

**2015 ANNUAL REPORT FOR THE UK BLACK CARBON NETWORK**

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Approved on behalf of NPLML by Martyn Sené, Operations Director

## EXECUTIVE SUMMARY

This report covers the operation of the UK Black Carbon Network and the data collected by the Network in 2015. The Aethalometer instrument used on the Network makes measurements of Black Carbon (BC) and 'UV component'.

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. By 2007 the network was making measurements at 21 sites. In 2008 the Black Smoke samplers were replaced by model AE22 Aethalometers. The Network was reduced to 20 sites in October 2009, when the Bradford site was closed after Defra reviewed its monitoring requirements. In early 2012 the Network underwent major restructuring to focus the network on targeted monitoring of specific emission sources. In late 2012 Aethalometers were installed at Goonhilly and Ballymena to investigate emissions from shipping and domestic fuel use, respectively. The Norwich Lakenfields site closed in May 2013, the Goonhilly site closed in November 2013 and the Cardiff 12 site closed in June 2014. In March 2015 new sites at Cardiff Centre and Glasgow High Street were installed, now allowing urban and roadside increments to be calculated for London, Birmingham and Glasgow. 14 sites made measurements in 2015.

The 2015 data capture for Aethalometer measurements was 98%. This is a remarkably high figure for an automatic monitoring network and shows the reliability of the Aethalometers and the effectiveness of daily on-line surveillance of the equipment.

2015 urban annual mean Black Carbon concentrations on the Network (with the corresponding 2014 concentrations in brackets) ranged from 0.8 (1.0)  $\mu\text{g.m}^{-3}$  at Dunmurry Kilmakee to 5.1 (6.6)  $\mu\text{g.m}^{-3}$  at Marylebone Road. Harwell (rural background) reported an average concentration of 0.3 (0.5)  $\mu\text{g.m}^{-3}$ , with similar values at the newly installed rural sites at Auchencorth Moss and Detling. The network mean for Black Carbon concentration was 1.3 (1.6)  $\mu\text{g.m}^{-3}$ .

The annual mean UV component concentrations ranged from 0.1 (0.1)  $\mu\text{g.m}^{-3}$  at Auchencorth Moss to 0.9 (1.1)  $\mu\text{g.m}^{-3}$  at Strabane. Roadside sites show many negative spikes in the UV component concentration, thought to be measurement artefacts caused by volatile components in fresh vehicle exhaust plumes. This effect is most prevalent at Marylebone Road. The network mean for UV component concentration was 0.3 (0.2)  $\mu\text{g.m}^{-3}$ . The figures in brackets are again the corresponding concentrations for 2014.

The new 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. The urban increment for Black Carbon was similar for all locations while the roadside increment was roughly proportional to road traffic volumes. Reductions in the roadside increment for Black Carbon have fallen in line with reductions in roadside increment for  $\text{PM}_{2.5}$  concentrations indicating similar emission sources. There was no significant urban or roadside increment in UV component concentration.

Daily averages of the measurements show that the highest concentrations of Black Carbon are found on weekdays, with the weekends generally having lower values. The hourly averages of Black Carbon broadly show a commuter traffic-based signature, with the exception of Strabane, Ballymena and Dunmurry, in Northern Ireland, which show elevated levels from 15:00hrs to 23:00hrs, which is indicative of local residential heating.

From the diurnal plots it can be seen that the main driver behind the UV component concentrations is

domestic fuel use, with elevated concentrations in the evenings at the urban background sites. Sites in Northern Ireland show the largest evening effect due to the higher use of coal, wood and solid fuel for domestic heating. From the daily averages it can be seen that the UV component concentration is fairly consistent on weekdays with greater concentrations at the weekend. It can also be seen that UV component concentrations are inversely proportional to ambient temperature providing 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 North Kensington, Harwell and Marylebone Road, with  $R^2$  values of 0.89, 0.83 and 0.90, respectively. For North Kensington, Harwell and Marylebone Road the slopes were 1.64, 1.67, & 1.23 with intercepts of -0.17, -0.03 & +0.28  $\mu\text{g}/\text{m}^3$ . 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 1.28.

Comparisons between particulate mass concentrations and Black Carbon concentrations showed that Black Carbon makes up a significant proportion of the particulate mass concentration at roadside sites. At Marylebone Road the Black Carbon concentration comprises 21% of the  $\text{PM}_{10}$  concentration and 32% of the  $\text{PM}_{2.5}$  concentration, while at Birmingham Tyburn roadside Black Carbon forms 12% and 18% of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  respectively. Glasgow High Street shows a similar proportion of BC to PM as Birmingham.

The relationship between collocated Aethalometer UV component measurements and Defra PAH Network BaP measurements has been used to predict BaP concentrations at Aethalometer sites where BaP is not directly measured. Only one site (Strabane) is consequently inferred to exceed the EU target value of 1.0  $\text{ng}\cdot\text{m}^{-3}$  for BaP based on this method. Using this relationship to infer BaP concentrations at Strabane in earlier years shows that the BaP target value would have been exceeded in 6 out of the last 7 years.

Monthly means of Black Carbon concentrations were examined over the period 2009 to 2015 to evaluate trends. Marylebone Road, Belfast Centre, Birmingham Tyburn and North Kensington have shown a significant downward trend in Black Carbon concentrations. At the three non-roadside sites this trend is likely to be influenced by meteorological conditions significantly lowering Black Carbon concentrations measured in 2015; with the last quarter being significantly wetter and warmer than in previous years. 2015 was the sixth wettest year since 1910. However, Marylebone Road has been showing reduced Black Carbon concentrations year on year since 2011, with 2015 annual mean concentration roughly half that of 2011. This drop in concentration is 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, HGCs, lorries and vans (Euro III to Euro IV). Low emission buses now make up 14% of the fleet.

The Marylebone Road UV component concentration shows a significant upward trend over the period 2009 to 2015. This trend should be treated with caution due to the low concentrations involved. It is probably related to the reduced Black Carbon concentrations over the last 4 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 2 years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which is not dominated by traffic, have fallen. There is also a significant downward trend in the Dunmurry data which is most likely to be caused by the warmer and wetter 2015 weather than by a significant change in emission sources.

CEN (TC 264 WG 35) are currently formulating a standard for the measurement of Elemental Carbon and Organic Carbon deposited on filters. The new standard should be published in late 2016. As part

of this work there were field validation trials where automatic instruments, including the Aethalometer, were evaluated in parallel with filter samplers.

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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. During 2007 the number of sites in the network expanded from 14 sites to 21 sites, as samplers were installed into mainly Automatic Urban and Rural Network (AURN) sites. By March 2007 all of the 21 sites were operational.

In 2008 the Black Smoke samplers were replaced by model AE22 Aethalometers. The Network was reduced to 20 sites in October 2009 when the Bradford site was closed after Defra reviewed its monitoring requirements.

In January 2012 a new contract was issued by Defra to run the Network, and this involved a major reorganisation of the monitoring sites. The focus of the Network changed to provide targeted monitoring of the major urban conurbations of London, Birmingham and Glasgow, plus additional sites to monitor specific emission sources. This involved the closing of 11 existing sites and the opening of 4 new sites, leaving a Network of 13 sites, plus Harwell which is run as part of Defra's Particle Numbers and Speciation Network. By targeting these urban areas a more detailed apportionment of emissions of Black Carbon emission sources should be provided.

Later in 2012 Defra took up two options to increase monitoring of specific emission sources which resulting in a temporary site being installed at Goonhilly in Cornwall to assess Black Carbon from shipping, and at Ballymena in Northern Ireland to extend the work on monitoring emissions from solid fuel and biomass burning. The Goonhilly monitoring continued until November 2013. Aethalometer measurements stopped at the Norwich Lakenfields site in May 2013 as the site was no longer provided additional information on Black Carbon emissions above that already collected from the other sites on the Network.

### 1.2 BLACK CARBON

Black Carbon (BC) is a measure of airborne soot-like carbon (in  $\mu\text{g}\cdot\text{m}^{-3}$ ) 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 Aethalometers<sup>1</sup>, 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. It consists of pure carbon in several forms. 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, whereas CO<sub>2</sub> has an atmospheric lifetime of more than 100 years.

The terminology to be used for 'Black Carbon' data is currently receiving attention within, for example, the Global Atmosphere Watch special aerosol advisory group. This is mainly concerned with

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<sup>1</sup> P Quincey, A relationship between Black Smoke Index and Black Carbon concentration, Atmospheric Environment 41 (2007) 7964–7968

highlighting the assumptions used to convert optical data to mass concentration data. 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 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,  $\sigma$  [ $\text{m}^{-1}$ ], 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 [ $16.6 \text{ m}^2 \text{ g}^{-1}$ ] 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 Aethalometers run on the Network operate at 2 wavelengths, 880nm and 370 nm. The 880nm wavelength is used to measure the Black Carbon (BC) concentration of the aerosol, while the 370nm 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 370nm source with a mass extinction coefficient of  $39.5 \text{ m}^2 \cdot \text{g}^{-1}$ . 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 Virkkula<sup>2</sup> 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.

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<sup>2</sup> A Virkkula et al, A Simple Procedure for Correcting Loading Effects of Aethalometer Data, Journal of Air and Waste Management Association, 57:1214-1222, 2007

### 1.3.2 Sampling

At all sites, ambient air is 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 is made by a PM<sub>2.5</sub> 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.

## 2.0 NETWORK INFRASTRUCTURE

The following sections present the design of the Network, describe its operation and the changes to the Network in 2015.

### 2.1 NETWORK SITES AND DESIGN

Figure 1 shows the locations of the Aethalometers during 2015. 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.

| Conurbation | Site Name                    | Site Classification |
|-------------|------------------------------|---------------------|
| Glasgow     | Auchencorth Moss             | Rural Background    |
|             | Glasgow Townhead             | Urban Background    |
|             | Glasgow High Street          | Traffic             |
| Birmingham  | Harwell                      | Rural Background    |
|             | Birmingham Tyburn Background | Urban Background    |
|             | Birmingham Tyburn Roadside   | Traffic             |
| London      | Harwell                      | 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 installation of a site at Glasgow High Street was completed on 4<sup>th</sup> March after delays due to planning restrictions.

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                  |

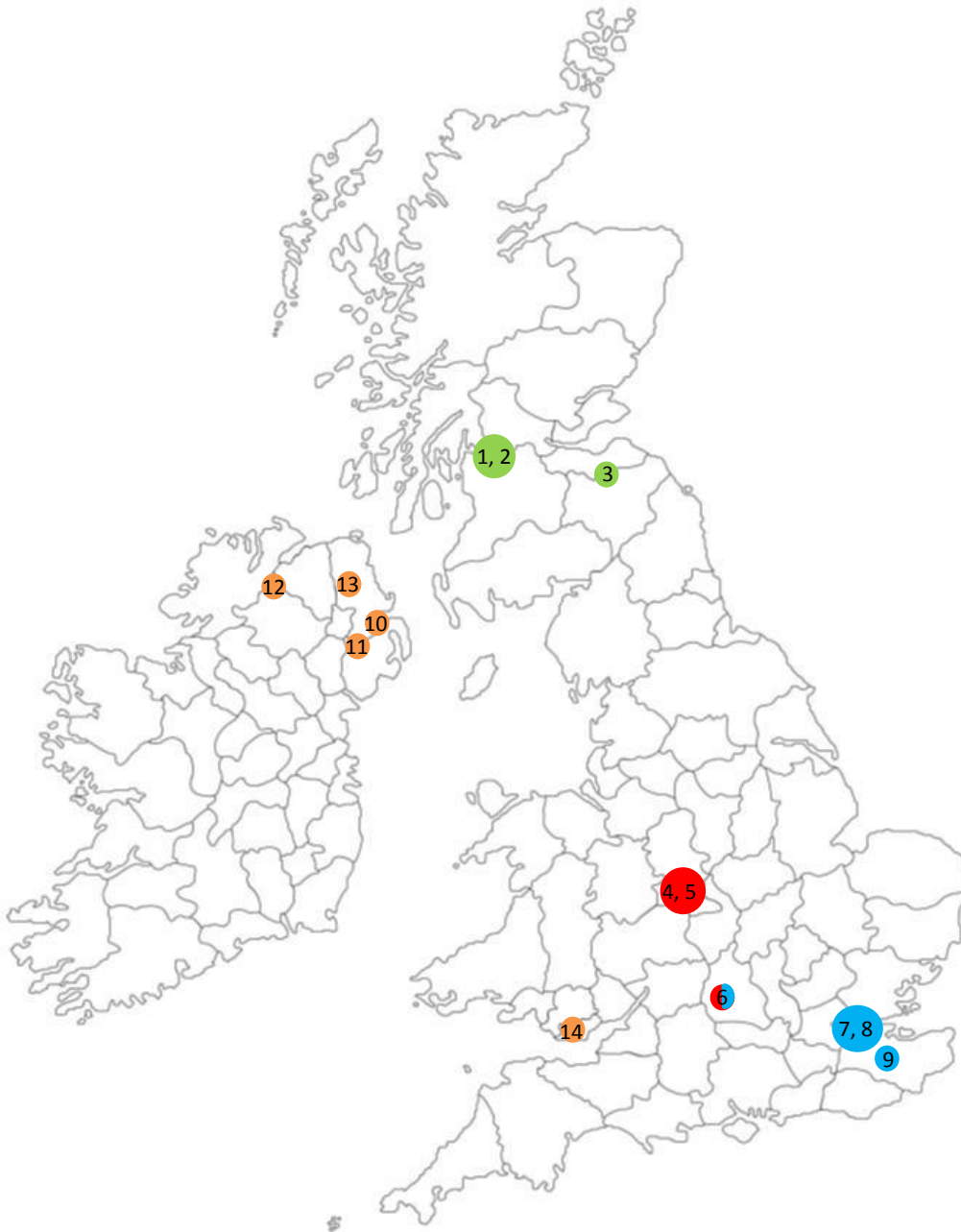
**Table 2 Sites to measure non-traffic related emission sources**

The Cardiff Centre site was commissioned on 13<sup>th</sup> March 2015 as a replacement for the Cardiff 12 site which was closed in June 2014. The gap in the monitoring was due to the Cardiff Centre site having to be upgraded with a larger enclosure to house new monitors.

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

**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 Tyburn Roadside   |
|   | 5   | Birmingham Tyburn Background |
| Birmingham Urban Area + London Urban Area | 6   | Harwell                      |
| 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               |



**Figure 1** Sites on the BC Network during 2015  
Key on previous page

## 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 CMCU 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.

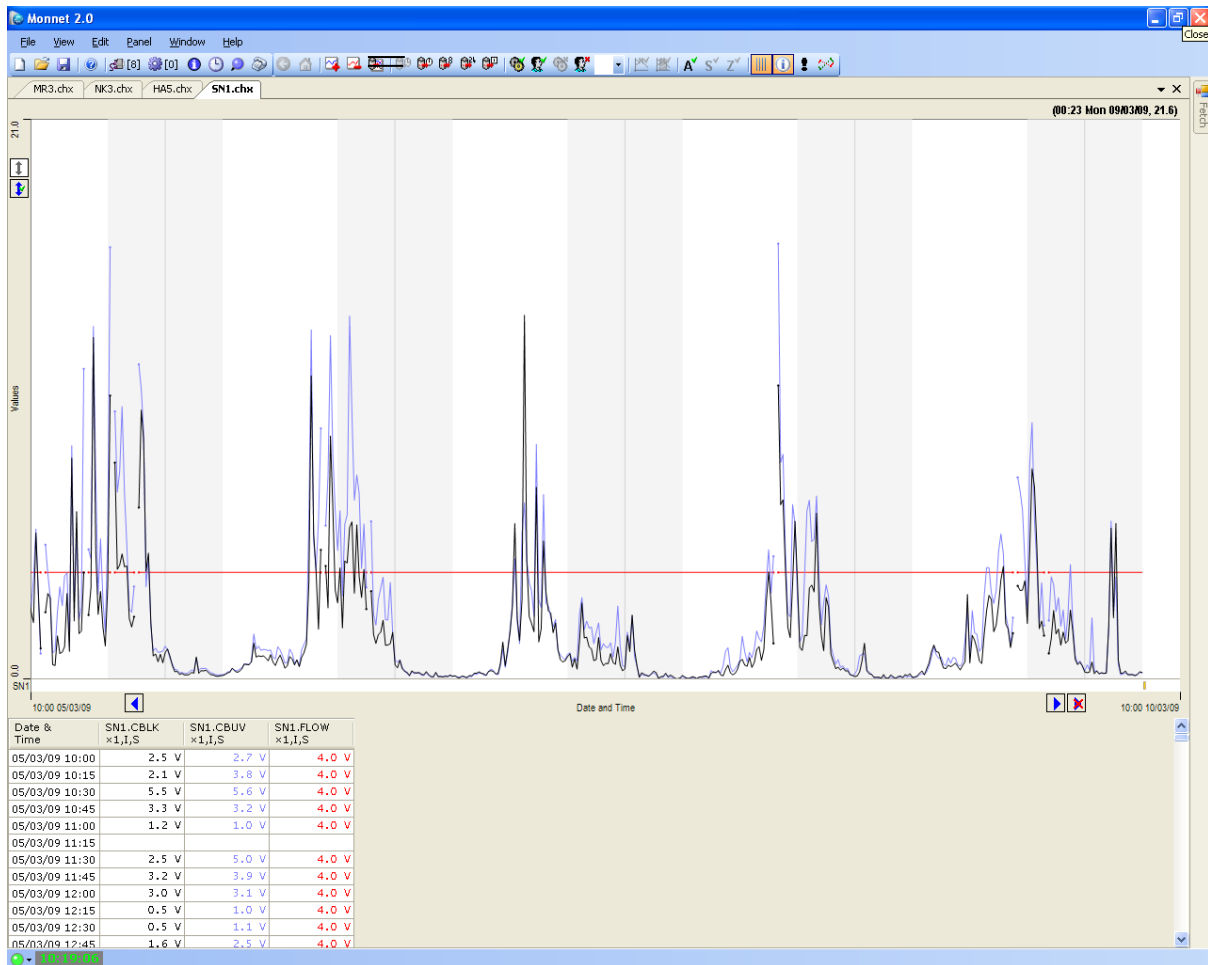
Measurements are collected from the 14 sites on the Network on a daily basis. Measurements of Black Carbon, UV carbon, flow, raw attenuation signals and tape life since the last data collection are requested from the Aethalometer and automatically loaded into King's database. The 5 minute mean measurements 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 is stable for more than 6 consecutive 15 minute periods.
- The data capture over the previous 24 hours is less than 90 %.
- The site was not manually checked the previous day.

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  $\text{NO}_x$  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            | 08/07/2015 | 862        |
| Ballymena                   | 28/07/2015 | 849        |
| Belfast                     | 28/07/2015 | 855        |
| Birmingham Roadside         | 20/07/2015 | 869        |
| Birmingham Urban background | 20/07/2015 | 859        |
| Cardiff Centre              | 13/03/2015 | 868        |
| Detling                     | 04/08/2015 | 865        |
| Dunmurry Kilmakee           | 28/07/2015 | 861        |
| Glasgow High Street         | 09/04/2015 | 860        |
| Glasgow Townhead            | 06/07/2015 | 856        |
| Harwell                     | 31/07/2015 | 851        |
| Marylebone Road             | 05/08/2015 | 866        |
| North Kensington            | 05/08/2015 | 867        |
| Strabane                    | 29/07/2015 | 848        |

**Table 3 Site Audit Visits**

##### 3.1.1 Sampler Leak Rate and Calibration of Sample Flow

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

The absolute value of the inlet flow measured during the leak test is 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%. Black Carbon concentrations are not corrected for leak rate, but the leak rate is included in the uncertainty budget.

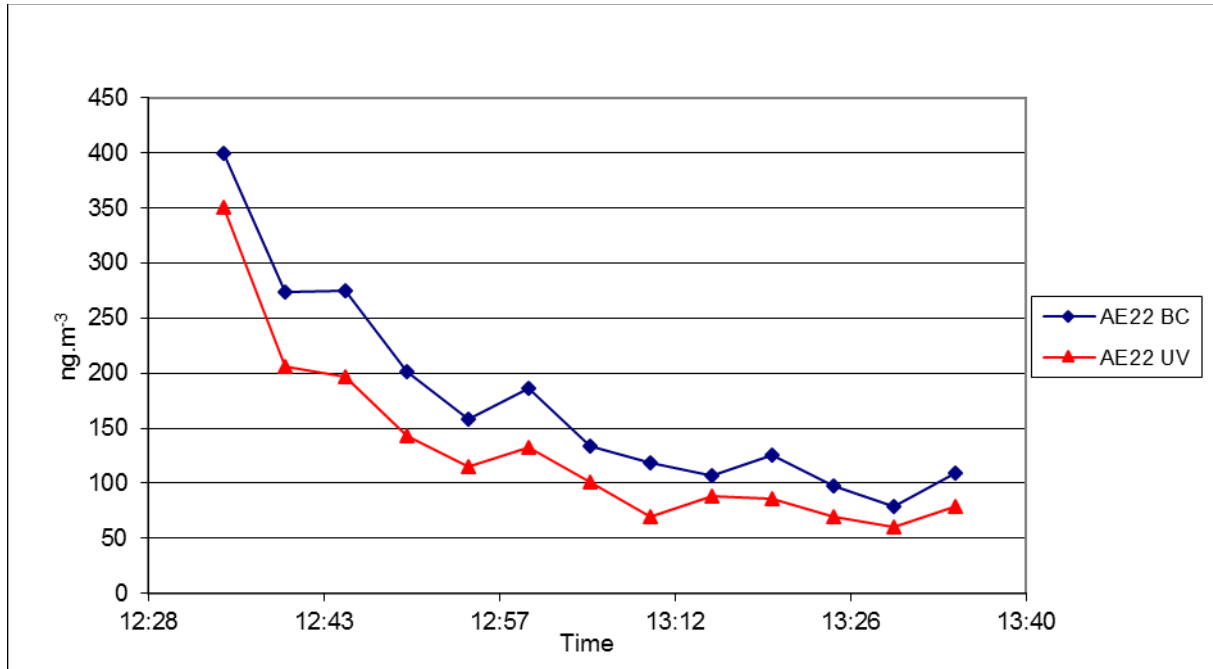
Table 4 gives the measured leak rates and sample flows for each site:

| <b>Site</b>                 | <b>Leak Rate,<br/>%</b> | <b>Indicated Flow,<br/>lpm</b> | <b>Inlet Flow,<br/>lpm</b> |
|-----------------------------|-------------------------|--------------------------------|----------------------------|
| Auchencorth Moss            | 0.0                     | 4.0                            | 3.997                      |
| Ballymena                   | 9.1                     | 4.0                            | 3.800                      |
| Belfast                     | 9.1                     | 3.9                            | 3.973                      |
| Birmingham Roadside         | 9.1                     | 4.0                            | 4.150                      |
| Birmingham Urban Background | 1.4                     | 4.0                            | 4.140                      |
| Cardiff Centre              | 4.8                     | 4.0                            | 4.270                      |
| Detling                     | 0.9                     | 4.0                            | 4.210                      |
| Dunmurry Kilmakee           | 4.8                     | 4.0                            | 4.010                      |
| Glasgow High Street         | 4.8                     | 4.0                            | 4.230                      |
| Glasgow Townhead            | 6.6                     | 4.0                            | 4.107                      |
| Harwell                     | 8.7                     | 4.0                            | 4.250                      |
| Marylebone Road             | 9.7                     | 4.0                            | 4.160                      |
| North Kensington            | 8.6                     | 4.0                            | 4.303                      |
| Strabane                    | 14.3                    | 3.9                            | 3.600                      |

**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 is 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 falls 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 North Kensington site.



**Figure 3 North Kensington 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  $\text{ng.m}^{-3}$ . These concentrations during the zero noise tests are normally at or below the stated detection limit of the instrument,  $0.1 \mu\text{g.m}^{-3}$  ( $100 \text{ng.m}^{-3}$ ). The zero noise is 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, $\text{ng.m}^{-3}$ | UV Channel, $\text{ng.m}^{-3}$ | Annual Average BC, $\text{ng.m}^{-3}$ |
|-----------------------------|------------------------|--------------------------------|---------------------------------------|
| Auchencorth Moss            | 0.06                   | 0.03                           | 0.2                                   |
| Ballymena                   | 0.63                   | 0.24                           | 1.0                                   |
| Belfast                     | 1.04                   | 0.42                           | 1.1                                   |
| Birmingham Roadside         | N/A                    | N/A                            | 1.1                                   |
| Birmingham Urban background | 0.18                   | 0.14                           | 2.1                                   |
| Cardiff Centre              | 0.47                   | 0.76                           | 0.9                                   |
| Detling                     | 0.98                   | 0.45                           | 0.4                                   |
| Dunmurry Kilmakee           | 2.06                   | 0.57                           | 0.8                                   |
| Glasgow High Street         | 0.21                   | 0.26                           | 1.4                                   |
| Glasgow Townhead            | 0.24                   | 0.11                           | 0.9                                   |
| Harwell                     | 0.22                   | 0.76                           | 0.3                                   |
| Marylebone Road             | 0.52                   | 0.32                           | 5.1                                   |
| North Kensington            | 0.20                   | 0.18                           | 1.0                                   |
| Strabane                    | 0.89                   | 0.54                           | 1.2                                   |

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

There was a problem with the data collection during the HEPA filter test at the Birmingham Tyburn

Roadside site audit which resulted in the test being invalid. It should be noted that the UV Channel in Table 5 is not the UV component concentration, but the result taken 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, and are continually being reviewed and improved for the network. The values obtained during the audits are considered acceptable.

## **4.0 MEASUREMENT UNCERTAINTY**

### **4.1 SAMPLE VOLUME**

From measurements at the site audit the sample volume can be determined with an uncertainty of  $\pm 9.9\%$ , expressed with a level of confidence of 95%. Included in this uncertainty are contributions from flow rate accuracy, repeatability, drift and leaks.

The leak rate is not used to correct the results, but is included as an uncertainty if the sampler passes the leak test at audit, using the manufacturer's tolerance for leak rate of 20%. For the uncertainty calculation, the average value of leak rate determined in the 2015 audits was used. As leak rate is considered to be a rectangular distribution, its contribution to the standard uncertainty in sample volume is 3.8%.

### **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 is a standard deviation of approximately  $\pm 0.14 \mu\text{g.m}^{-3}$  for hourly means, compared to the network mean of  $1.60 \mu\text{g.m}^{-3}$ . Converting this into a standard uncertainty represents a contribution of 10.7%,

### **4.3 CORRECTION FOR SPOT DARKENING**

The Virkkula<sup>2</sup> model was used to correct the measured concentrations to account for the nonlinearity of Aethalometer Black Carbon and UV component measurements with attenuation. This effect and its correction introduce an uncertainty into the measurements. At most sites the correction can be seen to work well on the 15-minute data, in that there is minimal discontinuity when the spot location changes, and the associated uncertainty is considered to be small compared to other components. At sites where the concentration is changing quickly, such as Marylebone Road, this uncertainty in the 15-minute data becomes significant although this is decreased when hourly mean concentrations are calculated.

