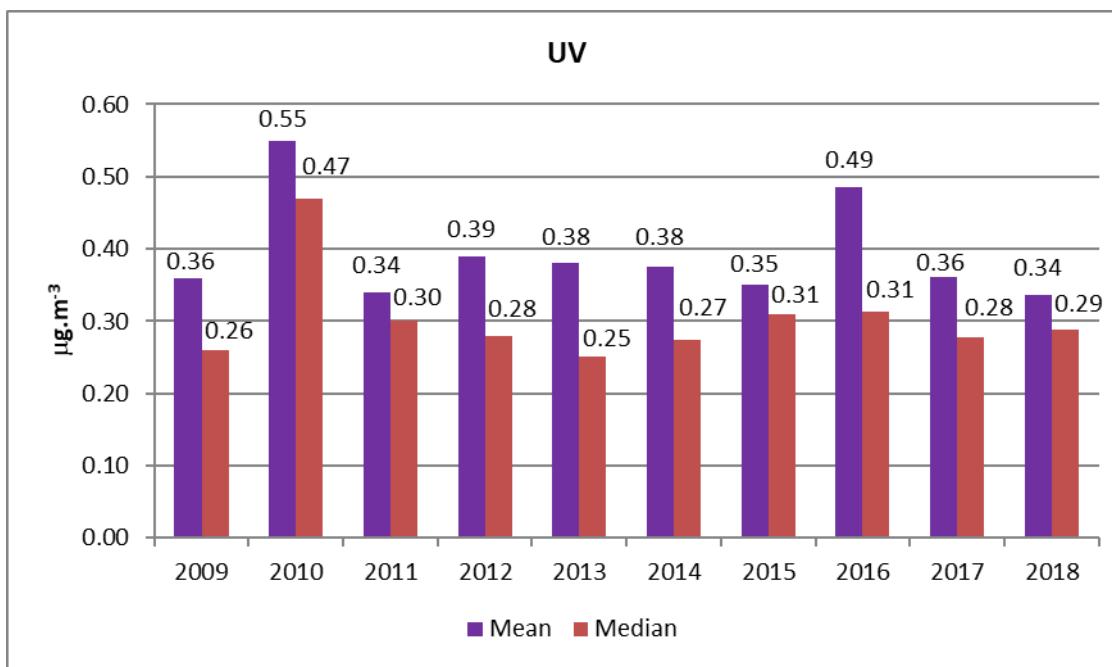


Figure 47 Annual average UV component concentrations for long-term sites

It can be seen that the network annual mean for Black Carbon has been generally decreasing over this time. This overall decrease is mainly driven by the drop in concentrations at Marylebone Road.

The annual average UV component concentrations varied from year to year with the maximum concentration seen in 2010. 2010 had very cold and snowy periods at the start and end of the year, especially in Northern Ireland, as shown in Figure 44, so elevated concentrations were probably due to domestic emissions.

6.0 EUROPEAN STANDARDISATION

The European standardisation body CEN has formulated a European Standard on the measurement of Elemental Carbon and Organic Carbon deposited on filters. This standard was published in March 2017 as EN 16909.

The CEN working group, CEN TC 264 WG 35, is now working to bring automatic black carbon analysers such as the Aethalometer used on the network within the standardisation process. A new work item for a draft standard has been approved by CEN.

7.0 RELATED RESEARCH ACTIVITIES

7.1 Airborne particles from wood burning in UK cities

Analysis of Black Carbon Network measurements were undertaken to determine the contribution of wood burning to PM in the UK from 2009 to 2016¹⁵. Funded by Defra, the study was prompted by new information that domestic wood burning in the UK had been systematically underestimated by a factor of three in the national emissions inventory and there was therefore an important need to properly quantify its contribution to the urban atmosphere. In addition to measuring black carbon concentrations, Aethalometers provide information on the light absorption of the collected particles at two different wavelengths. These measurements were processed to quantify the particle matter from wood burning.

As expected, air pollution from wood burning was found to be greatest in winter and almost absent in summer. In most cities wood burning PM concentrations were greater in evening and weekends, indicating residential combustion. Coupled with a poor correlation with daily temperature, this suggested that current urban wood burning was in large part decorative and was not being used for primary heating. Looking at annual averages, it was estimated that wood burning was the source of between 23 and 31 % of the urban derived PM_{2.5} in London and Birmingham, making control of wood burning an important urban issue.

Looking forward, large increases in biomass burning are projected from energy scenarios over the next two decades (Williams et al., 2017). An increase in the coverage of the aethalometer network would enhance our ability to track the impact of these changes. A surveillance programme would also need to include long-term measurements with multiple methods at key locations.

7.2 The EMPIR Black Carbon project

The EMPIR Black Carbon project, which will run from July 2017 to June 2020, is being lead by NPL¹⁶ and aims to establish SI traceability for optical measurements of Black Carbon. Measurement techniques that do not involve the collection of particles on filters can be made traceable by measuring well-defined physical parameters of the aerosol. Filter based techniques, as used in the Aethalometers, can then be standardised using fully-characterised reference materials. The project will develop and characterise these standard reference materials and validate a field calibration

15 FONT, A., BUTTERFIELD, D., and FULLER, G.W. Airborne particles from wood burning in UK cities. 2017.

https://uk-air.defra.gov.uk/assets/documents/reports/cat05/1801301017_KCL_Wood Burning Report_2017_FINAL.pdf

16 www.empirblackcarbon.com

method using them. Bringing standardisation to these measurements will increase their accuracy and allow for better interpretation and comparison between results. This will have a direct impact on the quality of Black Carbon measurements, including those made for the network.

7.3 Other Papers

Data from the Black Carbon Network are used in research by other institutions, including a study on Black Carbon concentration trends over the UK which identified similar trends and patterns to those described in this report¹⁷.

¹⁷ SINGH, V. et al. Trends of atmospheric black carbon concentration over the United Kingdom. *Atmospheric Environment*, 2018, **178**, 148-157.