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

The uncertainty due to the spot darkening cannot 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 are combined, the overall measurement uncertainty for hourly Black Carbon concentrations is 23.7%, 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  | 23.7% |
| Monthly | 10.0% |
| Yearly  | 9.9%  |

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 and is calculated from the results of the 2015 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 2015 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 have been corrected for spot darkening using the Virkkula method<sup>2</sup>.

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

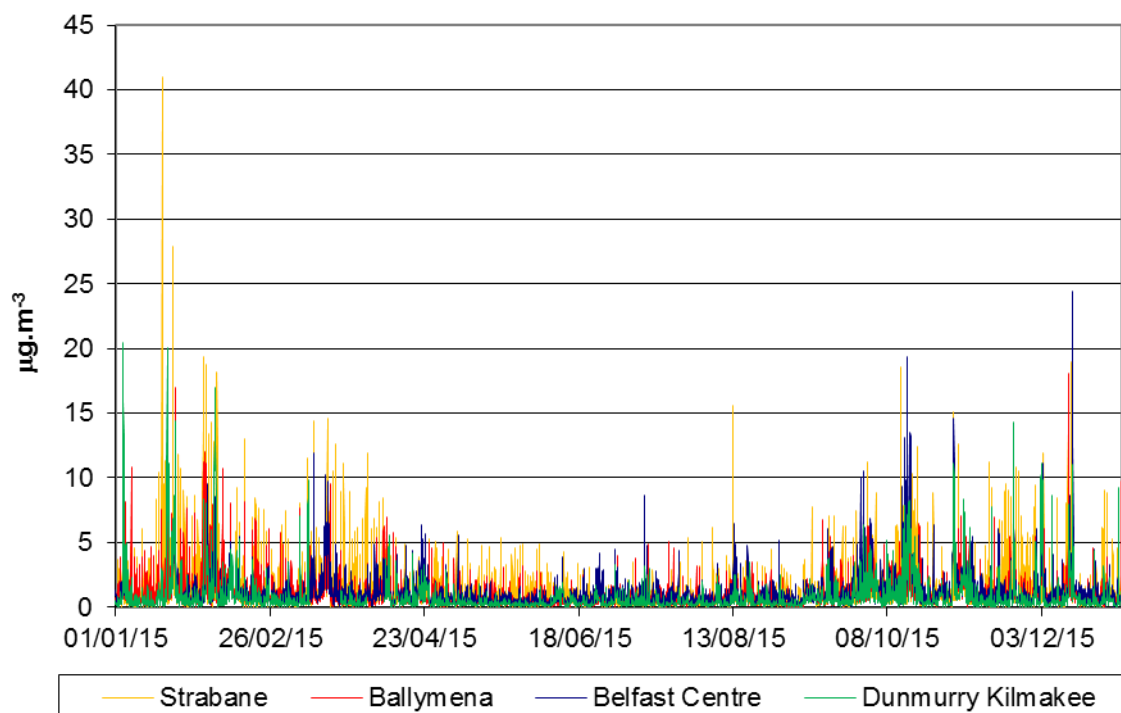
<http://uk-air.defra.gov.uk/>

### 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 by the UK Black Carbon Network for 2015. 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 45  $\mu\text{g.m}^{-3}$  to enable easy comparison between charts, except for Figure 8 which shows concentrations measured at rural locations.



**Figure 4** Black Carbon concentrations during 2015 in Northern Ireland

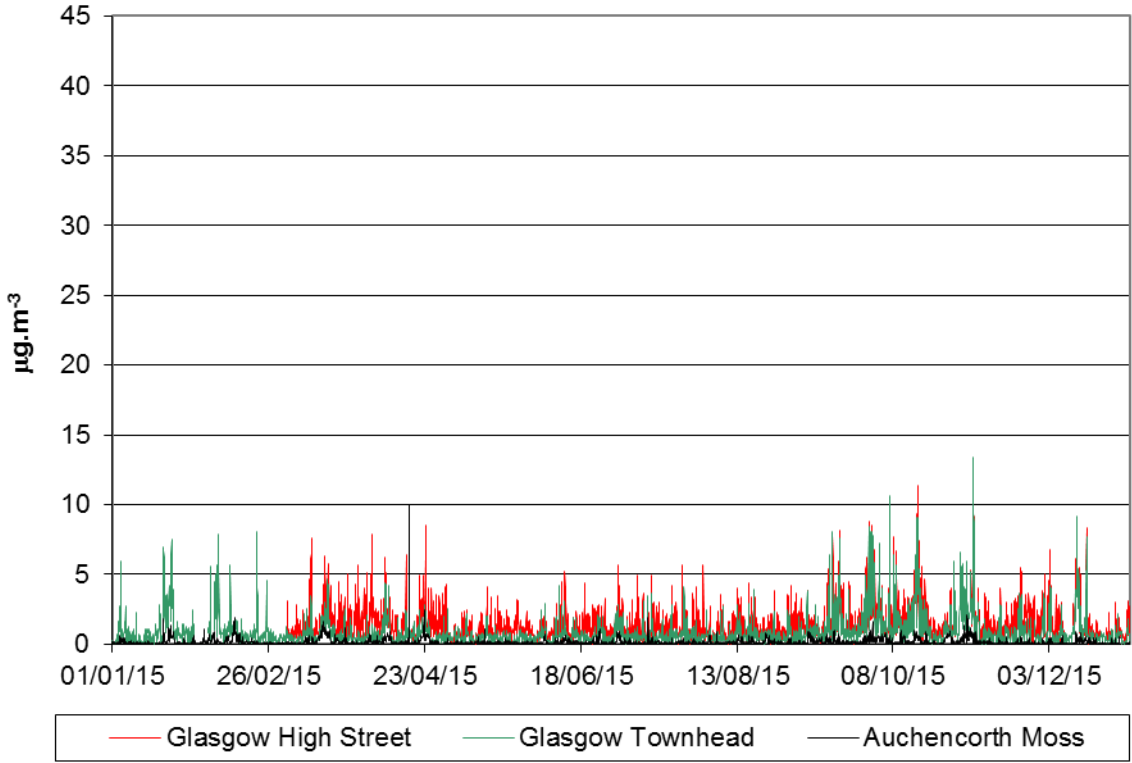


Figure 5 Black Carbon concentrations during 2015 in Scotland

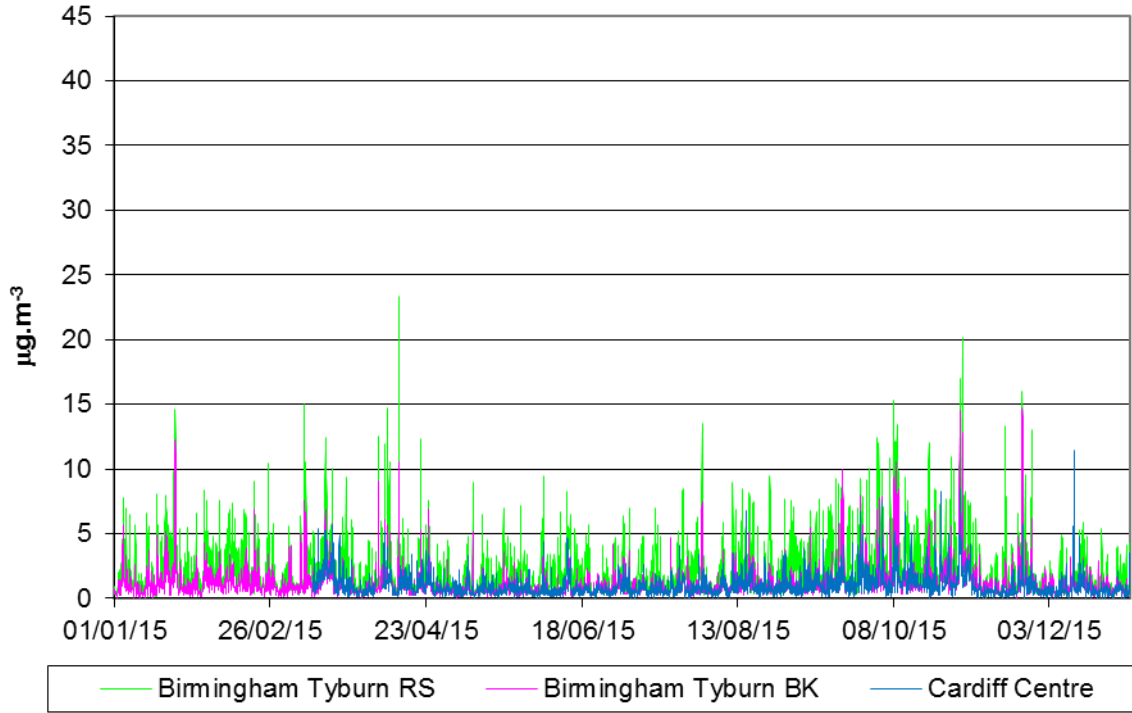
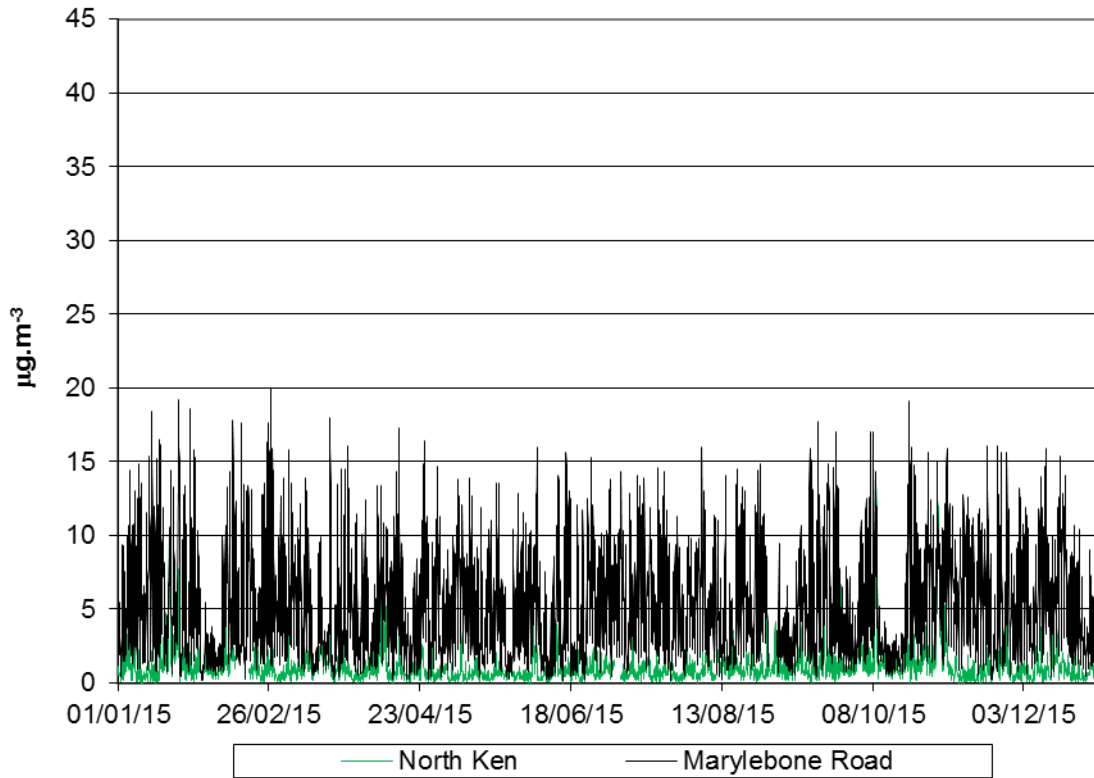
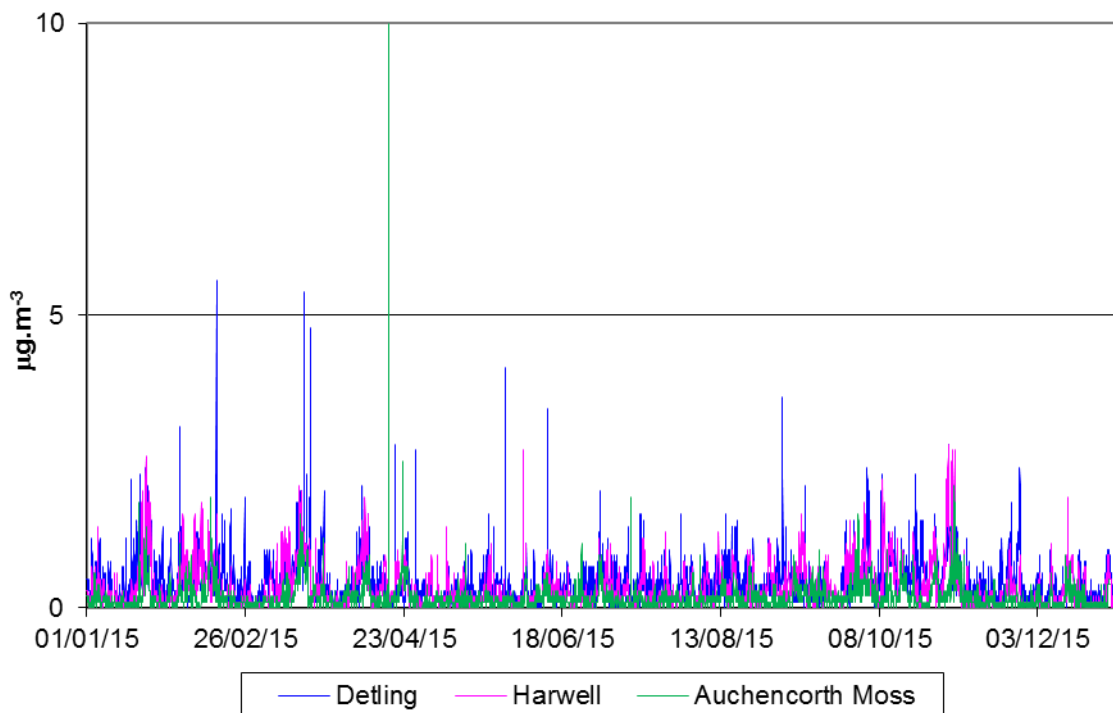


Figure 6 Black Carbon concentrations during 2015 in Wales and the Midlands





**Figure 7 Black Carbon concentrations during 2015 in London**



**Figure 8 Black Carbon concentrations during 2015 at Rural Locations**

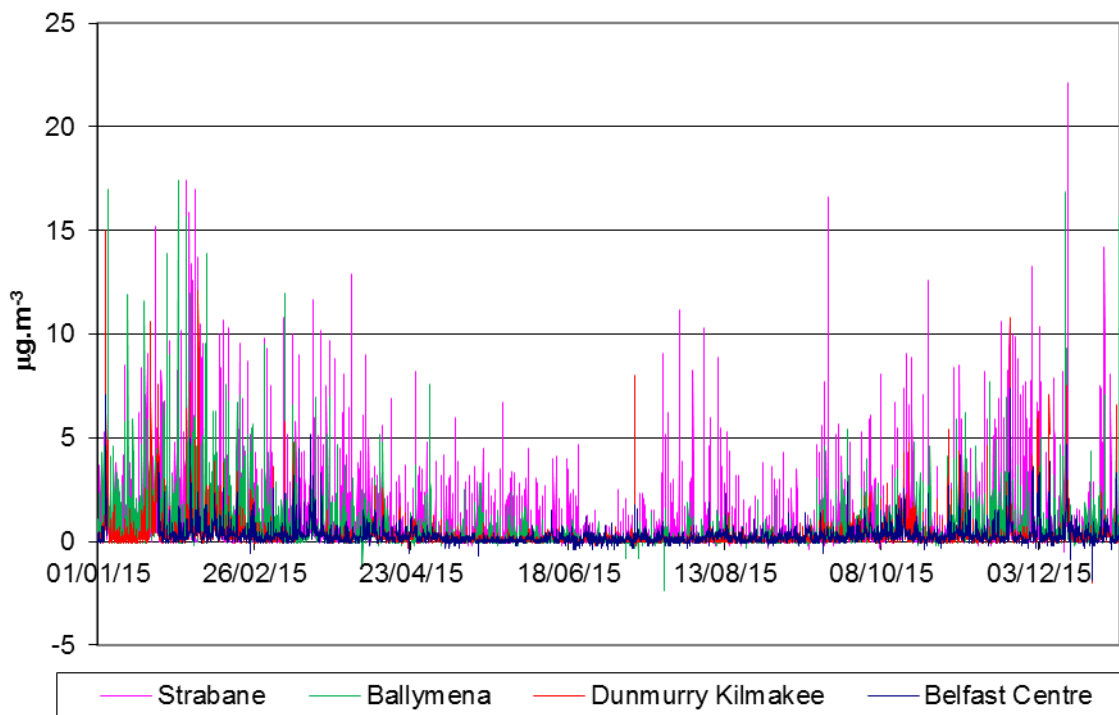
Northern Irish sites measured increased concentrations during the cold months in January, February, March, October, November and December.

Most UK sites measured elevated black carbon concentrations between 16<sup>th</sup> to 24<sup>th</sup> January due to cold still weather leading to poor dispersion of local sources followed by long range transport from 22<sup>nd</sup> January onwards.

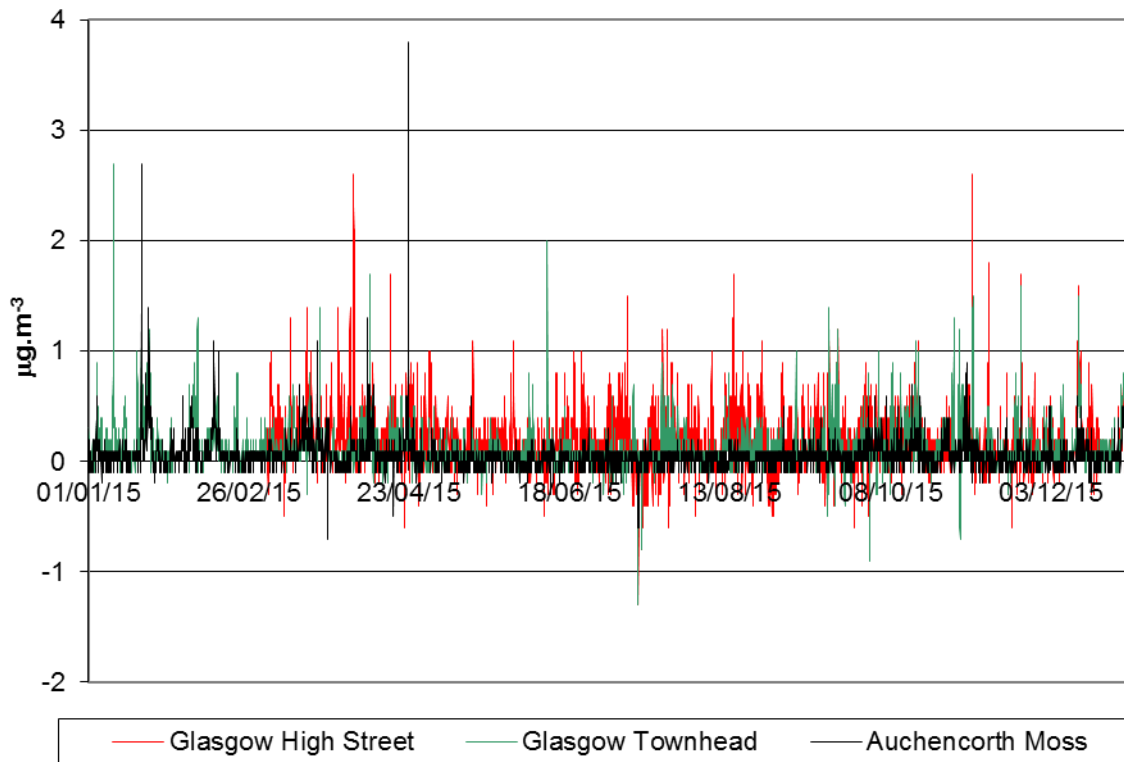
Elevated Black Carbon concentrations were also measured between 31<sup>st</sup> October and 2<sup>nd</sup> November at many sites across the UK, indicating emissions associated with early bonfire night celebrations. November 5<sup>th</sup> fell on a Thursday in 2015 so many celebrations were held the weekend before. Especially high concentrations were recorded at Birmingham Tyburn Background and Roadside sites with concentrations reaching over 20  $\mu\text{g.m}^{-3}$ . Concentrations of Black Carbon were not as high as in previous years where concentrations in excess of 40  $\mu\text{g.m}^{-3}$  have been measured.

### 5.1.2 UV component

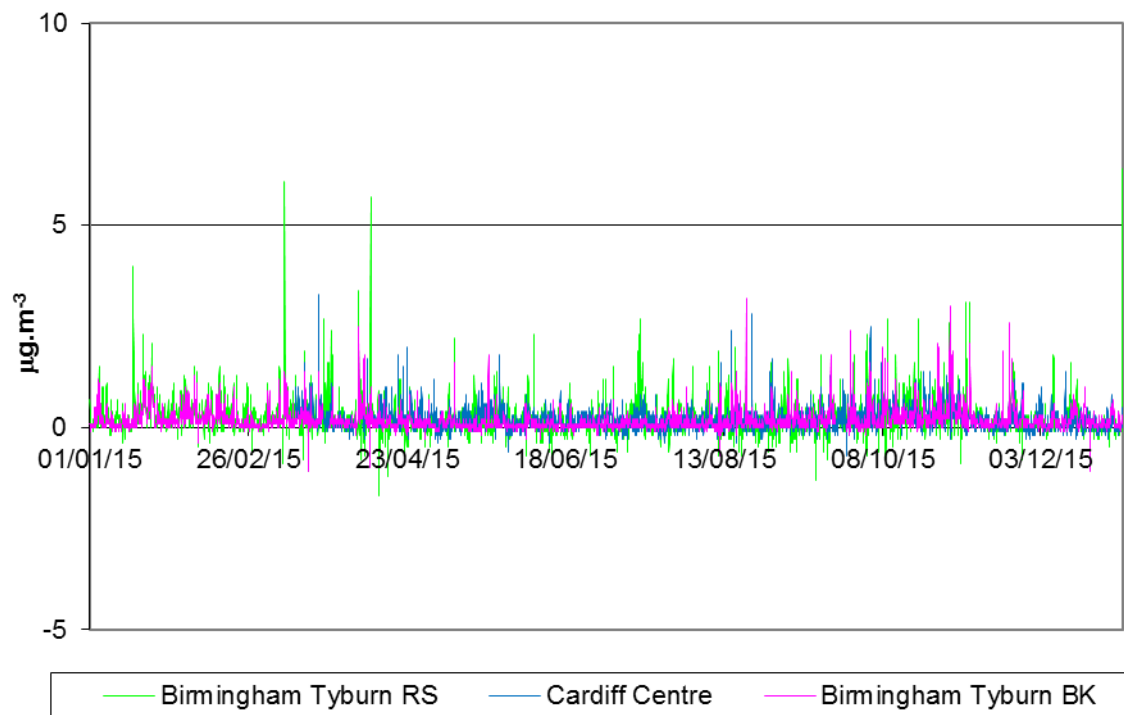
The following charts show the UV component concentrations measured by the UK Black Carbon Network for 2015. The time resolution of the measurements is hourly. Data has been split into regions of the UK for presentation purposes. The y-axis on the UV component time series graphs have not been fixed to 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 2015 in Northern Ireland**



**Figure 10 UV component concentrations during 2015 in Scotland**



**Figure 11 UV component concentrations during 2015 in Wales and the Midlands**

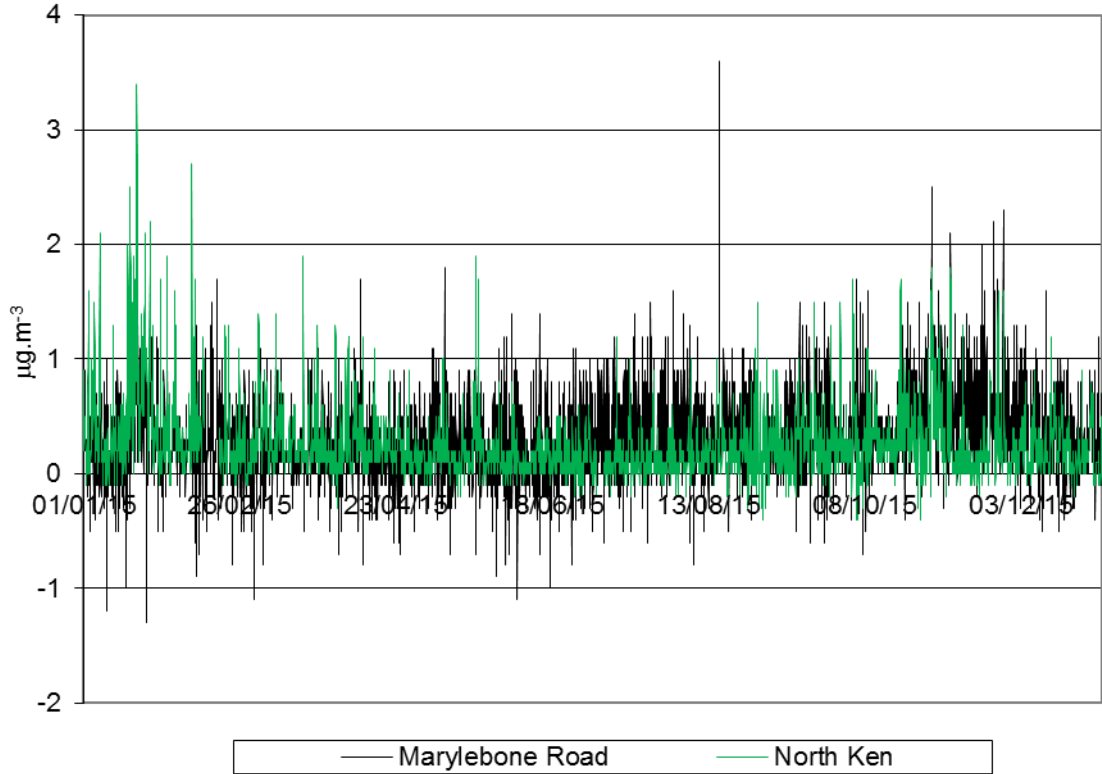


Figure 12 UV component concentrations during 2015 in London

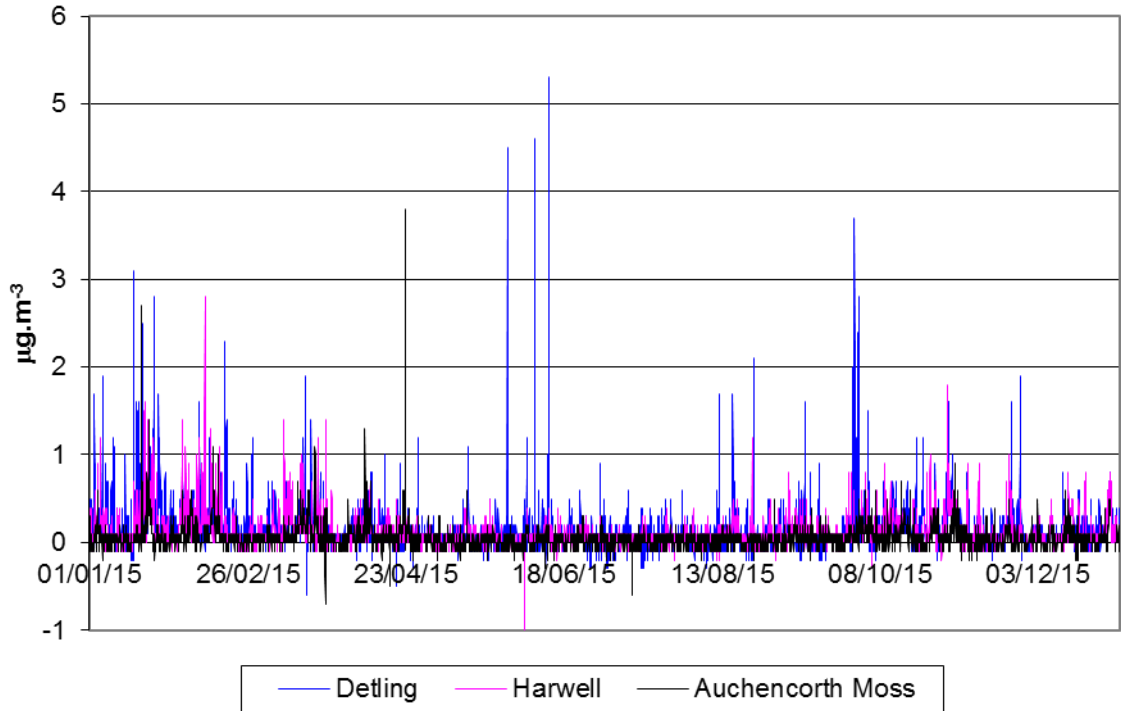


Figure 13 UV component concentrations during 2015 at Rural Locations

The Northern Irish sites measured increased UV component concentrations during the cold periods in January, February, March, October November and December. Evidence from the UV component concentrations for 2015 suggests that the heating season runs from January to mid-June and mid-

September to the end of the year in Strabane. However there are also elevated UV component concentrations during July and August in Strabane suggesting there are still significant emissions of UV absorbing particulate mass (either solid fuel or wood burning). In Ballymena the heating season finishes earlier in mid-April but starts again at the same time as Strabane. Concentrations across 2015 were lower than those measured in 2014, but with a slightly longer heating season.

Elevated UV component concentrations were also measured between 31<sup>st</sup> October and 2<sup>nd</sup> November at many sites across the UK, indicating emissions associated with early bonfire night celebrations. Hourly concentrations reached a maximum of 3  $\mu\text{g}\cdot\text{m}^{-3}$ . Concentrations of UV component were not as high as in previous years where concentrations in excess of 15  $\mu\text{g}\cdot\text{m}^{-3}$  have been measured.

UV component concentrations were also elevated during 16<sup>th</sup> to 24<sup>th</sup> January due to cold still weather leading to poor dispersion of local sources, followed by long range transport from 22<sup>nd</sup> January onwards.

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 370nm 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 boil off the tape and reduce the enhanced UV adsorption. If equilibrium between organic species deposit and boil off is not reached due to cleaner air being sampled, the amount of organic enhancement will drop and will lead to negative UV component concentrations.

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.

Equally, the UVPM calculation assumes an Ångström Coefficient of 1.0 for the wavelength dependant absorption of freshly emitted Black Carbon from traffic sources. Fuller *et al*<sup>3</sup> estimate this to be close to 0.96, which would 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 is also found at Birmingham Tyburn Roadside, which still has high traffic flows but less diesel based exhaust emissions than Marylebone Road. This effect is not prevalent at Birmingham Tyburn Background which is approximately 60m away from the roadside site.

## 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

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<sup>3</sup> Contribution of wood burning to PM10 in London, Fuller *et al*, ATMOSPHERIC ENVIRONMENT 87 (2014), Volume: 87, Pages: 87-94.

### 5.2.1 Black Carbon

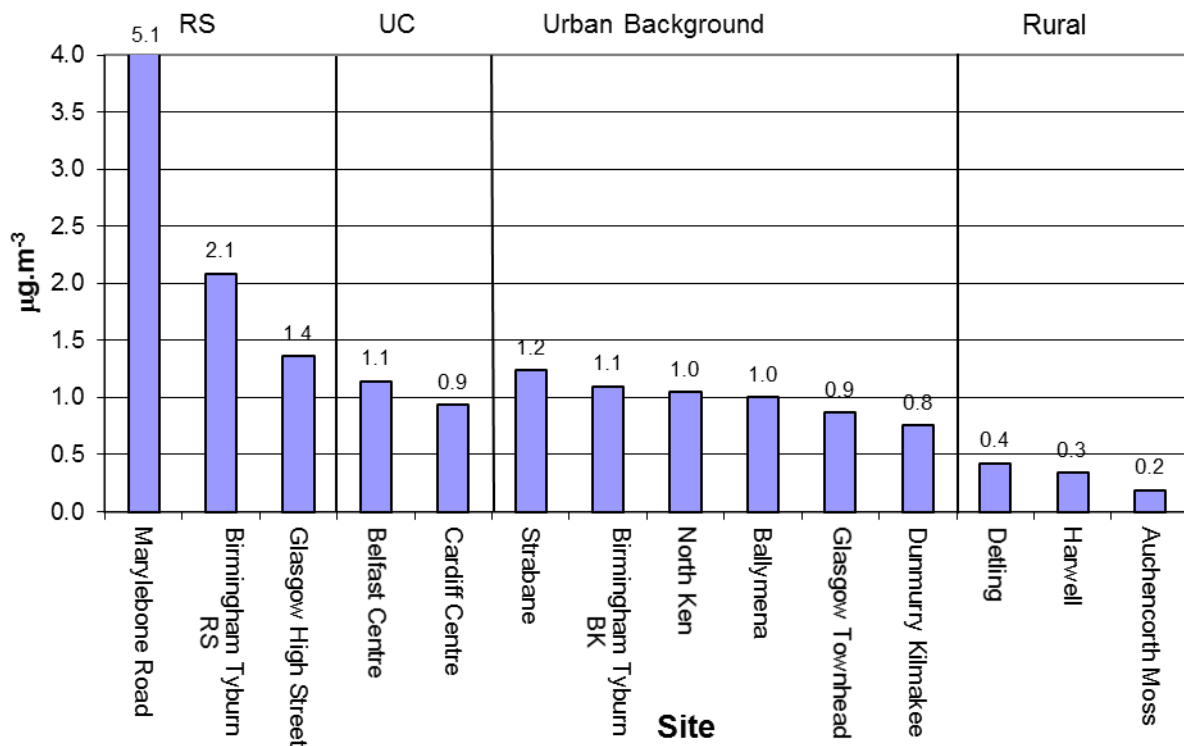
Table 6 gives the annual mean for each site for 2015.

| Site                 | Mean concentration<br>$\mu\text{g.m}^{-3}$ |
|----------------------|--|
| Auchencorth Moss     | 0.2  |
| Ballymena            | 1.0  |
| Belfast Centre       | 1.1  |
| Birmingham Tyburn BK | 1.1  |
| Birmingham Tyburn RS | 2.1  |
| Cardiff Centre       | 0.9  |
| Detling              | 0.4  |
| Dunmurry Kilmakee    | 0.8  |
| Glasgow High Street  | 1.4  |
| Glasgow Townhead     | 0.9  |
| Harwell              | 0.3  |
| Marylebone Road      | 5.1  |
| North Kensington     | 1.0  |
| Strabane             | 1.2  |

Note: Cardiff Centre and Glasgow High Street are not full calendar years.

**Table 6 Annual Mean Black Carbon Concentrations for 2015**

The annual mean concentrations are presented as a bar graph (Figure 14) to aid the comparison of sites:



**Figure 14 Annual Mean Black Carbon Concentrations for 2015**

Black Carbon concentration increments between rural, urban background and roadside sites for London, Birmingham and Scotland 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      | 0.7                             | 4.1      |
| Birmingham  | 0.8                             | 1.0      |
| Glasgow     | 0.7                             | 0.5      |

**Table 7 Increments in Black Carbon concentrations between rural, background and roadside sites in 2015**

It can be seen that the urban increment for all three areas is similar, while the roadside increment for London is much larger than that for Birmingham and Glasgow. This is due to the much larger traffic flow and different vehicle profile of the Marylebone Road site compared to the Birmingham Tyburn and Glasgow High Street roadside sites. Highways Agency traffic count data for 2015 for the three roads passing the monitoring stations are given in Table 8.

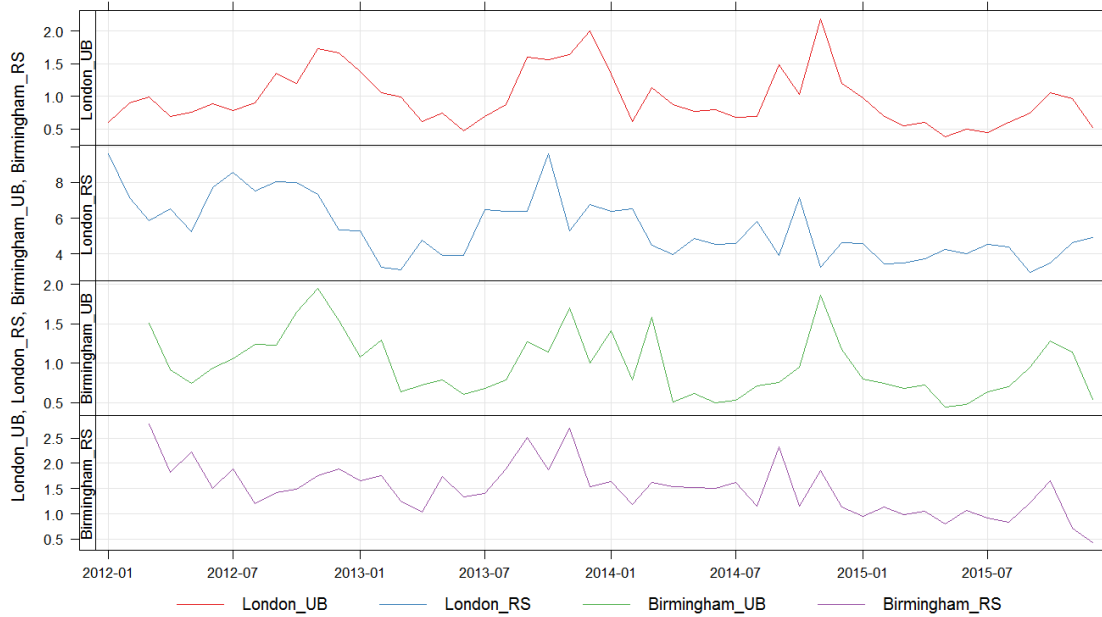
| Road (Count Point ID)       | Motor cycles | Cars Taxis | Buses Coaches | All HGVs | All Motor Vehicles |
|-----------------------------|--------------|------------|---------------|----------|--------------------|
| Marylebone Road (27236)     | 4,362        | 53,177     | 2,607         | 3,082    | 73,134             |
| Tyburn Road (56399)         | 147          | 24,317     | 249           | 1,647    | 31,693             |
| Glasgow High Street (10821) | 42           | 12,486     | 211           | 398      | 15,312             |
| Ratio London to Birmingham  | 30           | 2.2        | 10.5          | 1.9      | 2.3                |
| Ratio London to Glasgow     | 104          | 4.3        | 12.4          | 7.7      | 4.8                |
| Ratio Birmingham to Glasgow | 3.5          | 1.9        | 1.2           | 4.1      | 2.1                |

**Table 8 2016 Average daily traffic count data for Marylebone, Tyburn Road and Glasgow High Street Roadside sites**

The Marylebone Road roadside increment in Black Carbon concentration in 2015 was a factor of 4.1 higher than the Tyburn Road increment, somewhat higher than the ratio of numbers of cars / taxis and HGVs, between the sites at 2.2 and 1.9 respectively. However there were 10.5 times more buses and coaches and 30 times more motor cycles passing the Marylebone Road site compared with the Birmingham site, which would indicate that these are probably the predominant source of Black Carbon emissions at Marylebone Road. Changes in emissions from London buses and taxis are discussed in section 5.5.1.

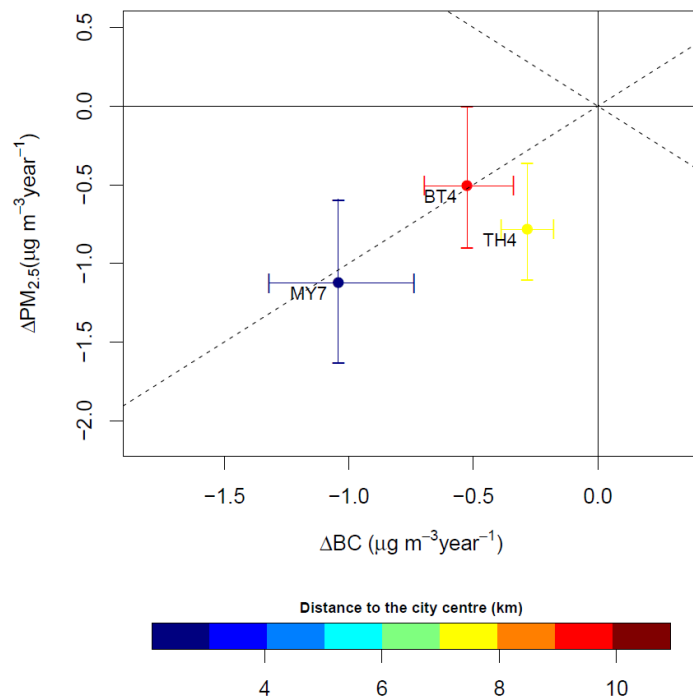
The Birmingham roadside increment is twice that of Glasgow, which is similar to the ratio in all motor vehicles indicating a similar emission profile for each site.

Figure 15 shows how the urban and roadside increments in London and Birmingham have changed over the period 2012 to 2015. The average urban background at both locations is roughly stable with increases during the heating season indicating the contribution from domestic heating. The roadside increment for London has clearly dropped over the whole period, while the Birmingham roadside increment was roughly stable for 2012 to 2014 and reduced in 2015.



**Figure 15 Urban and roadside increments for London and Birmingham for the period 2012 to 2015**

The change of London roadside increment in Black Carbon is also correlated to a reduction of the London roadside increment in PM<sub>2.5</sub> concentration<sup>4</sup> as shown in Figure 16 where a 1:1 correspondence is valid.



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

**Figure 16 Relationship between London roadside Black Carbon and PM<sub>2.5</sub> increments**

<sup>4</sup> Font, A.; and Fuller, G. W.: Did policies to abate atmospheric emissions from traffic have a positive effect in London? Using a large population of monitoring sites to investigate recent changes in air pollution. Submitted to Environmental Pollution May16.



This indicates similar emission sources (road transport) for Black Carbon and PM<sub>2.5</sub> roadside increments, as expected.

### 5.2.2 UV component

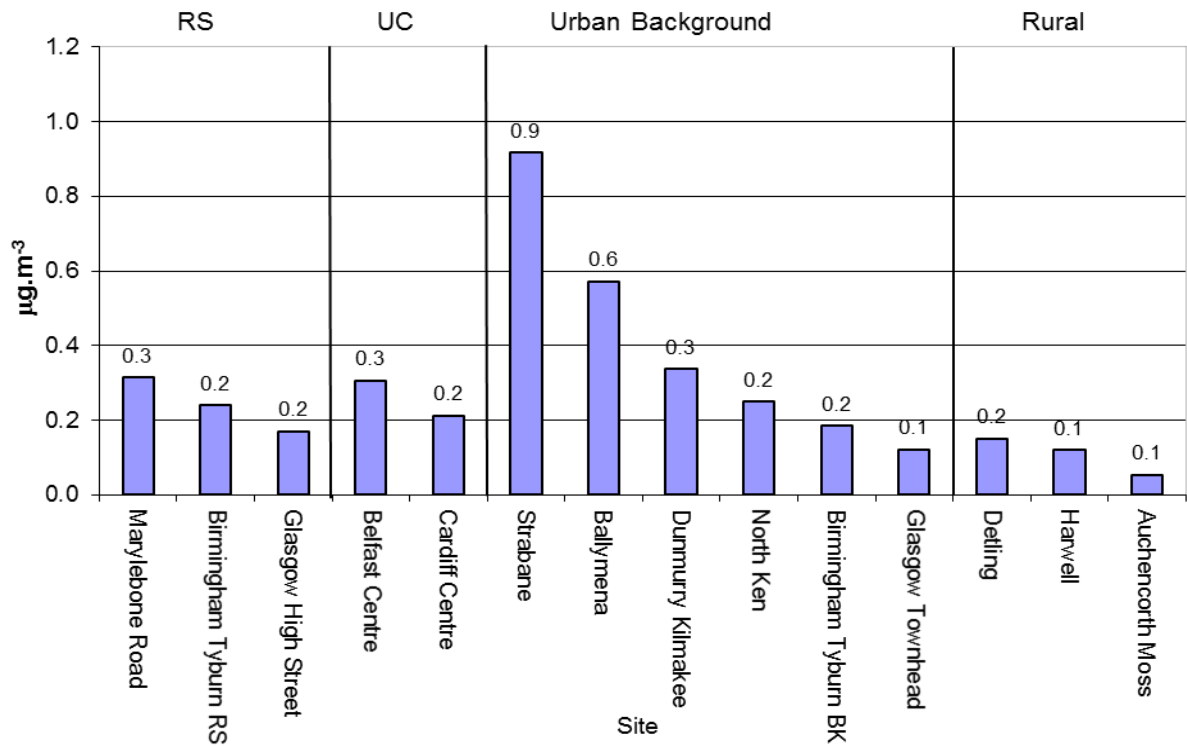
Table 9 gives the annual average for each site for 2015.

| Site                 | Mean concentration<br>$\mu\text{g}\cdot\text{m}^{-3}$ |
|----------------------|---|
| Auchencorth Moss     | 0.1   |
| Ballymena            | 0.6   |
| Belfast Centre       | 0.3   |
| Birmingham Tyburn BK | 0.2   |
| Birmingham Tyburn RS | 0.2   |
| Cardiff Centre       | 0.2   |
| Detling              | 0.2   |
| Dunmurry Kilmakee    | 0.3   |
| Glasgow High Street  | 0.2   |
| Glasgow Townhead     | 0.1   |
| Harwell              | 0.1   |
| Marylebone Road      | 0.3   |
| North Kensington     | 0.2   |
| Strabane             | 0.9   |

Note: Cardiff Centre and Glasgow High Street are not full calendar years.

#### Table 9 Annual Mean UV component Concentrations for 2015

The annual mean concentrations are presented as a bar graph (Figure 17) to aid the comparison of sites:



**Figure 17 Annual Mean UV component Concentrations for 2015**

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

| Conurbation | Increment, $\mu\text{g}\cdot\text{m}^{-3}$ |          |
|-------------|--|----------|
|             | Urban                                      | Roadside |
| London      | 0.1  | 0.1      |
| Birmingham  | 0.1  | 0.0      |
| Scotland    | 0.1  | 0.1      |

**Table 10 Increments in UV component concentrations between rural, background and roadside sites in 2015**

There is no significant difference in increments between 2014 and 2015.

It can be seen that the urban and roadside increments at all sites are similar, as domestic emissions in the three areas are of a similar magnitude due to similar fuel types and that road traffic is 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. The results are shown in Table 11.

| Site      | Increment compared to Belfast,<br>$\mu\text{g.m}^{-3}$ | Increment compared to Belfast,<br>% |
|-----------|--|-------------------------------------|
| Dunmurry  | 0.0  | 10                                  |
| Ballymena | 0.3  | 86                                  |
| Strabane  | 0.6  | 200                                 |

**Table 11 Increment in UV component concentration in Northern Ireland**

The increments at Dunmurry, Ballymena and Strabane are not surprising as domestic heating in Belfast has predominantly been gas fired since 2000, while there is a history of solid fuel usage for secondary heating in Dunmurry, and a significant usage of non-smokeless fuel in Strabane. Ballymena is supplied by natural gas, but the benzo[a]pyrene (BaP) concentrations measured by the PAH Network are higher than might be expected. A similar increase in the UV component is also found here. Correlations between BaP and the UV component can be seen in Section 5.4.2. It can therefore be concluded that there is a significant emission source of both these components in the Ballymena area, probably from the use of non-smokeless fuel. The monitoring site is not within Ballymena's smoke control zone but is within its Air Quality Management Area for PM<sub>10</sub>.

All three increments have decreased compared to 2014. Dunmurry has decreased by from 0.1  $\mu\text{g.m}^{-3}$  to 0.0  $\mu\text{g.m}^{-3}$ , Ballymena from 0.5  $\mu\text{g.m}^{-3}$  to 0.3  $\mu\text{g.m}^{-3}$  and Strabane from 0.8  $\mu\text{g.m}^{-3}$  to 0.6  $\mu\text{g.m}^{-3}$  between 2014 and 2015. This may be due to the warmer weather in the last quarter of 2015 or reduced use of solid fuel for domestic heating due to reduced fuel poverty.

### 5.2.3 Data Capture

Table 12 gives the data capture for each site for 2015. Due to the Network changes during 2015 the time coverage for the complete calendar year for each site has also been given.

| Site                 | Data Capture<br>% | Time Coverage<br>% |
|----------------------|-------------------|--------------------|
| Auchencorth Moss     | 99                | 99                 |
| Ballymena            | 100               | 100                |
| Belfast Centre       | 95                | 95                 |
| Birmingham Tyburn BK | 98                | 98                 |
| Birmingham Tyburn RS | 98                | 98                 |
| Cardiff Centre       | 99                | 80                 |
| Detling              | 98                | 98                 |
| Dunmurry Kilmakee    | 95                | 95                 |
| Glasgow High Street  | 96                | 80                 |
| Glasgow Townhead     | 98                | 98                 |
| Harwell              | 99                | 99                 |
| Marylebone Road      | 99                | 99                 |
| North Ken            | 97                | 97                 |
| Strabane             | 98                | 98                 |

**Table 12 Data capture rates of the Aethalometers for 2015**

The average data capture for the Network is 98% with no sites obtaining a data capture below 95%. Cardiff Centre and Glasgow High Street have time coverages for the year of 80% as both sites were installed in early March 2015.

### 5.3 TEMPORAL VARIATIONS

The following section presents analysis of the 2015 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 are made using the data from 2009 – 2015, to avoid bias introduced by single year measurements, y-axes vary by site.

All results have been grouped by site classification. The site order for the Roadside and Urban Centre 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 2015 data are presented in Figures 18 to 21.

The 2009-2015 data is presented in Figures 22 to 25.

#### 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 22-25 is for display purposes only, to allow the uncertainty in the mean value to be seen more clearly.

#### Acknowledgement

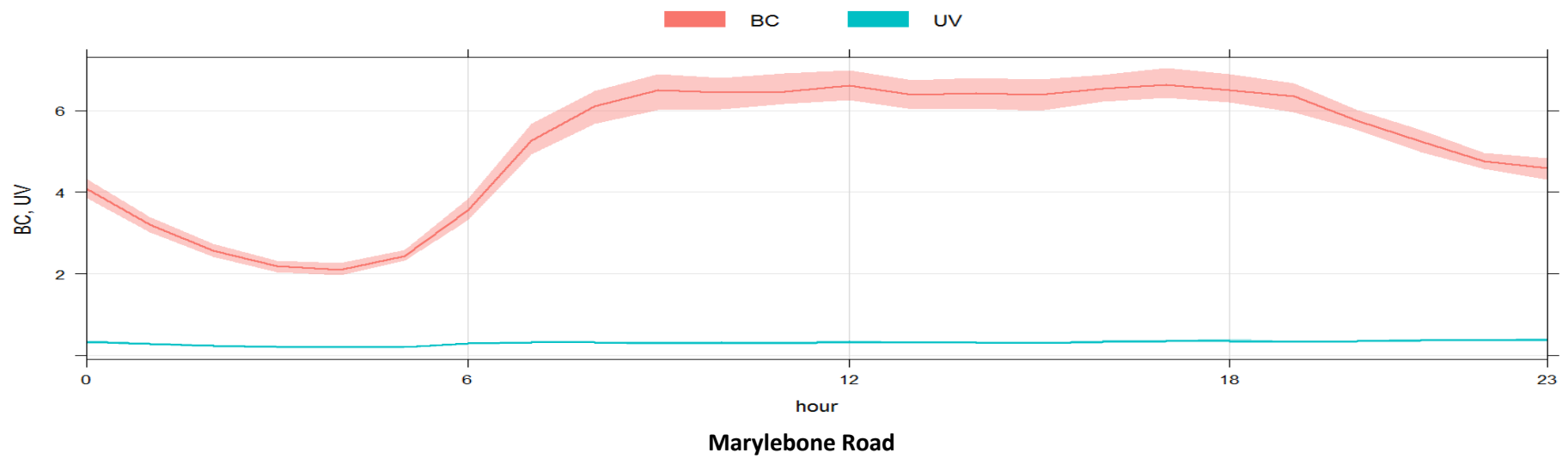
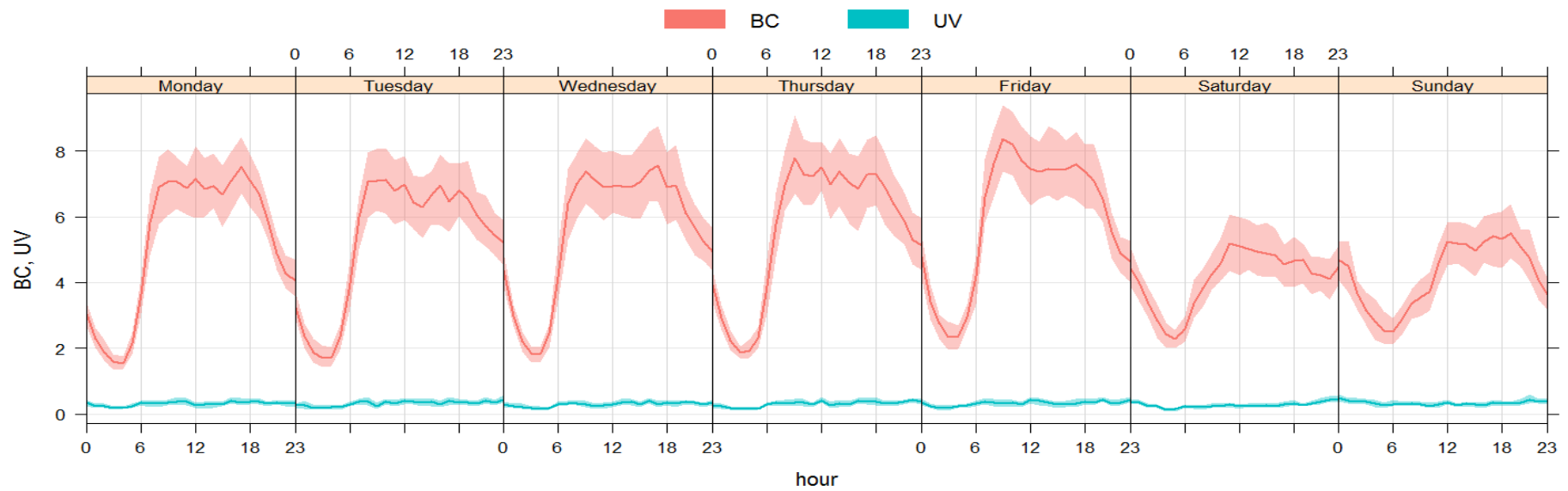
Figures 18 to 25 are generated using the Open-Air Tools run on the R software platform<sup>5,6</sup>.

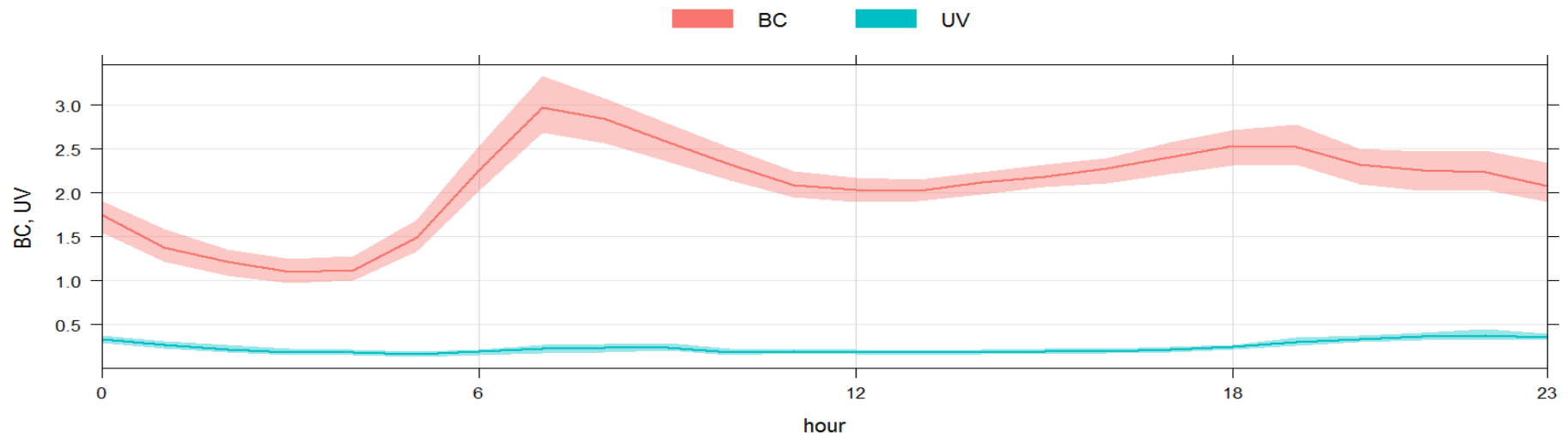
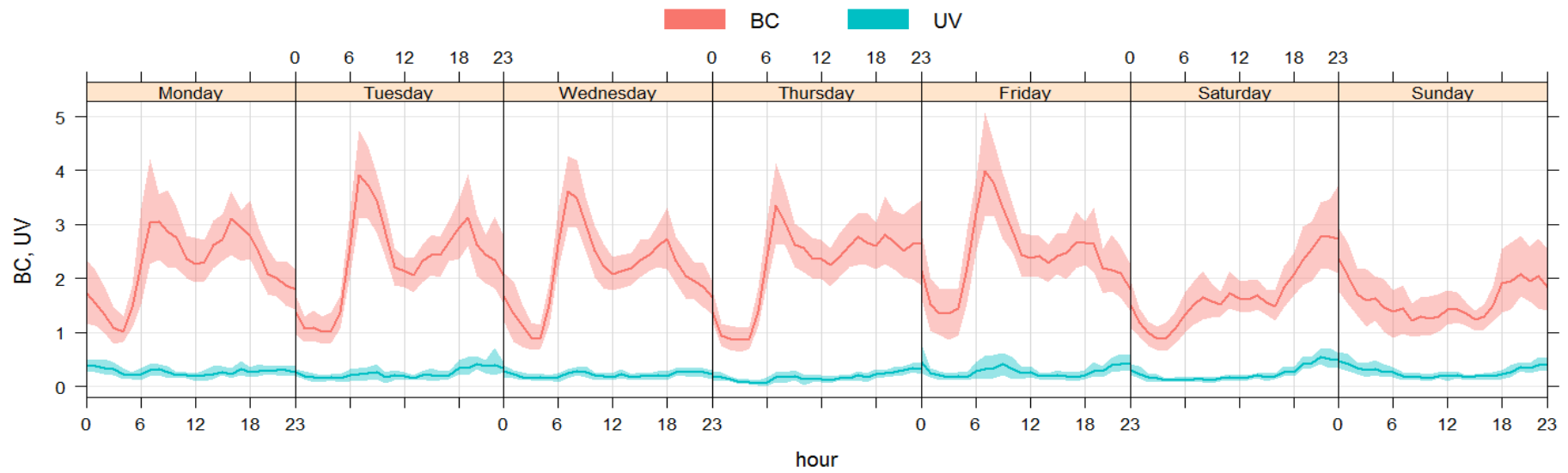
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5 DC Carslaw and K Ropkins, (2012) OpenAir --- an R package for air quality data analysis, Environmental Modelling & Software. Volume 27-28, 52-61.

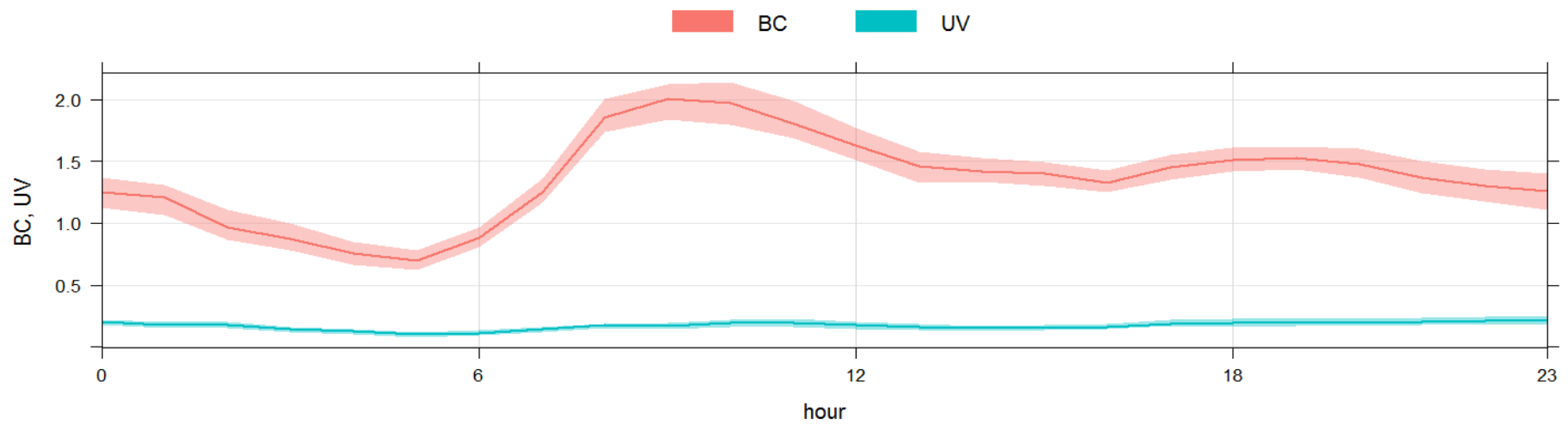
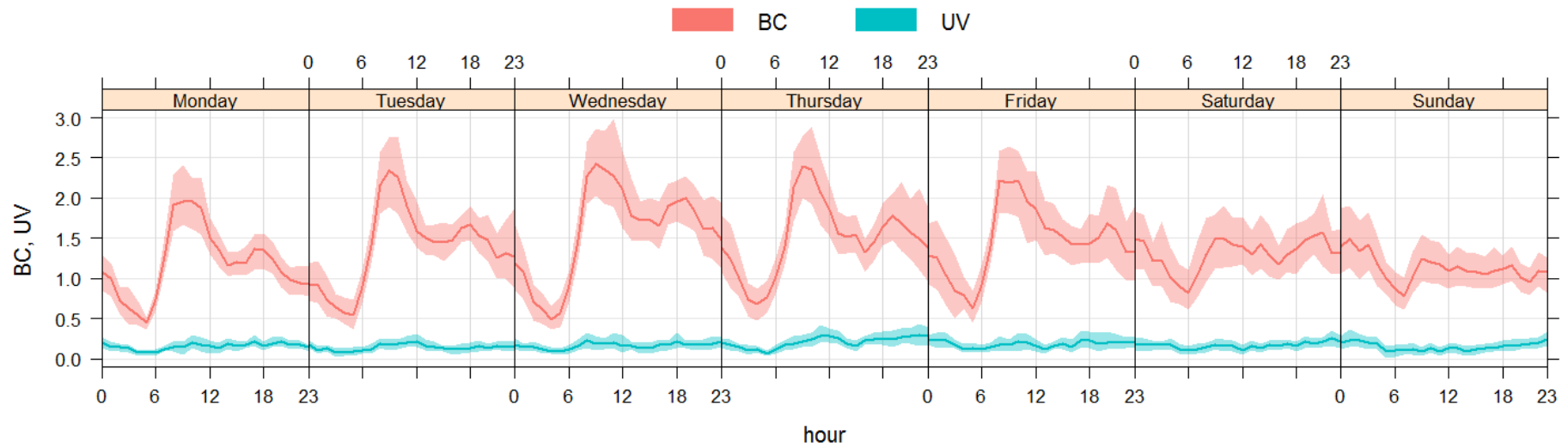
6 DC Carslaw and K Ropkins (2015). OpenAir: Open-source tools for the analysis of air pollution data, R package version 1.1-5

### 2015 Data





Birmingham Tyburn



Glasgow High Street

Figure 18 Roadside Sites

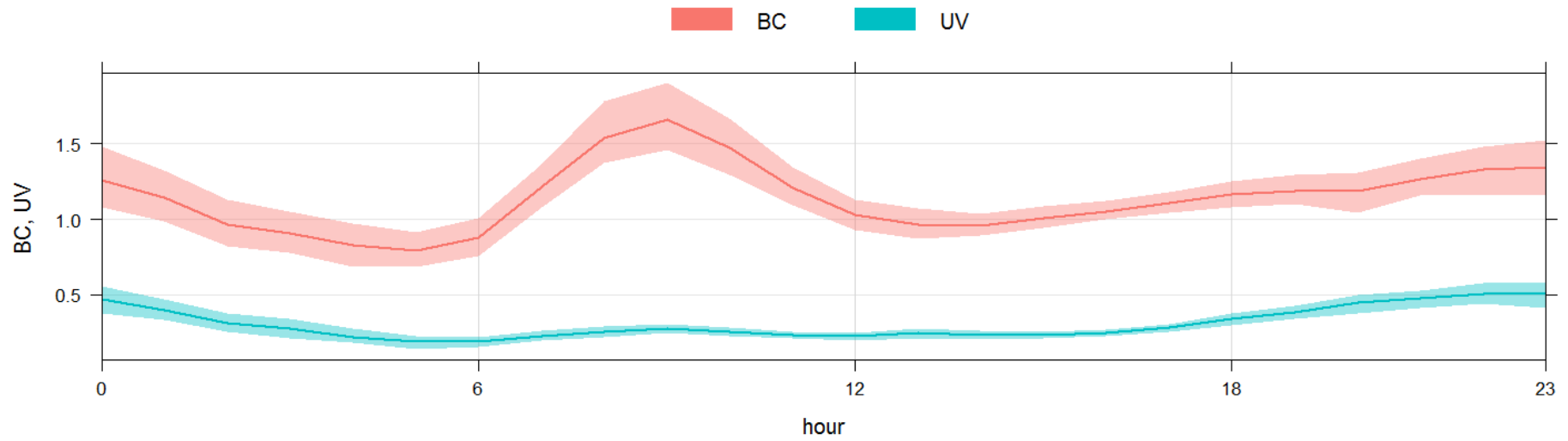
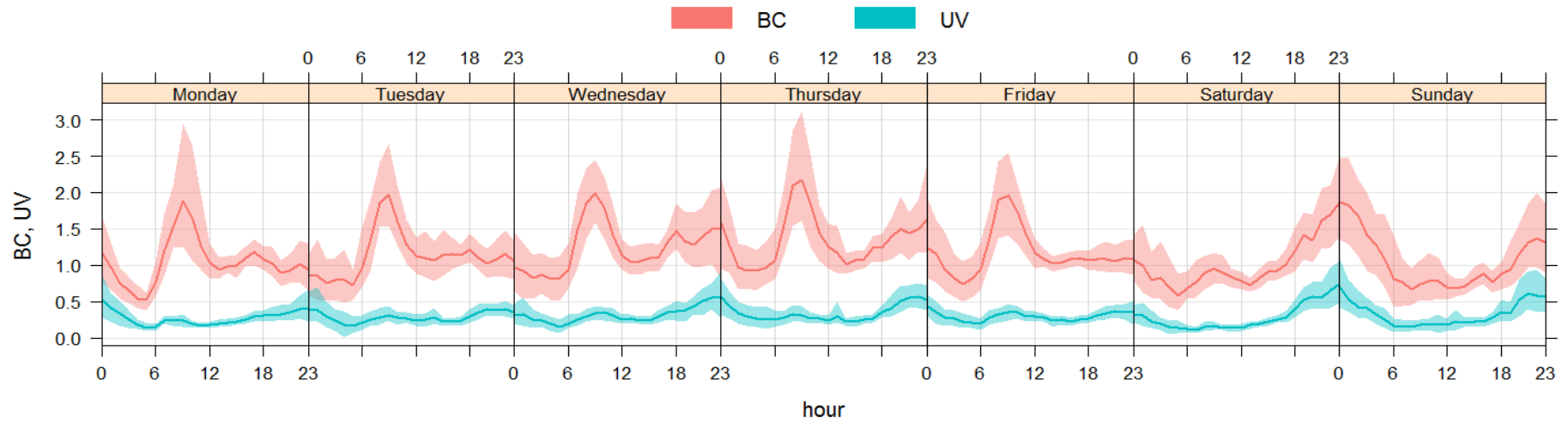
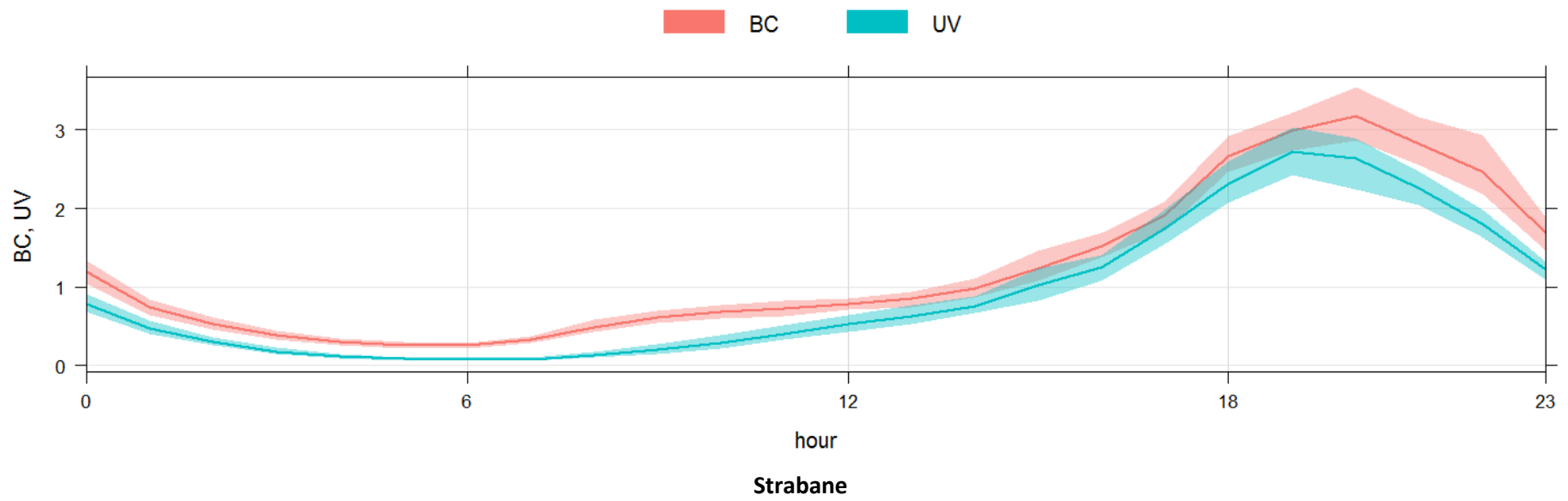
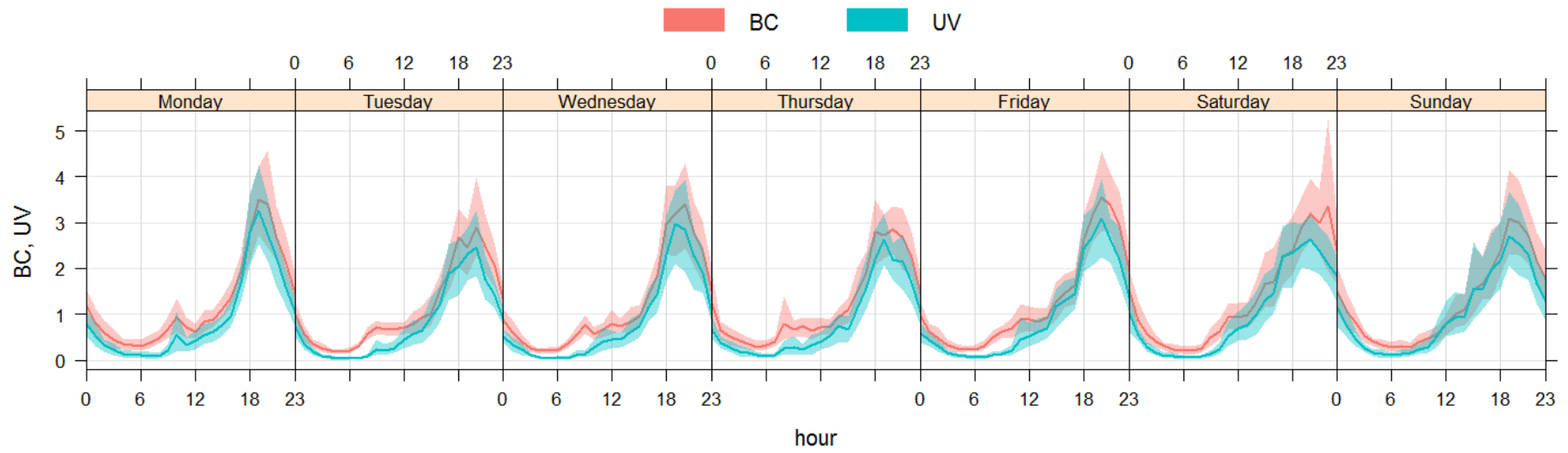
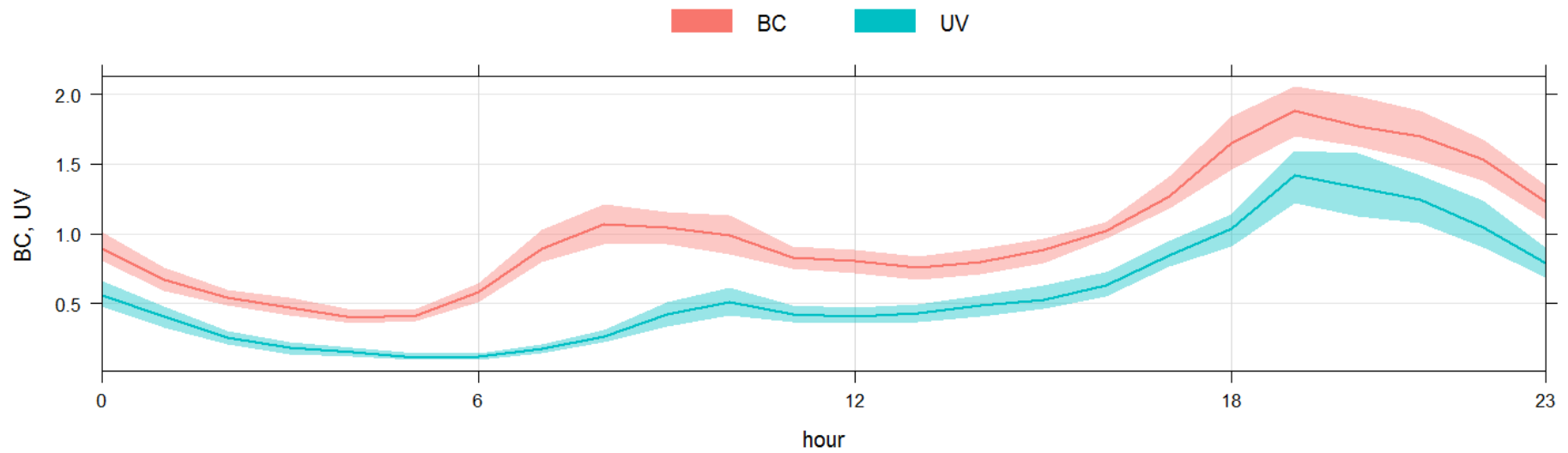
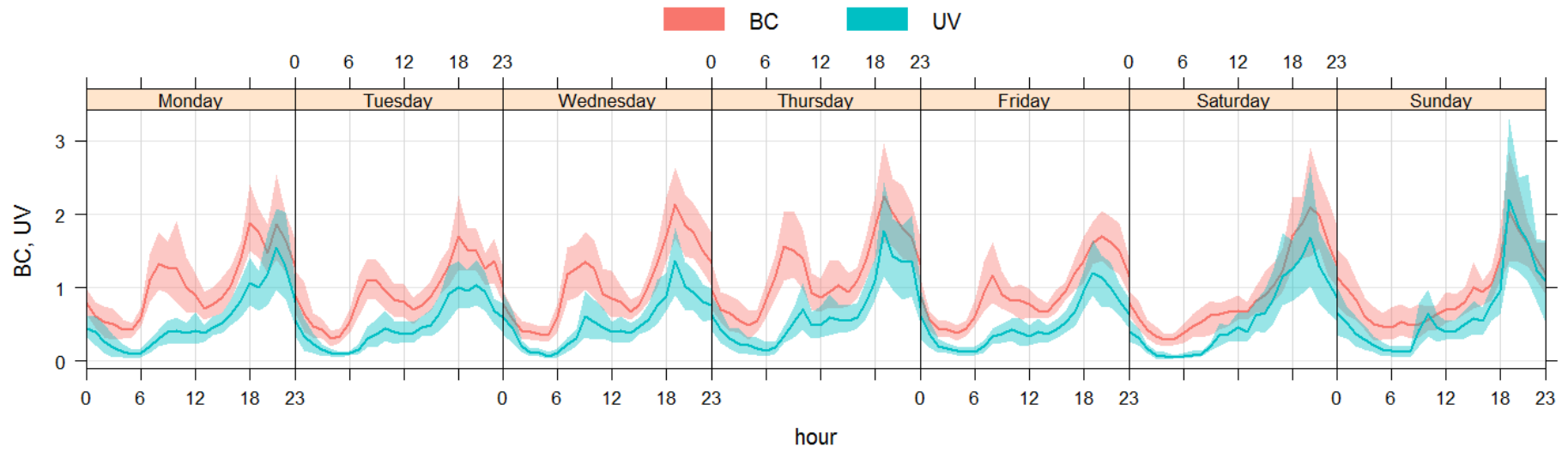


Figure 19 Urban Centre Sites

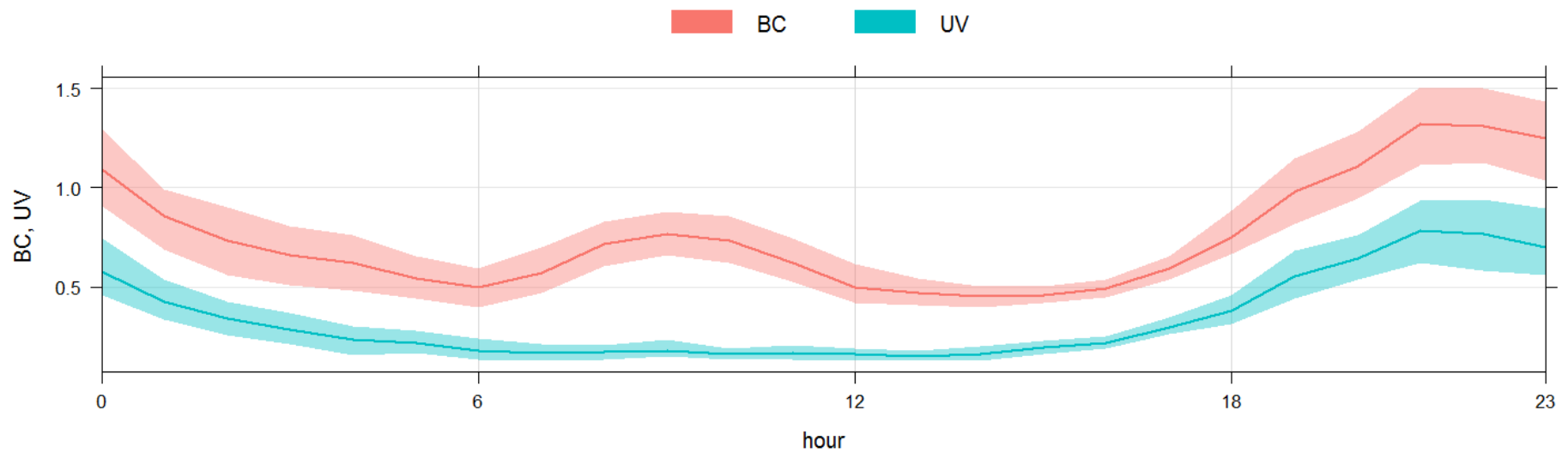
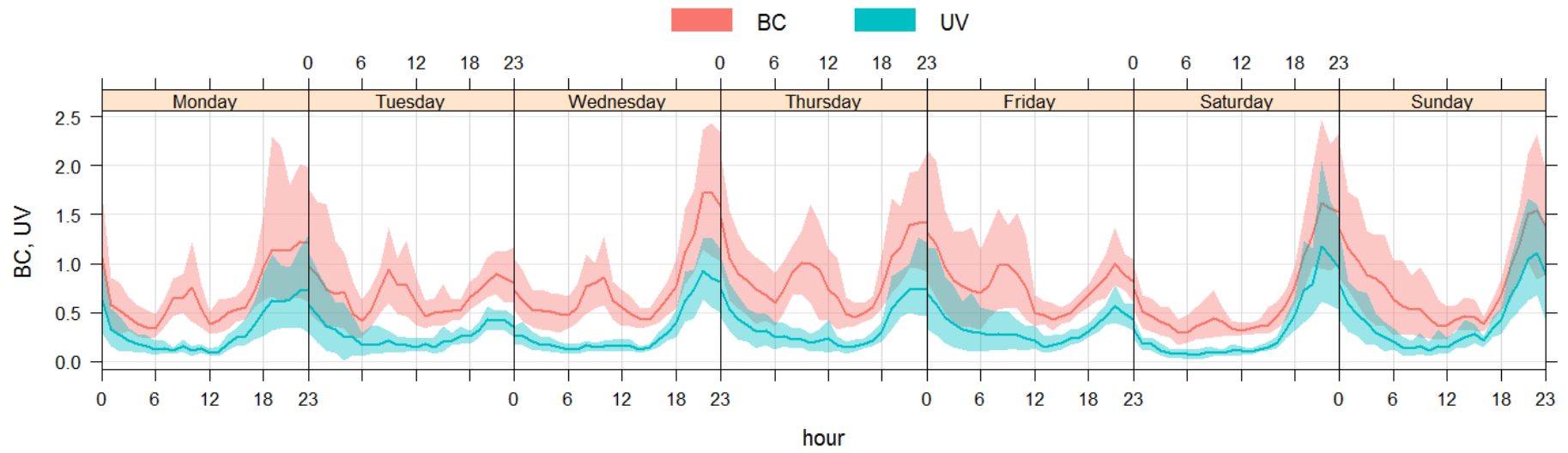
Belfast Centre



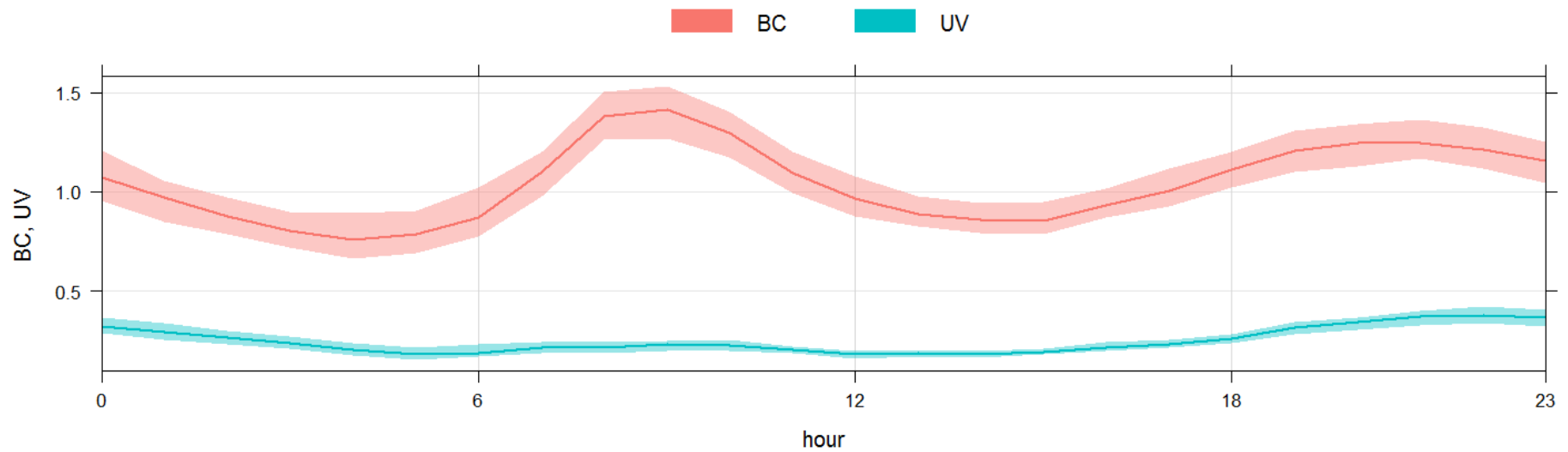
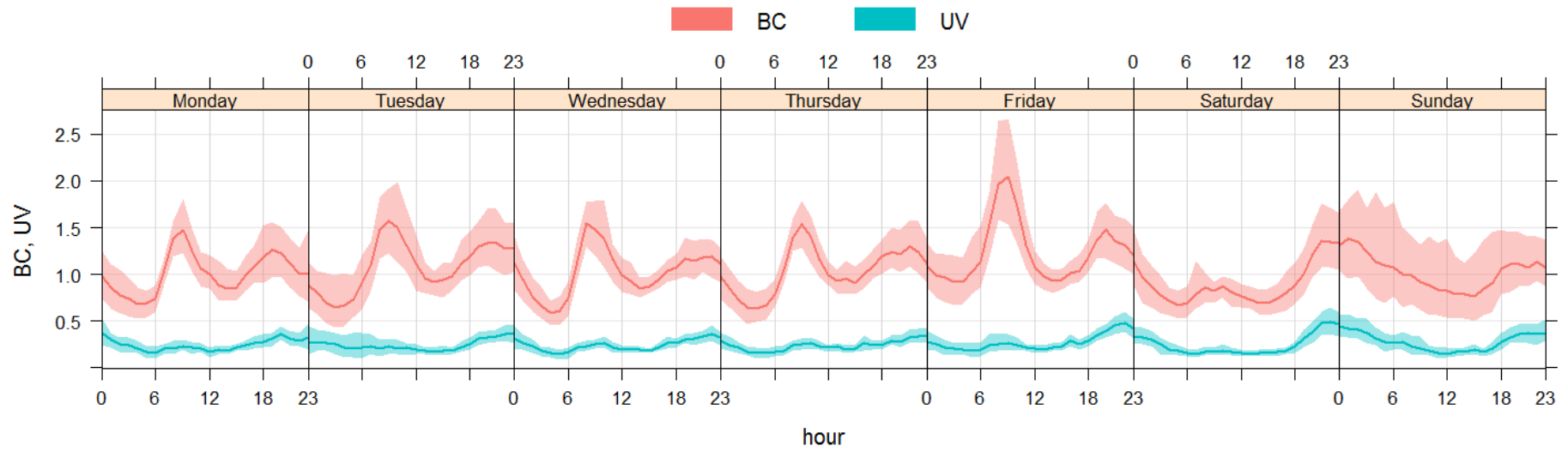




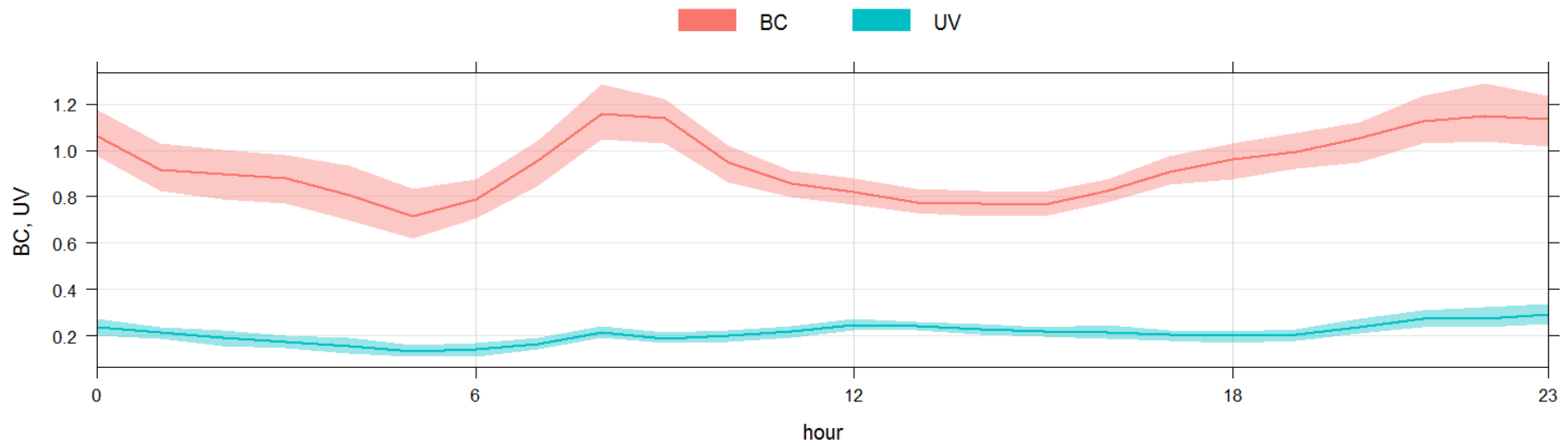
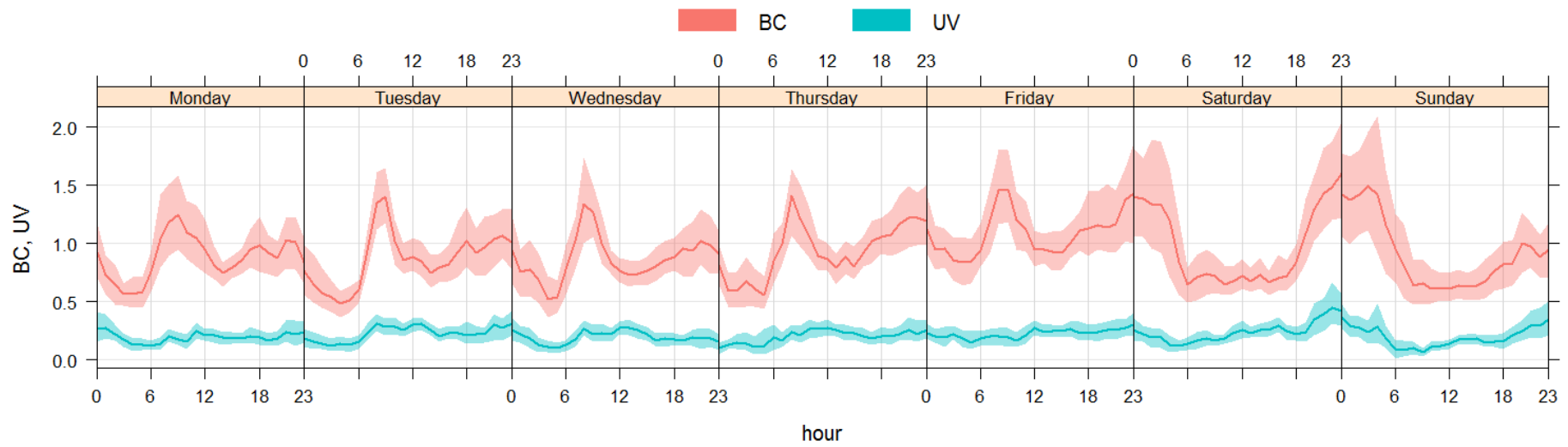
**Ballymena**



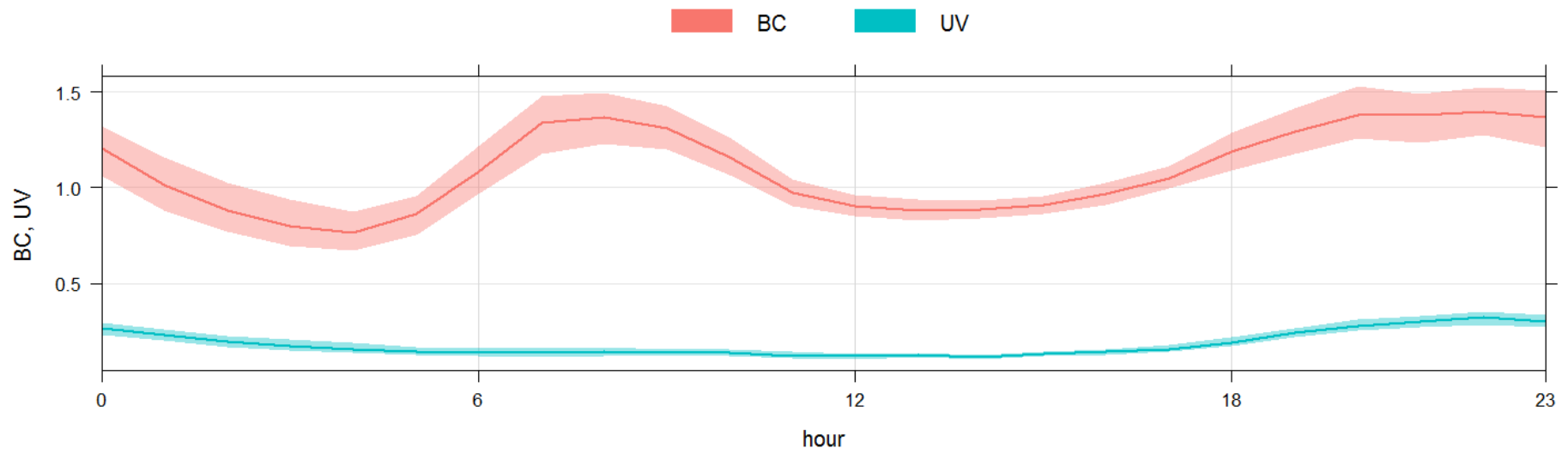
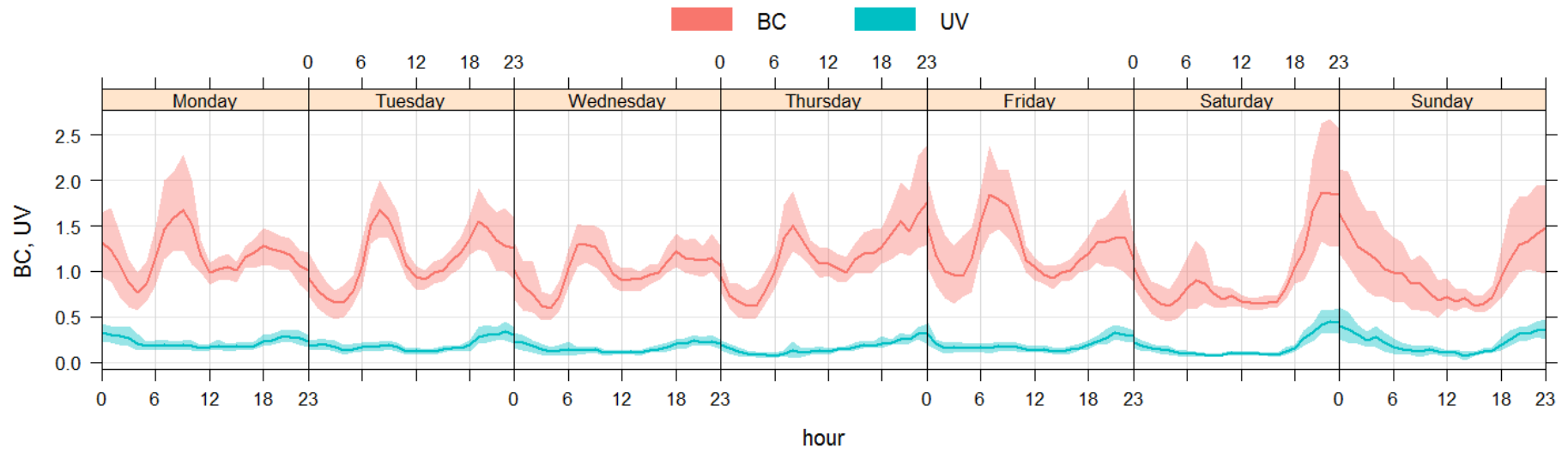
Dunmurry Kilmakee



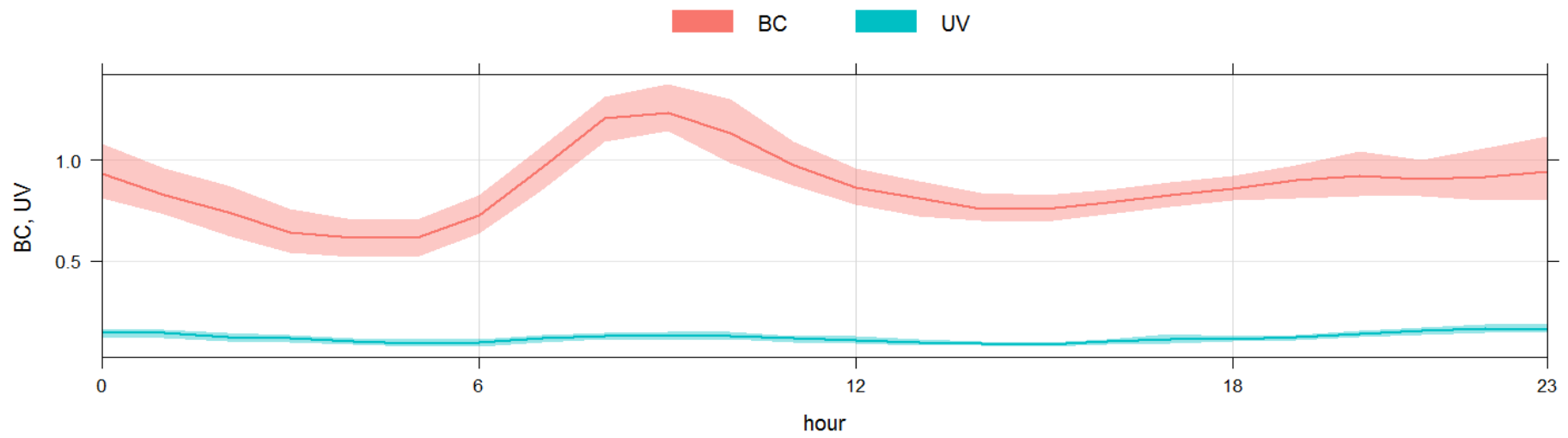
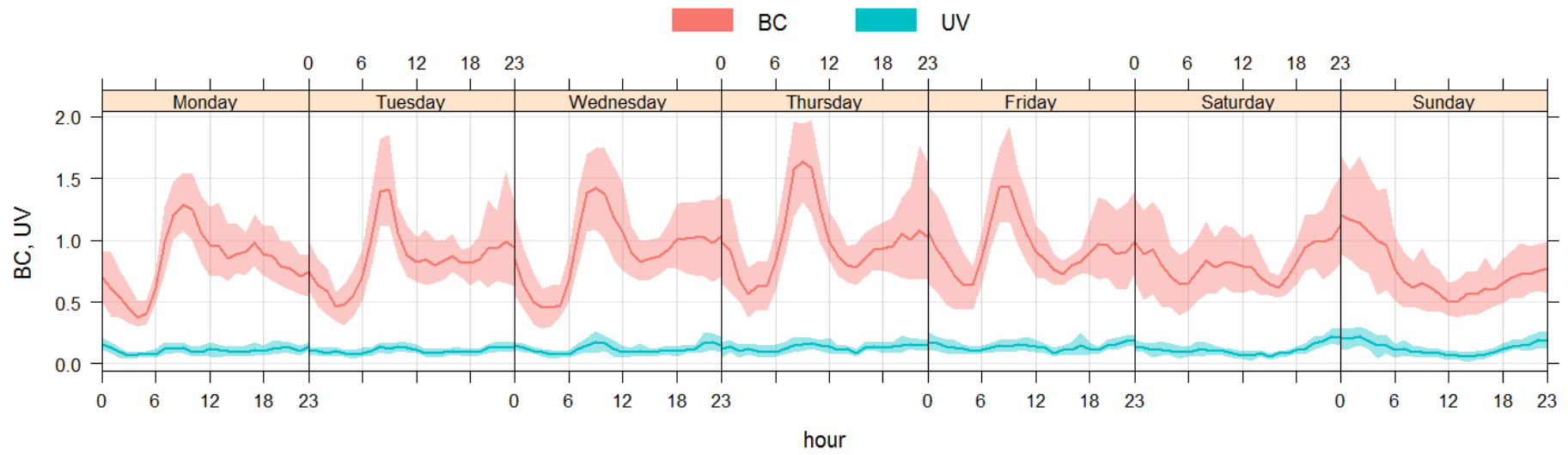
North Kensington



Cardiff Centre

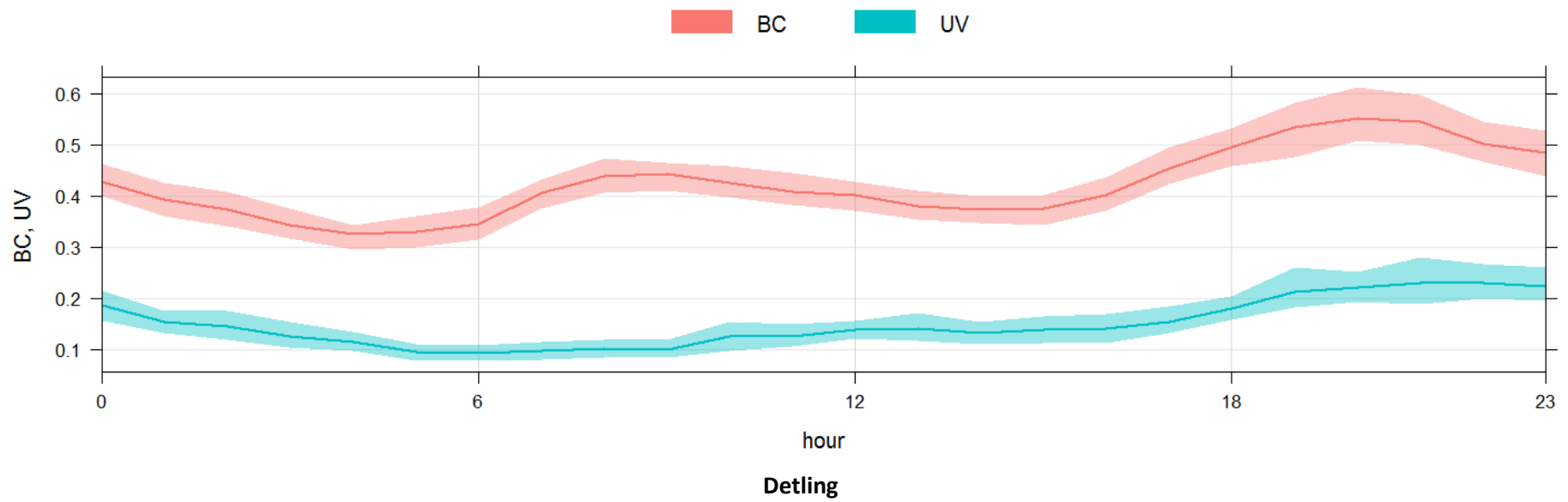
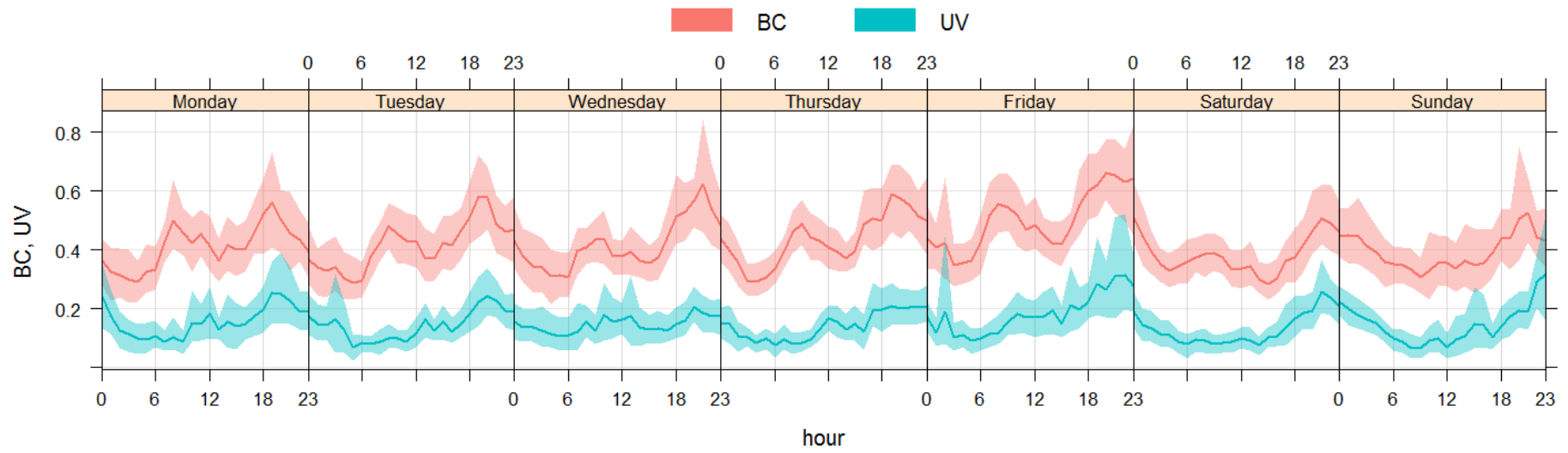


**Birmingham Tyburn**

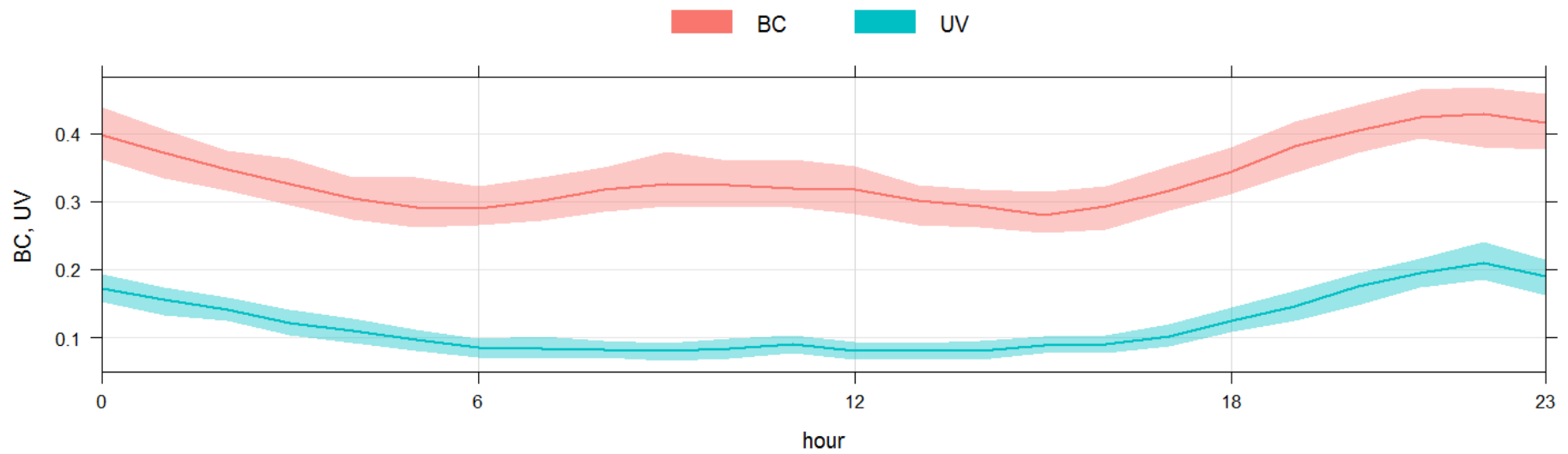
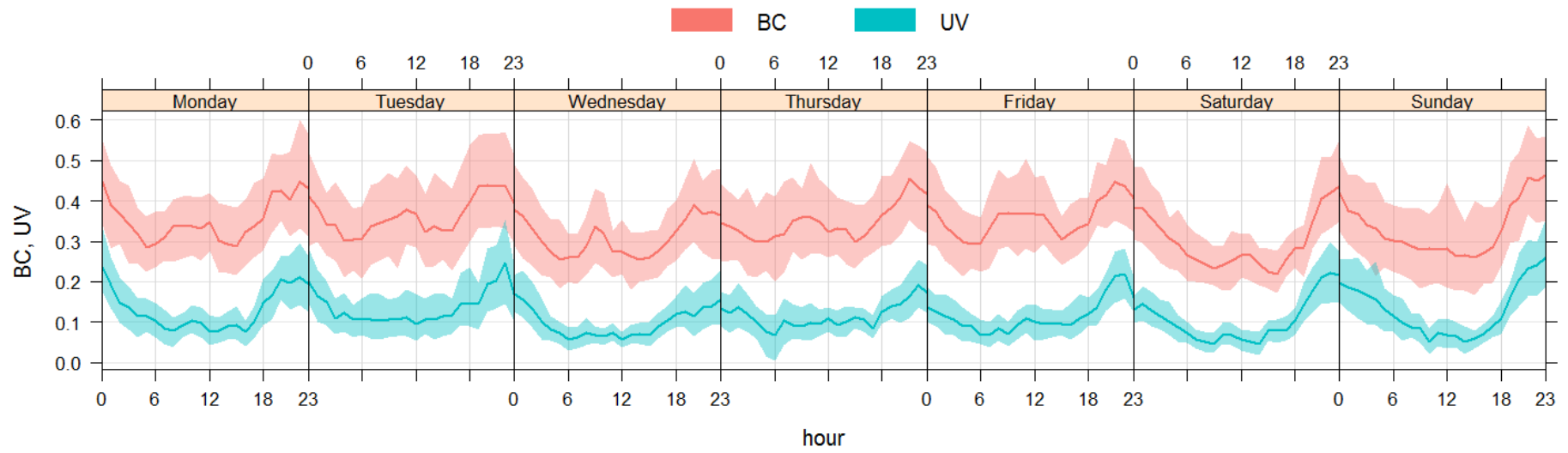


Glasgow Townhead

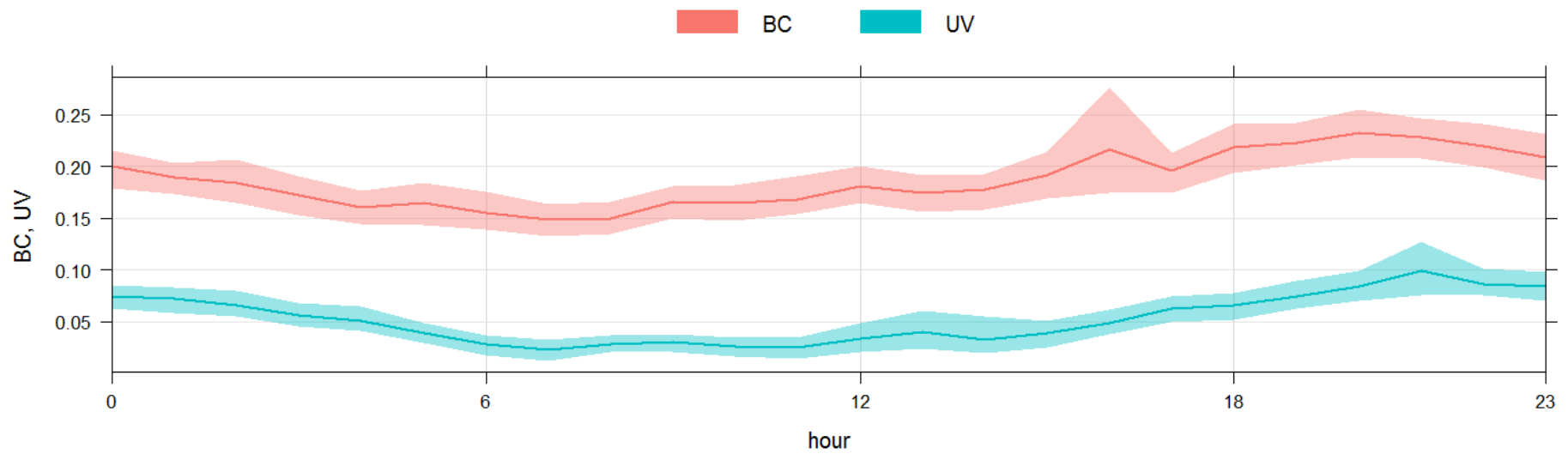
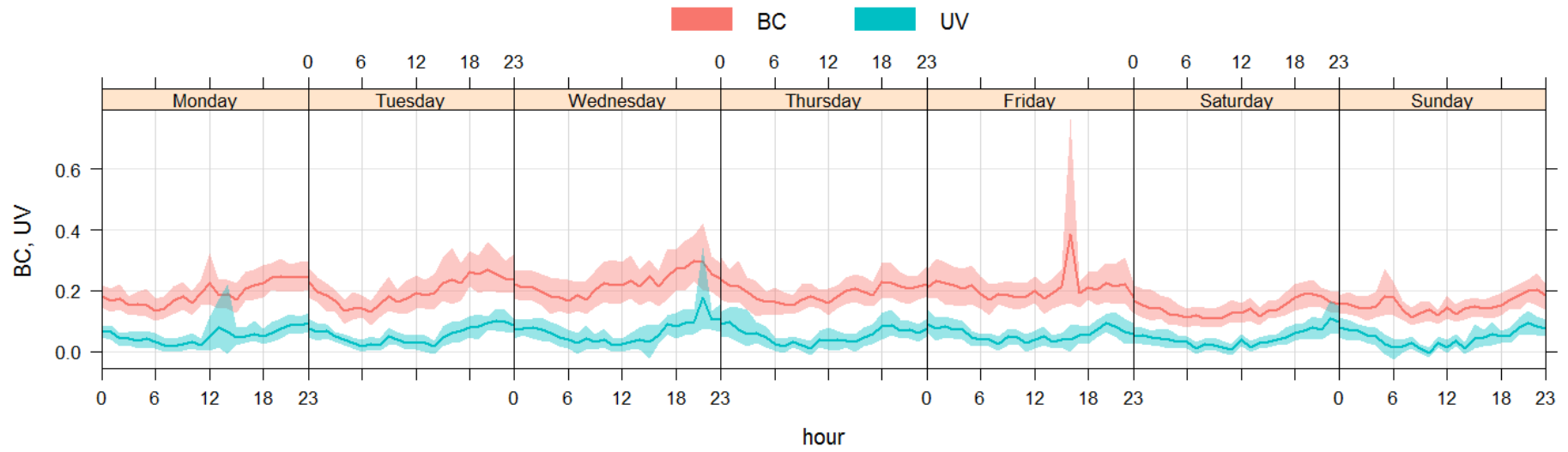
Figure 20 Urban Background Sites







Harwell



Auchencorth Moss

Figure 21 Rural Sites

2009 – 2015 Data

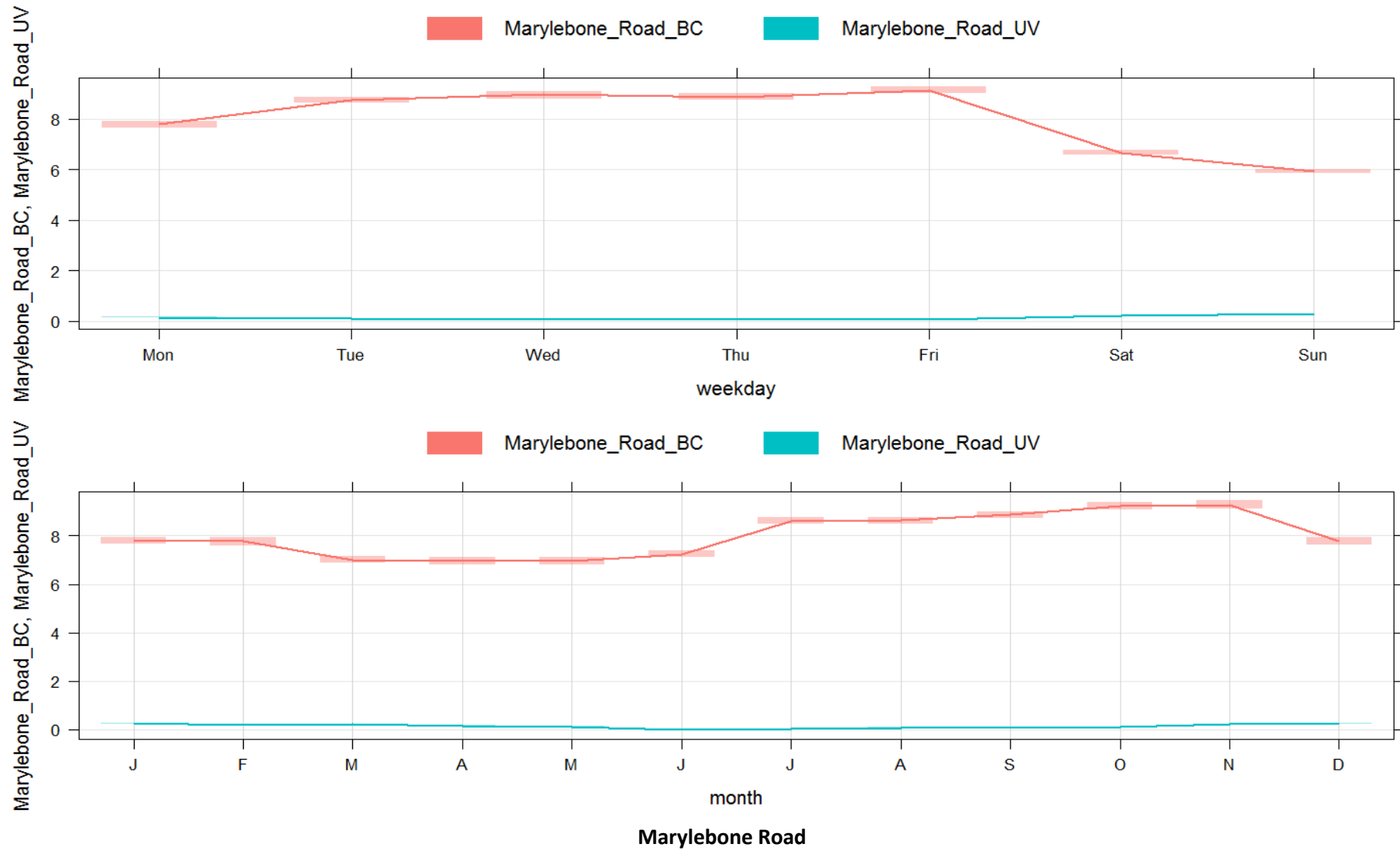


Figure 22 Roadside Sites

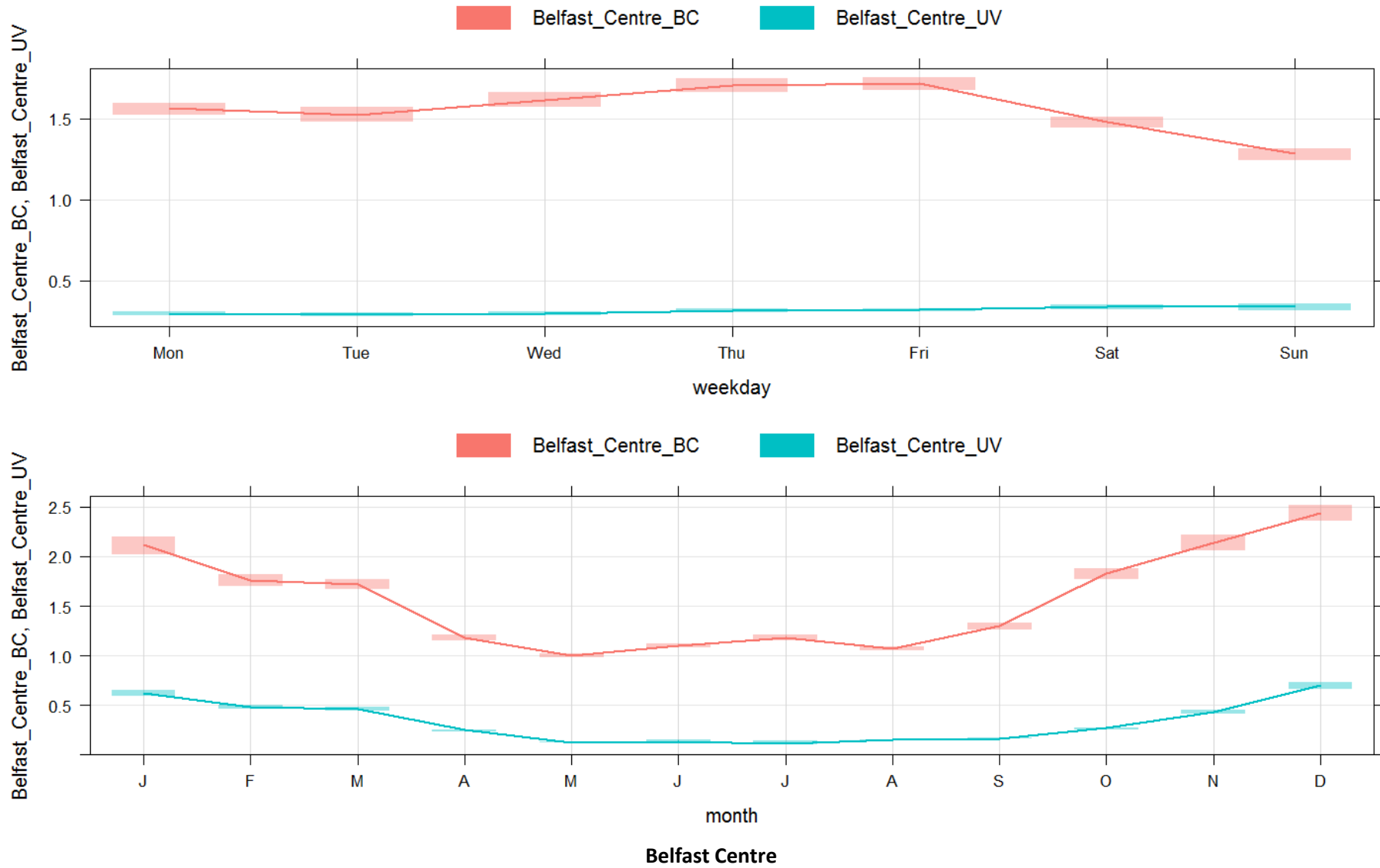
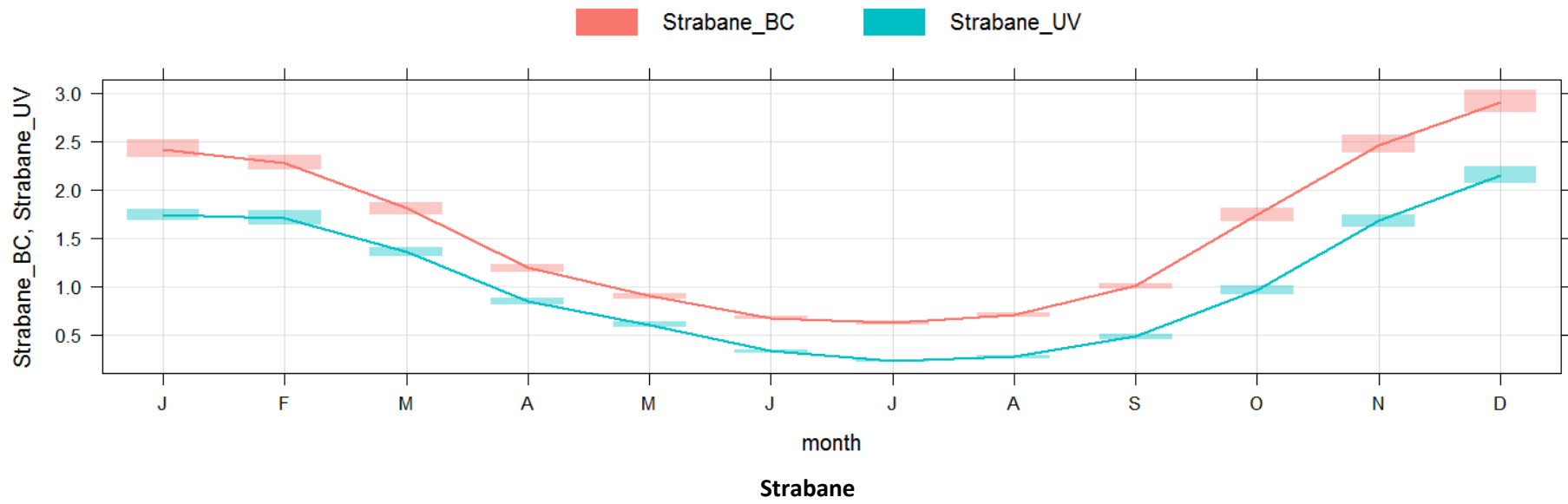
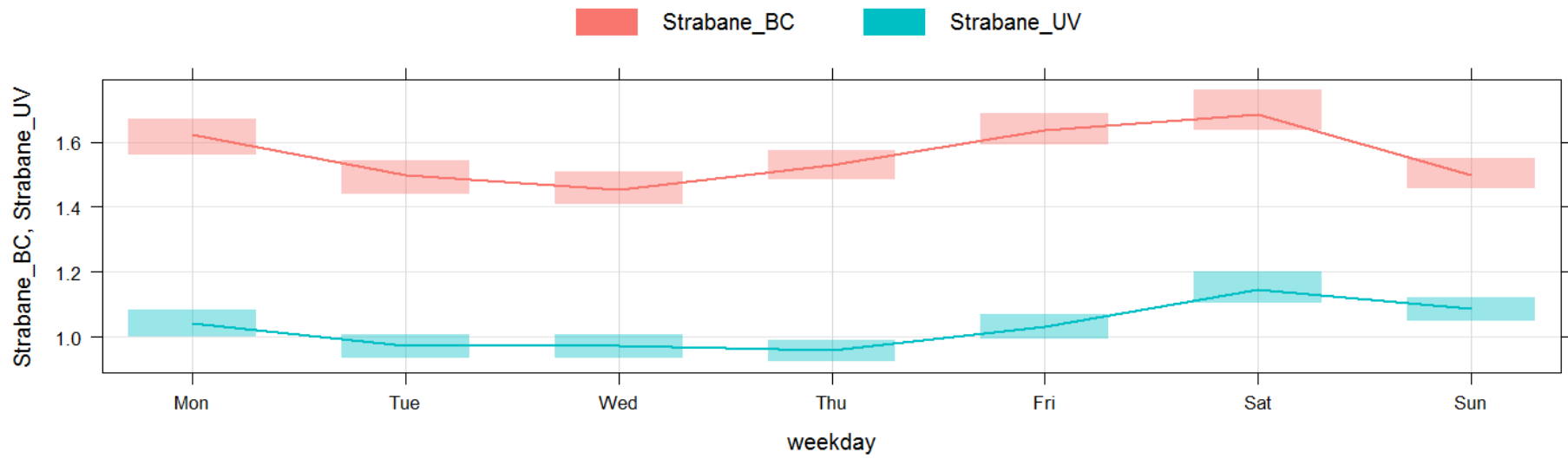
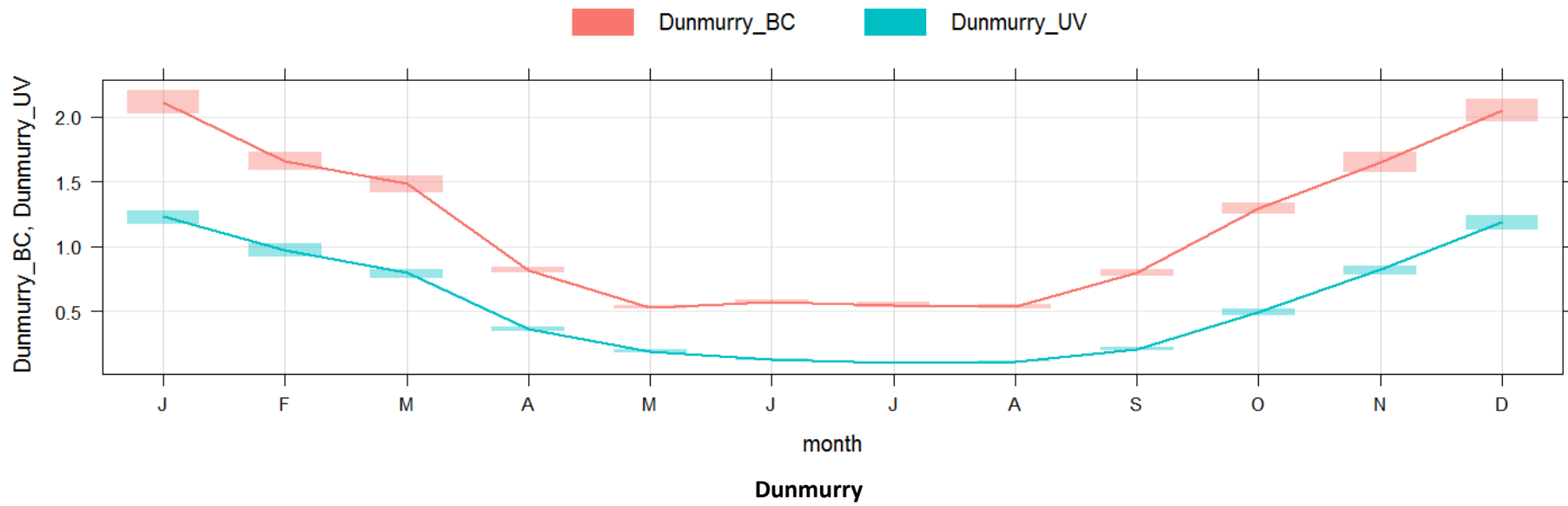
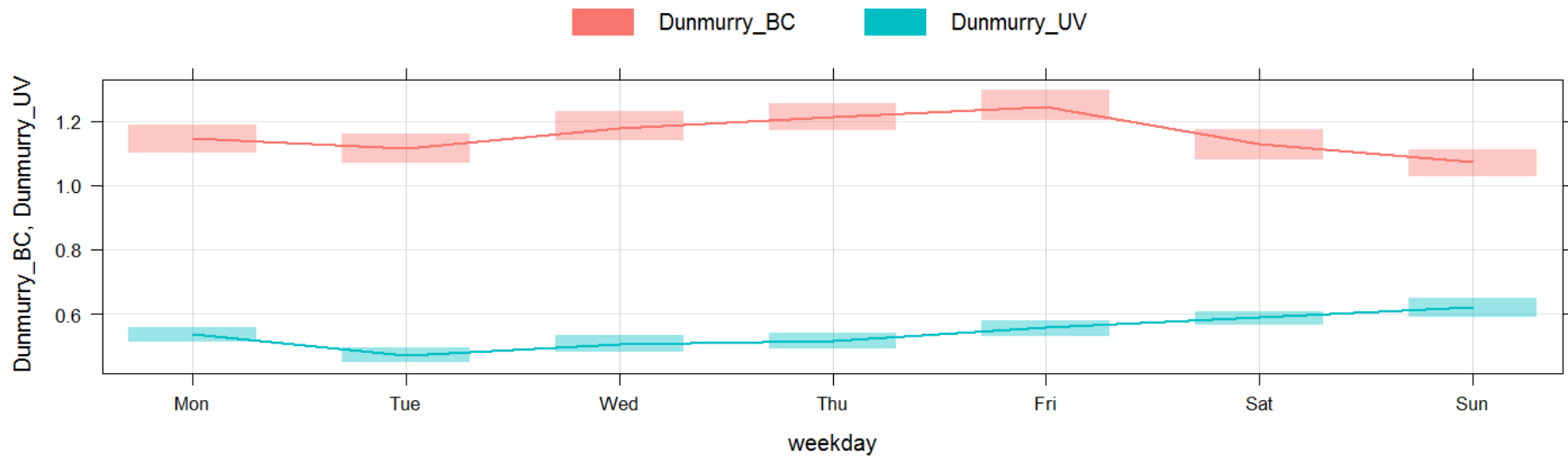
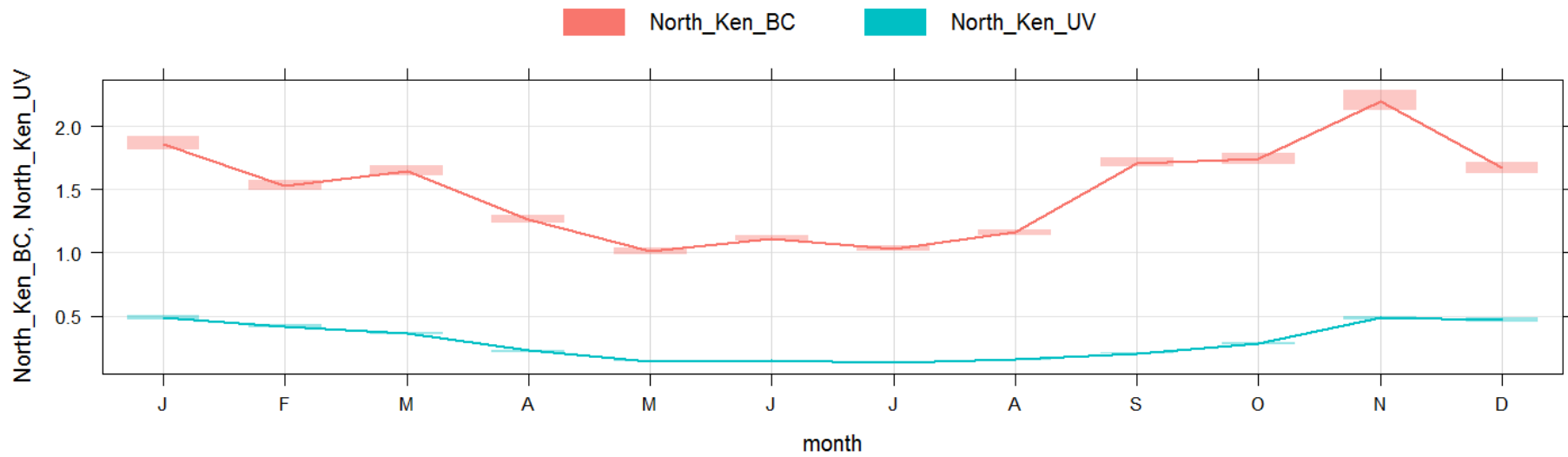
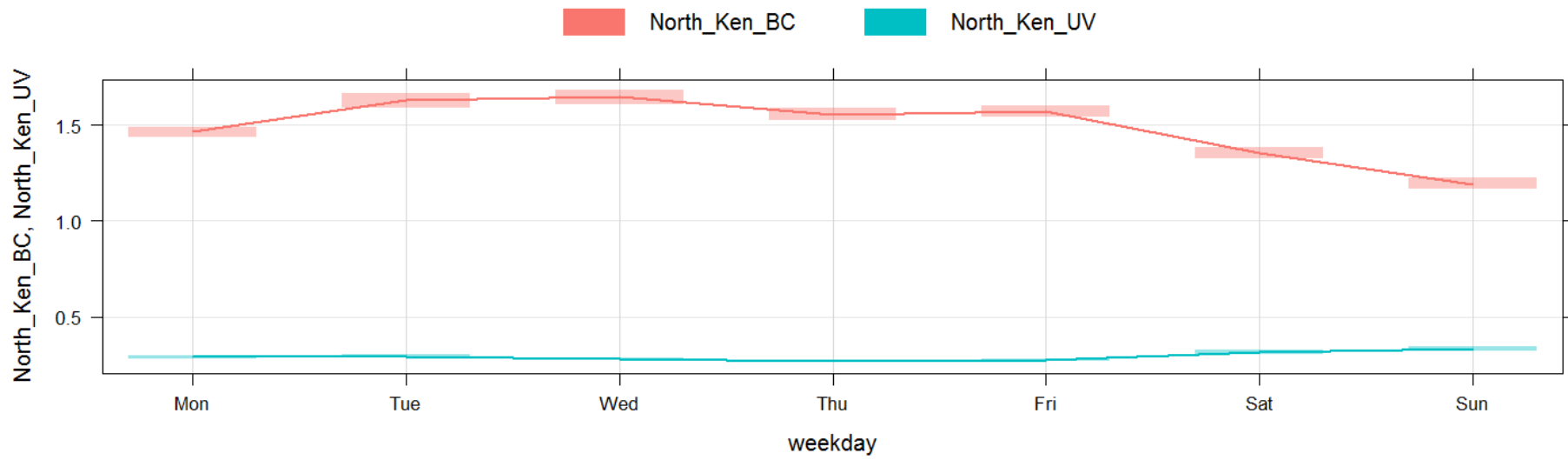


Figure 23 Urban Centre Sites







North Kensington

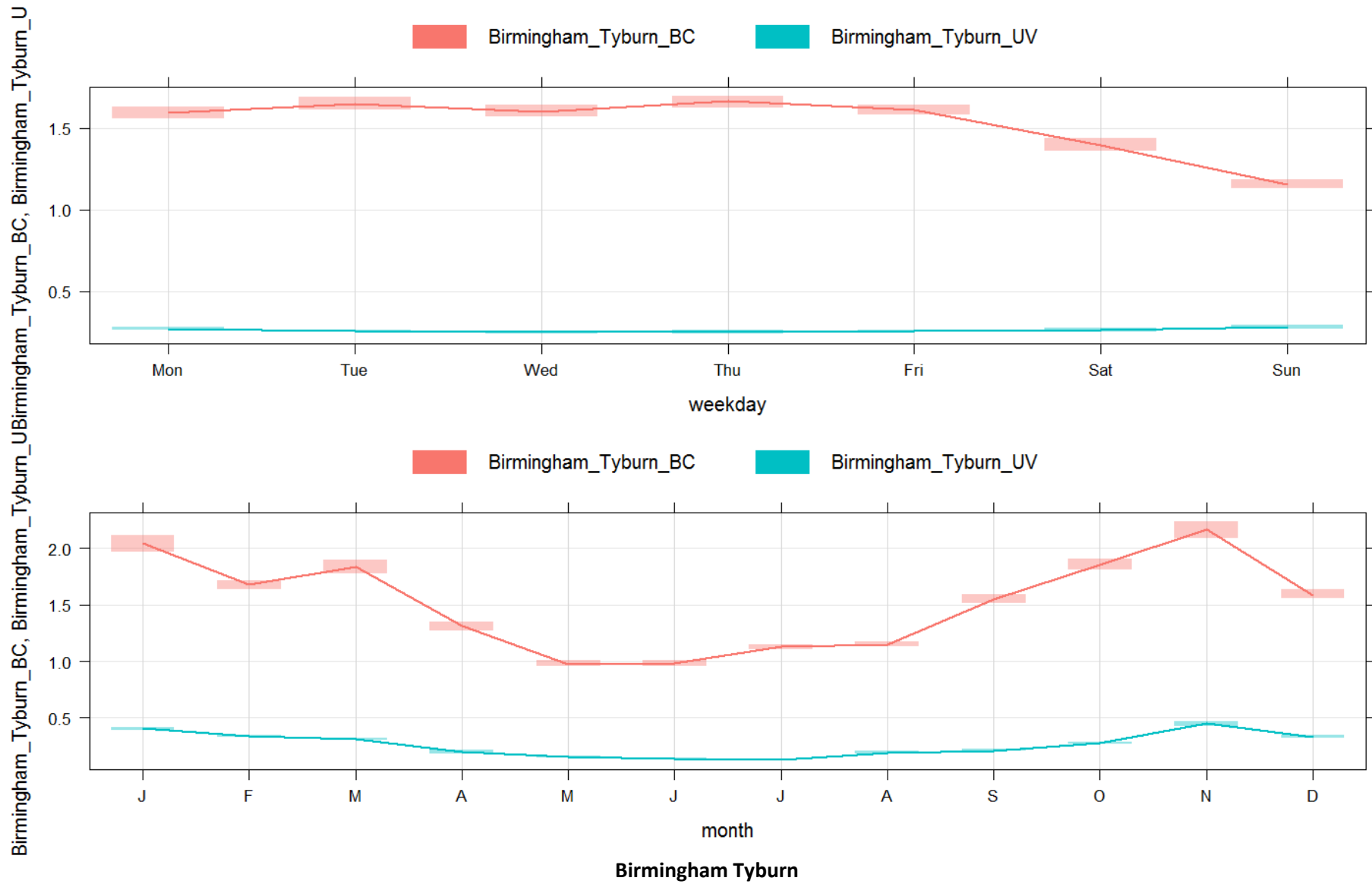
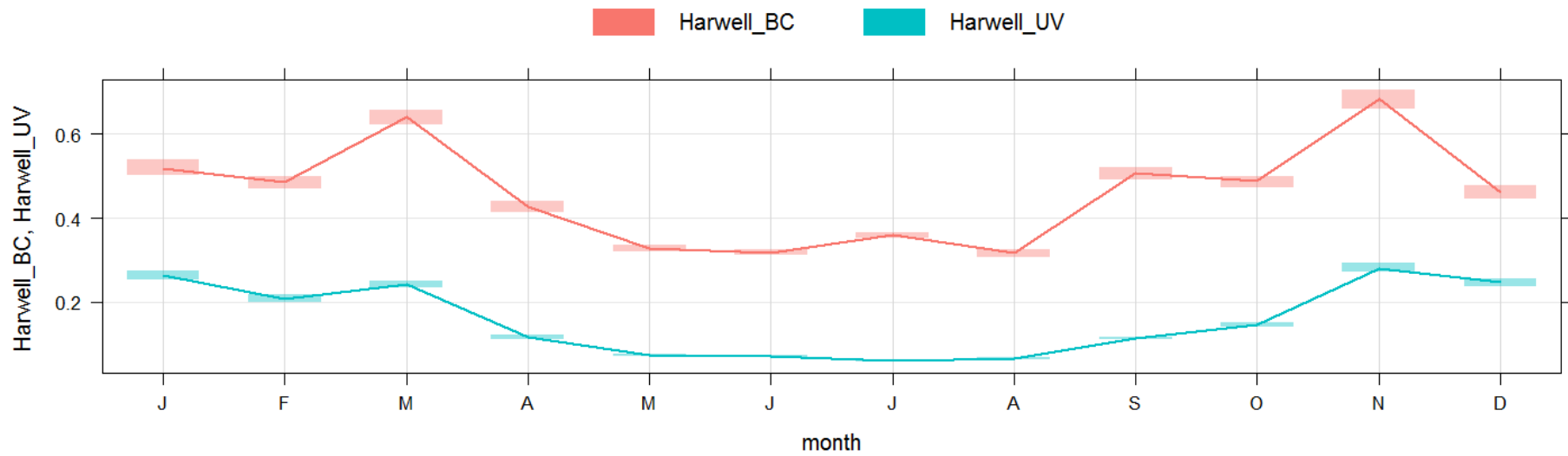
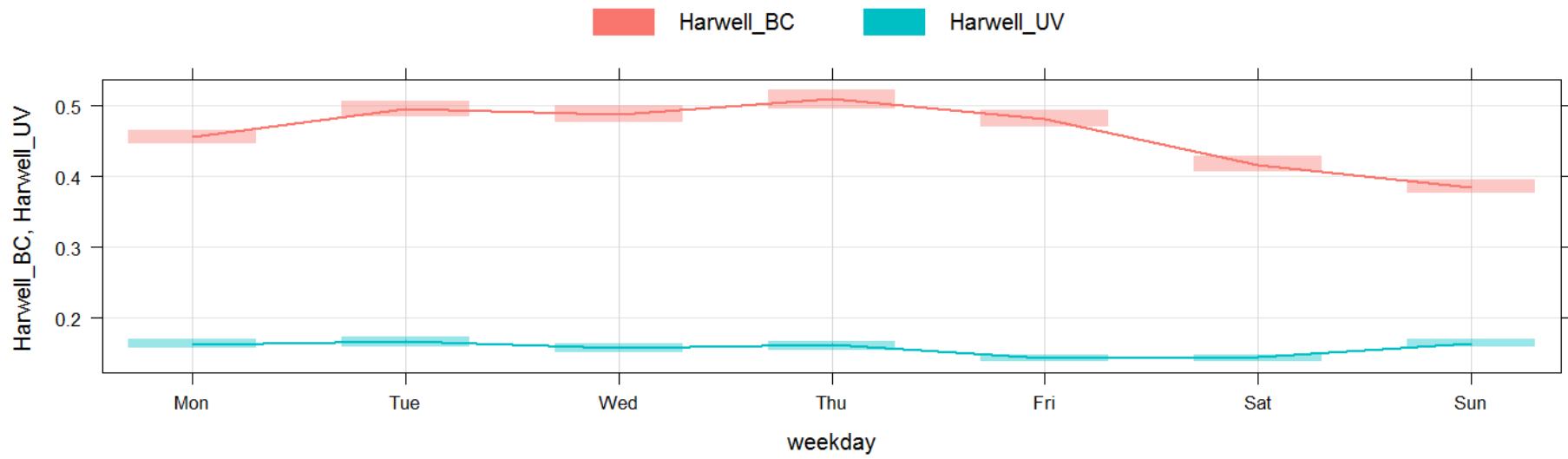


Figure 24 Urban Background Sites





Harwell

Figure 25 Rural Sites

### Roadside sites

It can be seen that the Black Carbon concentrations at the roadside sites follow the expected profile for traffic movements through the day, with raised concentrations in the morning and evening rush hours.

Marylebone Road shows a decrease in Black Carbon concentrations at the weekend, when compared with weekdays. This is in line with the reduced traffic and change in vehicle fleet over the weekend, there is a considerable drop in the number of heavy goods vehicles at the Marylebone Road site over the weekend.

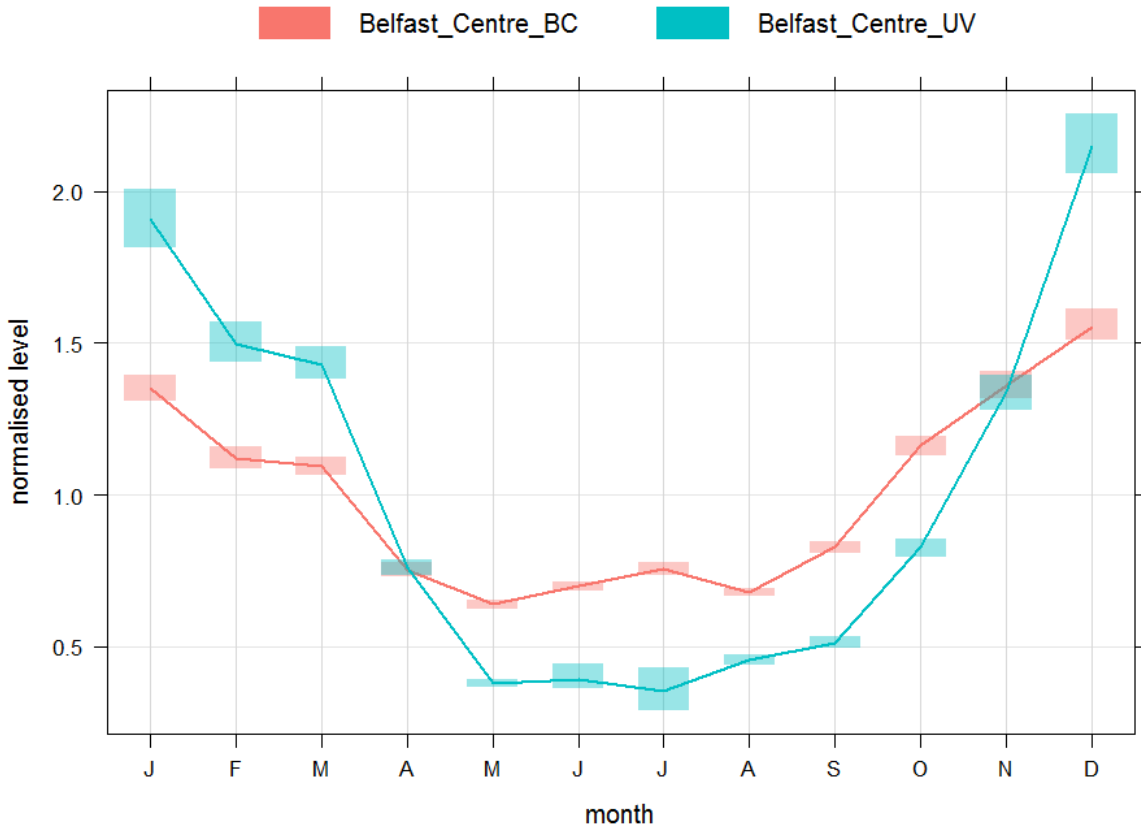
Birmingham Tyburn shows more of the traditional morning rush hour peak of Black Carbon in the morning, followed by a drop in concentrations during the day, with a smaller peak for the evening rush hour. Weekend concentrations are also much lower and relatively flat during the day. As with Marylebone Road there is little UV component signature, however Birmingham shows a slight increase in the evenings of the weekend, indicating possible local solid fuel / wood burning secondary heating.

Glasgow High Street also shows the traditional morning and evening rush hour peaks of Black Carbon, with lower, more stable concentrations at the weekend.

### Urban Centre sites

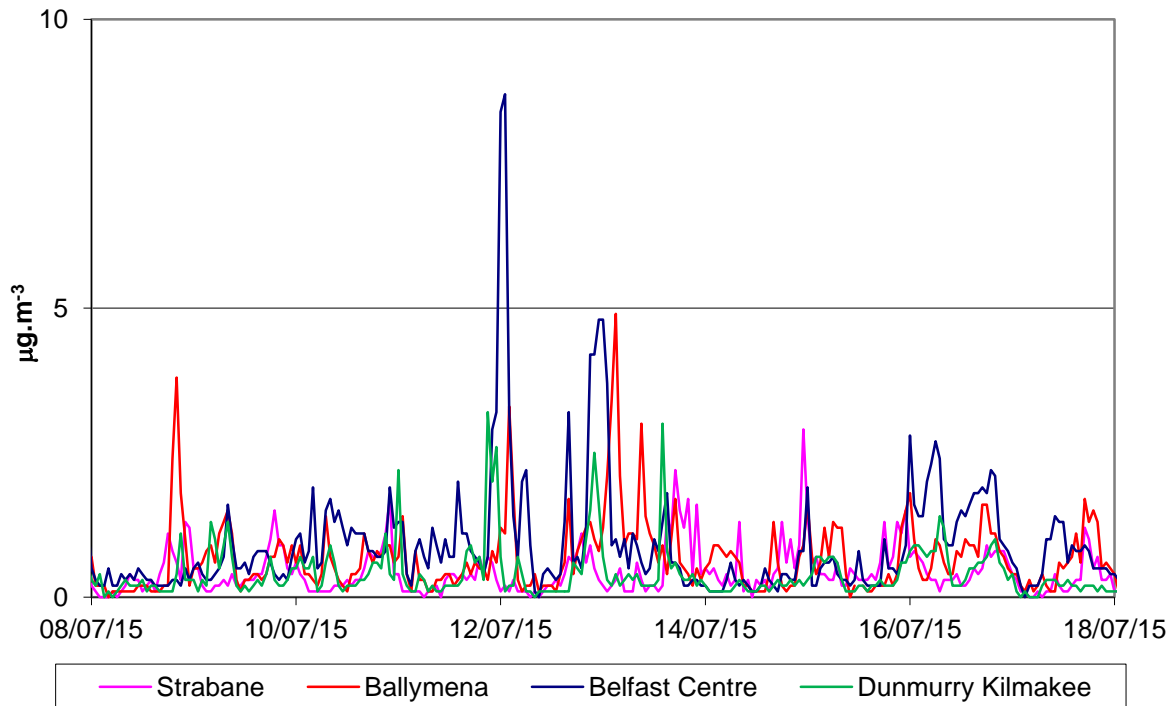
Belfast centre also shows an increase in Black Carbon concentrations coinciding with the morning rush hour, but with concentrations remaining fairly constant from 17:00 hours until midnight. Black Carbon concentrations also show an increase late on Saturday evening / early Sunday morning, which can be assumed to be due to evening leisure journeys.

Black Carbon concentrations are generally lower at the weekends compared with working days. Both Black Carbon and UV component concentrations show some seasonality 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 are relatively consistent through the year then normalising the concentrations should reveal any monthly variability in either the Black Carbon or UV component emissions. This can be seen in Figure 26.



**Figure 26** Normalised monthly variability at Belfast for the period 2009 - 2015

The increase in Black Carbon in July is due to localised burning around the period of 12 July as shown by Figure 27.

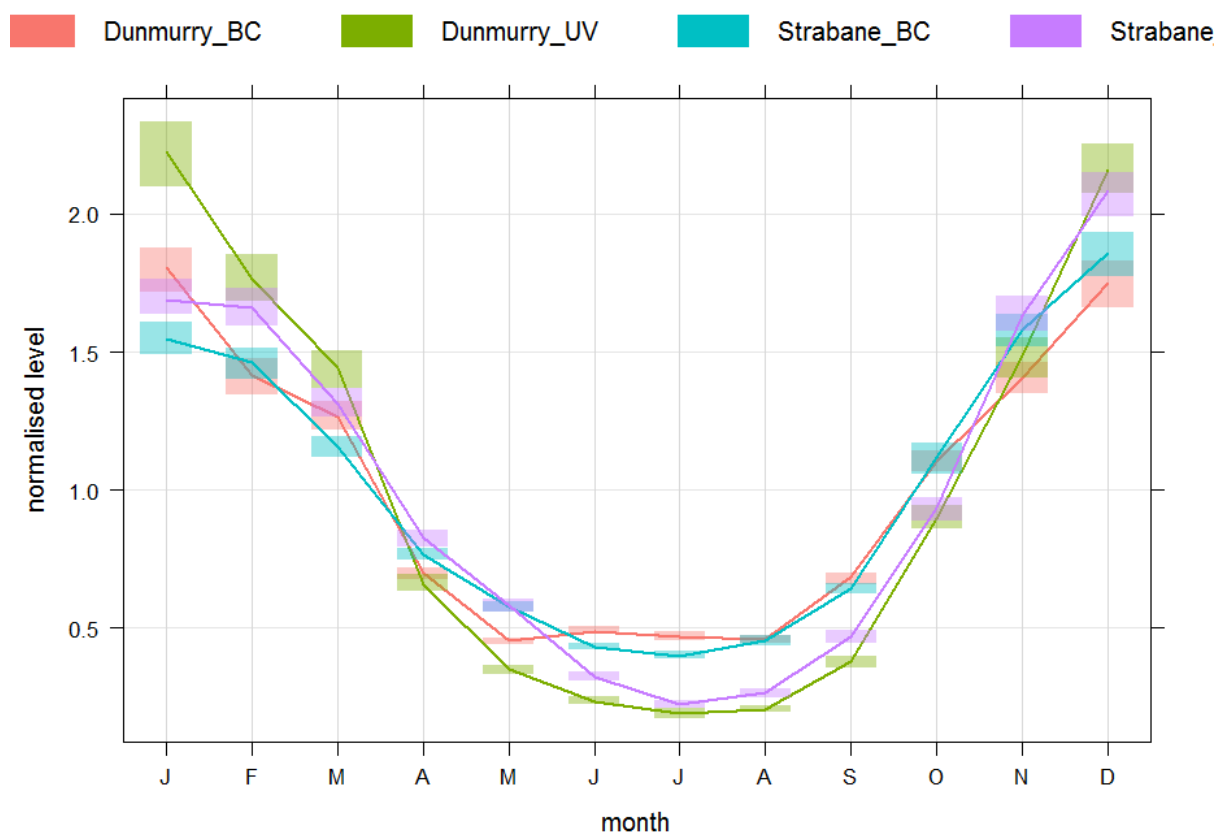


**Figure 27** Black Carbon concentrations in Northern Ireland around the period of 12<sup>th</sup> July

Urban Background sites

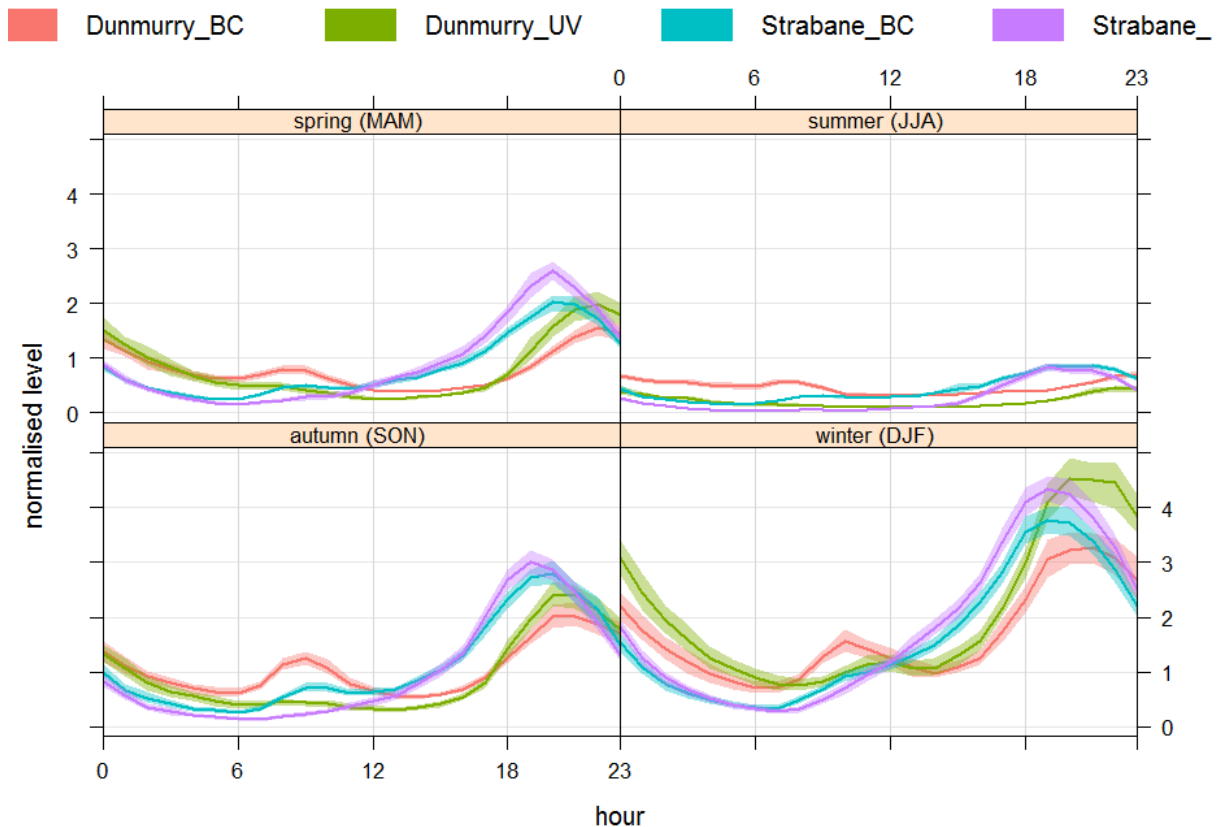
Urban background sites can be split into two categories: those predominantly influenced by emissions from domestic heating and those away from main roads with mixed influences from both road traffic sources and emissions from domestic sources.

Concentrations measured at Strabane, Ballymena and Dunmurry Kilmakee in Northern Ireland are dominated by emissions from domestic heating. UV component and Black Carbon concentrations follow similar hourly, daily and seasonal trends and have similar absolute concentrations. Strabane is not on the natural gas supply and domestic heating mainly comes from oil. Strabane is in a smokeless zone, however there is evidence that this is being ignored in some areas of the town and residents are burning smoky coal. Due to the large emission factors of PAHs from smoky coal<sup>7</sup> compared to oil and gas, it does not take many houses burning this coal to have a big influence on ambient concentrations. At Strabane 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 Ballymena and Dunmurry the morning rush hour is picked up in the Black Carbon concentrations, but the evening rush hour is masked by domestic emissions. Figure 28 gives the normalised monthly variability and Figure 29 gives the hourly variability



**Figure 28** Normalised monthly variability at Strabane and Dunmurry for the period 2009 - 2015

7 UK National Atmospheric Emissions Inventory



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 29** Seasonal Black Carbon and UV component concentrations measured at Strabane and Dunmurry for the period 2009 - 2015

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

Concentrations measured at Birmingham Tyburn, Cardiff Centre, North Kensington and Glasgow Townhead show expected peaks in Black Carbon concentrations during the morning rush hour, with little increase in UV component concentrations. However the evening rush hour peak is less pronounced and high Black Carbon concentrations continue into the evening. At the weekend there are elevated Black Carbon concentrations late into Saturday evening / early Sunday morning, combined with a similar increase in UV component concentration indicating domestic emission sources, probably from secondary heating.

#### Rural sites

The rural background site concentrations are lower than the other site classifications, as expected. The effect of local heating can also be seen in the concentrations at the Detling suburban site. Detling and Harwell sites display the morning rush hour in the Black Carbon concentration to a small extent, while at Auchencorth Moss there is little change in concentration over the day in both Black Carbon and UV component.

Over the longer dataset, Harwell shows some seasonality in the UV component concentration associated with domestic emissions, and reduced Black Carbon concentrations at weekends associated with lower local traffic flows.

## **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 measurements where these instruments were collocated with the Aethalometer.

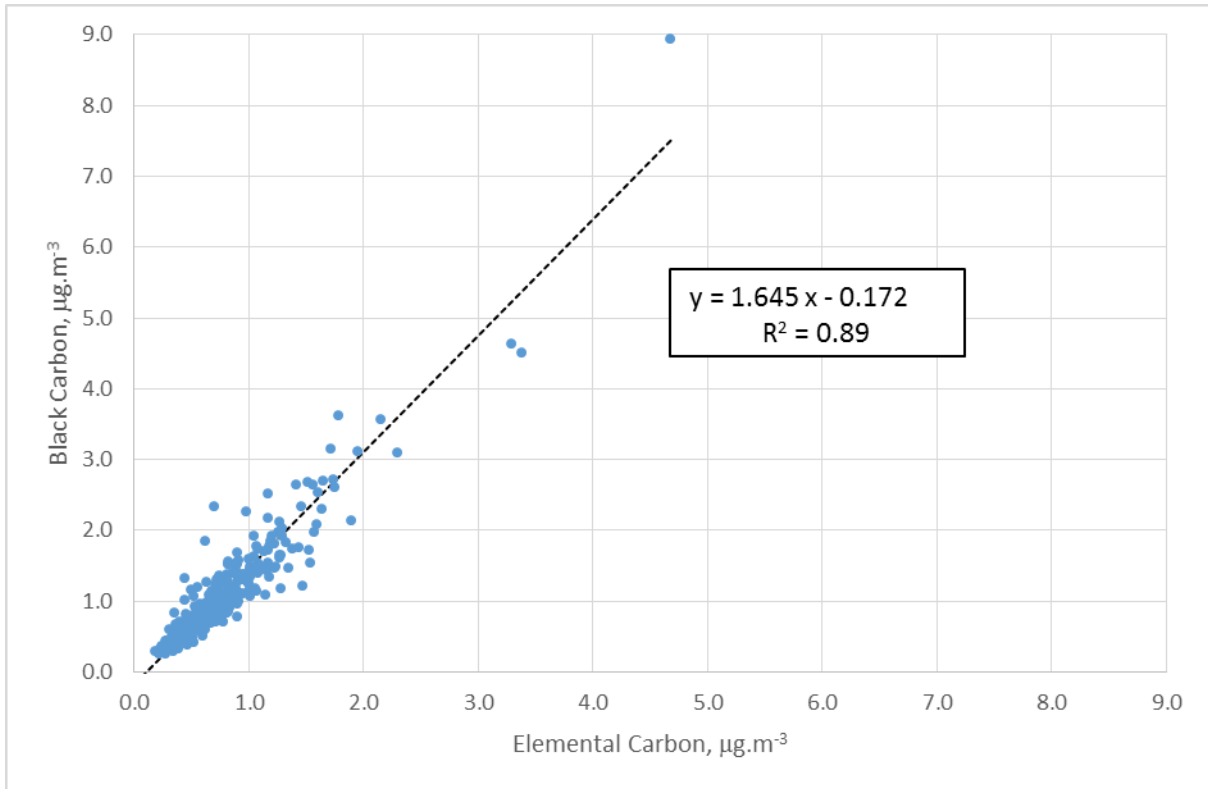
### **5.4.1 Elemental Carbon**

Daily Elemental Carbon (EC) measurements are made at the North Kensington, Marylebone Road and Harwell sites by the Particle Concentration and Number Network<sup>8</sup>. 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 30 to 32. The regression is calculated according to the Reduced Major Axis (RMA) method<sup>9</sup>, 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 dependent on each other, so there is no real separation into dependent and independent variables. Also deviations between fitted and observed data values will occur in both *x* and *y* directions due to random measurement uncertainties.

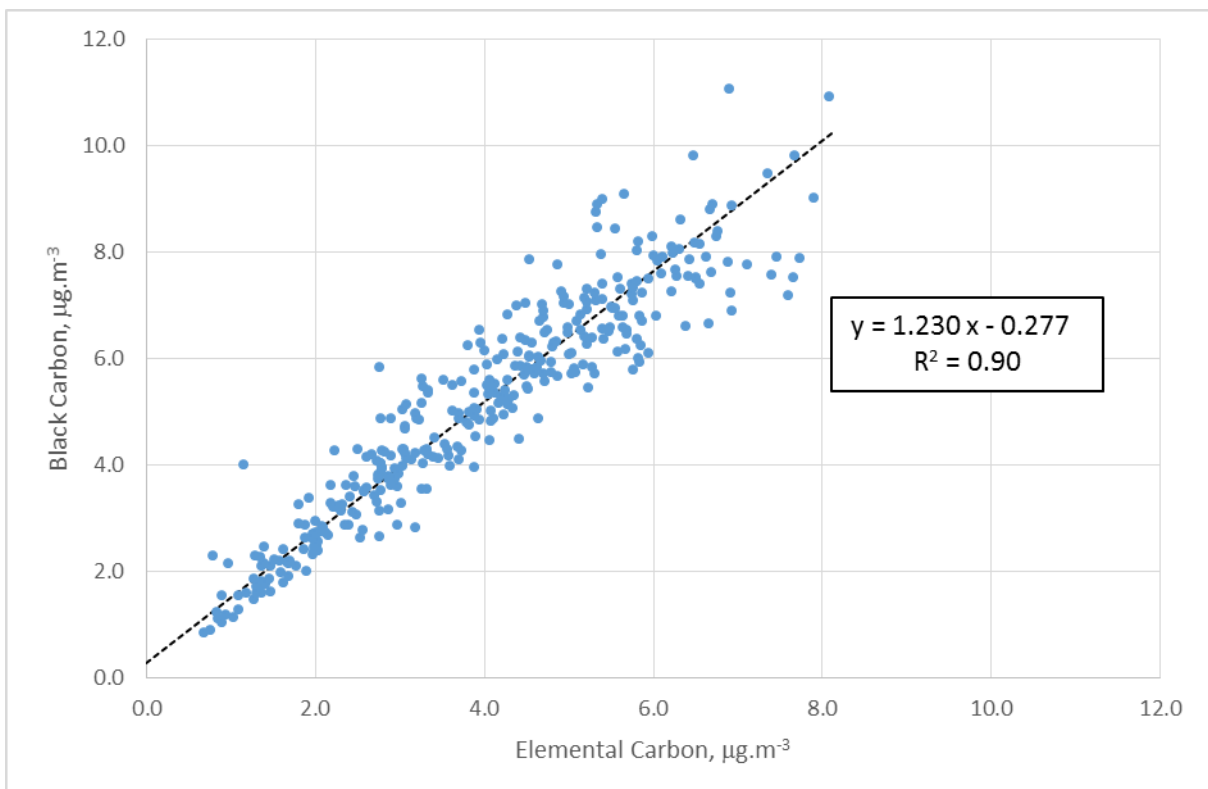
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<sup>8</sup> S Beccaceci et al, Draft NPL REPORT, 2015 Annual Report for Airborne Particulate Concentrations and Numbers in the United Kingdom (phase 3), June 2015

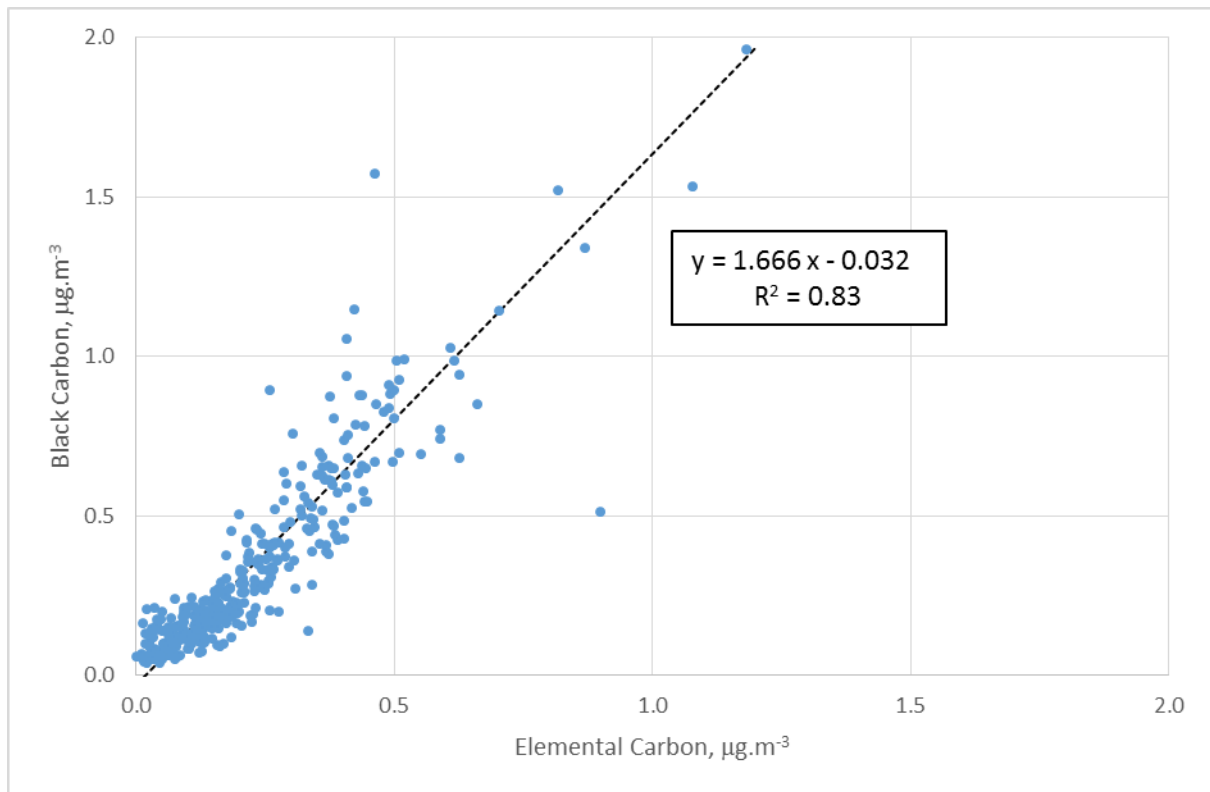
<sup>9</sup> G.P. Ayers, Comment on regression analysis of air quality data, Technical Note, Atmospheric Environment, 35 (2001) 2423 - 2425



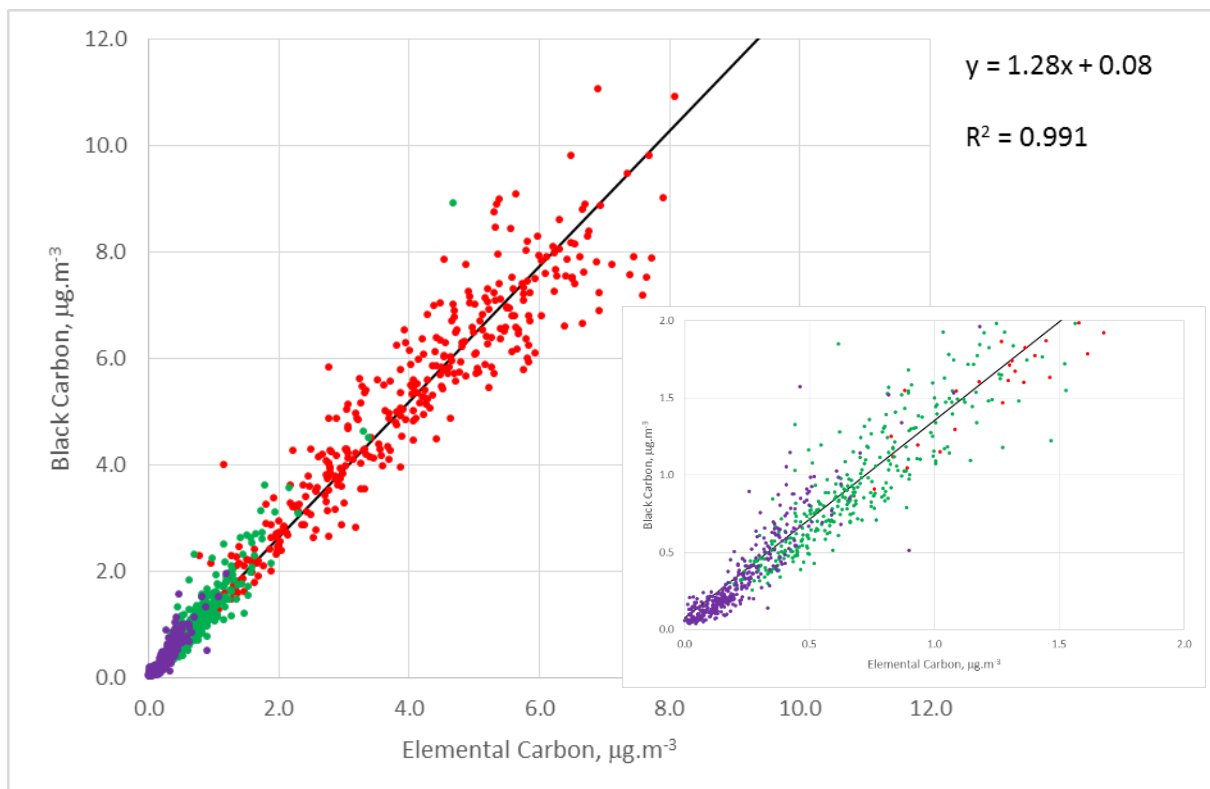
**Figure 30** 2015 EC and BC Measurements at North Kensington



**Figure 31** 2015 EC and BC Measurements at Marylebone Road



**Figure 32** 2015 EC and BC Measurements at Harwell



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

**Figure 33** 2015 EC and BC Measurements for all 3 sites. The inset graph shows points close to the origin.



It can be seen that there are good linear relationships ( $R^2 > 0.8$ ) between the Elemental Carbon and Black Carbon concentrations measured at all three sites. The relationship between Black Carbon and Elemental Carbon is markedly 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 33. In all cases the intercept value is relatively small, indicating that there is 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       |                | North Kensington |                | Marylebone Road |                |
|------|---------------|----------------|------------------|----------------|-----------------|----------------|
|      | Relationship  | R <sup>2</sup> | Relationship     | R <sup>2</sup> | Relationship    | R <sup>2</sup> |
| 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          |

\*There is not enough BC data collected at Harwell in 2009 to form a reliable relationship as the Aethalometer was only installed in November 2009.

**Table 13 Relationships between Black Carbon and Elemental Carbon over the period 2009 – 2015**

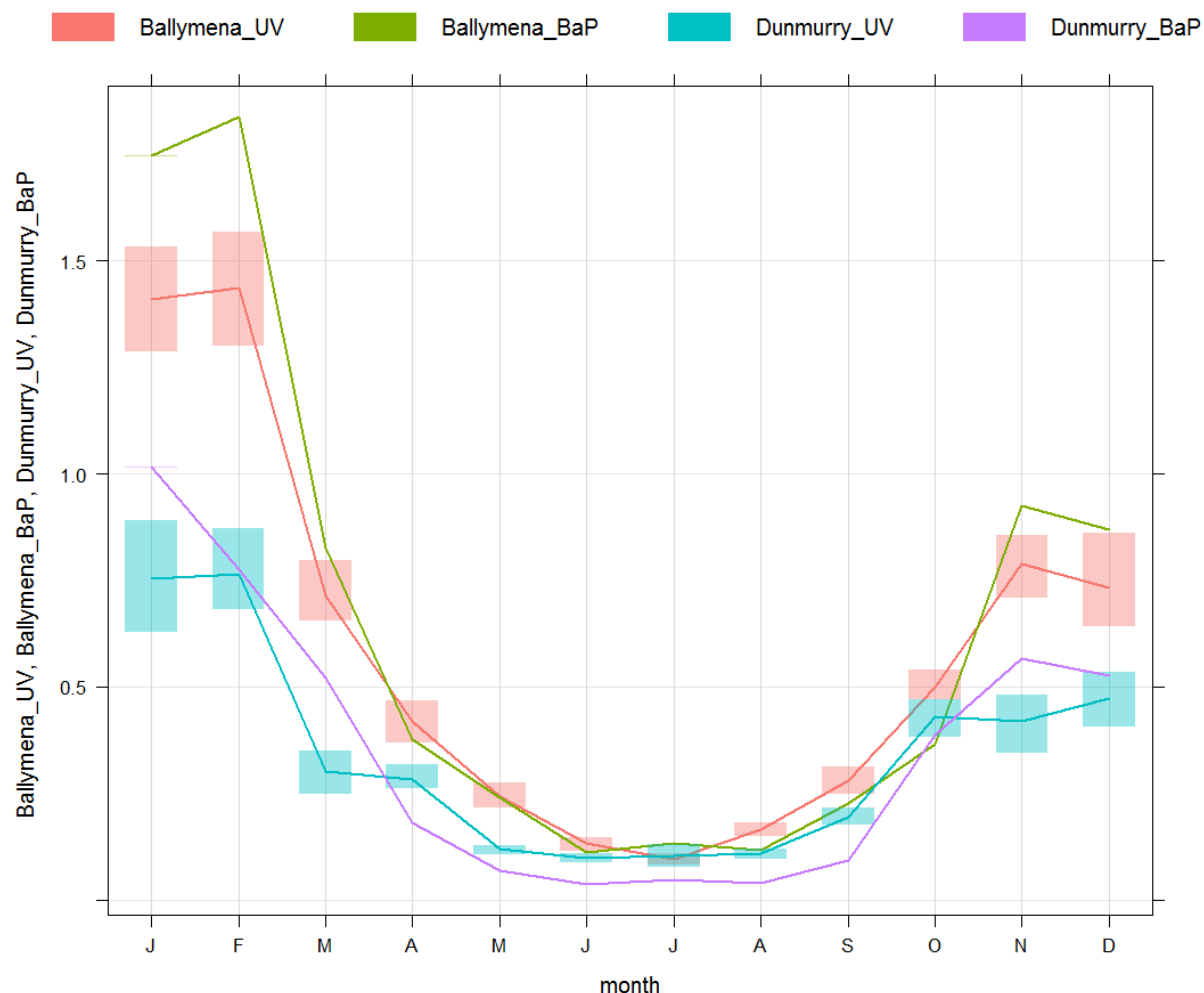
It can be seen that the slopes, i.e. the measured amounts of Black Carbon relative to Elemental Carbon, have been roughly constant at Marylebone Road, at  $1.4 \pm 0.1$ , but this dropped to around 1.2 in 2015. A drop in slope was also seen at Harwell in 2015 (2.0 to 1.7), where previously the slope had been increasing from 2010 to 2014 (from around 1.3 to 2.0). The slope over the last 3 years at North Kensington has been roughly constant at  $1.6 \pm 0.1$ .

There are three possible general explanations for the change in slope at these two sites, if it is real. (1) The method used for Black Carbon measurement has changed such that reported concentrations are now relatively lower than they used to be; (2) the method used for Elemental Carbon measurement has changed such that reported concentrations are now relatively higher than they used to be; and (3) the nature of the soot-like pollution has changed in a way that causes larger differences between the methods.

The Black Carbon method has not changed in any significant way over the years. The OCEC Sunset analyser was upgraded in early 2014 with a new optical system with a more powerful laser. This should have improved the determination of the OC / EC split point and improved the quality of the EC results. This seems to be apparent from the reduced scatter in the Marylebone Road data in 2014 and 2015 compared to 2013, while the slope there did not change significantly, and this upgrade would not explain the increases in slope observed before the upgrade took place. The changing slopes may simply reflect the uncertainties in regression lines when the observed amount of scatter is present.

### 5.4.2 Polycyclic Aromatic Hydrocarbons (PAH)

Monthly concentrations of benzo[a]pyrene are measured at Auchencorth Moss, Ballymena, Birmingham Tyburn Background, Dunmurry Kilmakee, Glasgow Townhead, Harwell and Marylebone Road under the UK PAH Network<sup>10</sup>. To illustrate that BaP and the UV component have similar emission sources, the monthly average concentrations at Ballymena and Dunmurry of both measurements are displayed in Figure 34.



Note: The units are different for the two quantities. The PAH measurements are presented in  $\text{ng.m}^{-3}$  while the UV component is presented as  $\mu\text{g.m}^{-3}$ , however it can be seen that there is good temporal agreement between the two species due to similar emission sources such as solid fuel burning.

**Figure 34 UV component and Benzo[a]pyrene concentrations measured at Ballymena and Dunmurry in 2015**

The shaded area on the y-axis 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. As the PAH measurements are a monthly average there is no spread in the

<sup>10</sup> NPL 2015 Annual Report for UK PAH Network.

result over the month and therefore no uncertainty displayed in the y-value. The shaded area on the x-axis is for display purposes only, to allow the uncertainty in the mean value to be seen more clearly.

A recent paper exploring the relationship between collocated Aethalometer UV component measurements and Defra PAH Network BaP measurements, by R Brown *et al*<sup>11</sup>, determined the following quadratic relationships between the two pollutants.

$$BaP = a.UV^2 + b.UV + c \quad \text{Eqn 2}$$

Where

BaP = predicted BaP concentration in ng.m<sup>-3</sup>;

UV = measured UV component concentration in µg.m<sup>-3</sup>

Table 14 gives the coefficients a, b & c for the different site types.

| 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 2015 BaP concentration at each Aethalometer site based on the measured annual UV component concentration.

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11 R Brown, D Butterfield, S Goddard, D Hussain, P Quincey & G Fuller, Wavelength dependent light absorption as a cost effective, real-time surrogate for ambient concentrations of polycyclic aromatic hydrocarbons, Atmospheric Environment 127 (2016) 125-132.

| Site                 | Class | UV component,<br>$\mu\text{g.m}^{-3}$ | Predicted BaP,<br>$\text{ng.m}^{-3}$ | Measured<br>BaP    |
|----------------------|-------|---------------------------------------|--------------------------------------|--------------------|
|                      |       |                                       | site specific                        | $\text{ng.m}^{-3}$ |
| Auchencorth Moss     | R     | 0.05                                  | 0.0                                  | 0.0                |
| Ballymena            | NI    | 0.57                                  | 0.6                                  | 0.6                |
| Belfast Centre       | NI    | 0.31                                  | 0.3                                  |                    |
| Birmingham Tyburn BK | UR    | 0.18                                  | 0.1                                  | 0.2                |
| Birmingham Tyburn RS | UR    | 0.24                                  | 0.2                                  |                    |
| Cardiff Centre       | UR    | 0.21                                  | 0.1                                  |                    |
| Detling              | R     | 0.15                                  | 0.1                                  |                    |
| Dunmurry Kilmakee    | NI    | 0.34                                  | 0.3                                  | 0.4                |
| Glasgow High Street  | UR    | 0.17                                  | 0.1                                  |                    |
| Glasgow Townhead     | UR    | 0.12                                  | 0.0                                  | 0.1                |
| Harwell              | R     | 0.12                                  | 0.0                                  | 0.1                |
| Marylebone Road      | MY    | 0.31                                  | 0.4                                  | 0.2                |
| North Ken            | UR    | 0.25                                  | 0.2                                  |                    |
| Strabane             | NI    | 0.92                                  | 1.1                                  |                    |

**Table 15 Predicted 2015 annual BaP concentration based on measured UV component concentrations**

It can be seen that the predicted is within  $0.1\text{ng.m}^{-3}$  or better at all of the sites where BaP is measured. As the 2015 Black Carbon and BaP data were not included in the dataset to calculate the a, b & c coefficients in Eqn 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.

| Year | UV component concentration | Predicted BaP concentration $\text{ng.m}^{-3}$ |
|------|----------------------------|--|
| 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  |

**Table 16 Predicted BaP concentrations from UV component concentration at Strabane for the period 2009 to 2015.**

Six out of the last seven years have had predicted BaP concentrations above the  $1.0\text{ng.m}^{-3}$  target value in the EC Directive 2004/107/EC<sup>12</sup> relating to ambient BaP concentrations. The average concentration over the last 7 years is predicted to be  $1.3\text{ng.m}^{-3}$ .

<sup>12</sup> 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.

A detailed analysis of PAH concentrations in Northern Ireland is contained in an NPL Report for the Department for the Environment Northern Ireland<sup>13</sup>.

### 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. Two different types of instruments provide particulate mass concentrations across the Black Carbon Network: TEOM FDMS Model CB drier, and MetOne BAM – reference equivalent. As different automatic methods provide different results, the concentrations reported by these automatic instruments have been separated into instrument type and the results shown in Table 17.

| Site                            | BC<br>$\mu\text{g.m}^{-3}$ | TEOM<br>FDMS<br>PM <sub>10</sub><br>$\mu\text{g.m}^{-3}$ | TEOM<br>FDMS<br>PM <sub>2.5</sub><br>$\mu\text{g.m}^{-3}$ | MetOne<br>BAM<br>PM <sub>10</sub><br>$\mu\text{g.m}^{-3}$ | Percent BC<br>PM <sub>10</sub><br>% | Percent BC<br>PM <sub>2.5</sub><br>% |
|---------------------------------|----------------------------|--|---|---|-------------------------------------|--------------------------------------|
| Marylebone Road                 | 5.1                        | 24   | 16  |   | 21                                  | 32                                   |
| Birmingham Tyburn<br>Roadside   | 2.1                        | 17   | 12  |   | 12                                  | 18                                   |
| Glasgow High Street             | 1.4                        | 16   | 9   |   | 9                                   | 16                                   |
| Birmingham Tyburn<br>Background | 1.1                        | N/A  | N/A   |   | N/A                                 | N/A                                  |
| Belfast Centre                  | 1.1                        | 16   | 10  |   | 7                                   | 11                                   |
| North Kensington                | 1.0                        | 20   | 11  |   | 5                                   | 9                                    |
| Glasgow Townhead                | 0.9                        | N/A  | 7   |   | N/A                                 | 13                                   |
| Harwell                         | 0.3                        | 15   | 9   |   | 2                                   | 3                                    |
| Auchencorth Moss                | 0.2                        | 7  | 9   |   | 3                                   | 2                                    |
| Detling*                        | 0.4                        | 19   |   |   | 21                                  |                                      |
| Strabane*                       | 1.2                        |  |   | 12  | 10                                  |                                      |

Note: \* Local Authority run site; may not have identical QAQC procedures to AURN datasets. MetOne data is taken from the Air Quality Northern Ireland web site where it is stated to be Reference Equivalent.

Grey shaded cells indicate no measurements were made.

**Table 17 Comparison of Annual Black Carbon and Particulate Mass Concentrations**

Data capture for the Glasgow Townhead PM<sub>10</sub> TEOM-FDMS and Birmingham Tyburn Urban Background's PM<sub>10</sub> and PM<sub>2.5</sub> TEOM-FDMS systems were below the 75% required for a valid annual average. 2015 data capture for both of these 3 analysers was 44%, 62% & 61%, respectively.

It can be seen that the PM<sub>10</sub> and PM<sub>2.5</sub> mass concentration measured at Marylebone Road, Birmingham Tyburn roadside and Glasgow High Street sites have a much higher percentage of Black Carbon than the other sites. Black Carbon represents a large proportion of the total particulate mass

13 D Butterfield, R Brown, NPL REPORT AS66, Polycyclic Aromatic Hydrocarbons in Northern Ireland, February 2012.

at sites influenced by road traffic emissions. Any substantial significant reduction in Black Carbon emissions from road traffic should therefore lead to a measurable reduction in PM<sub>10</sub> and PM<sub>2.5</sub> mass concentrations as found by Font<sup>4</sup> *et al* (Figure 16).

## 5.5 TRENDS

Short and long-term trends in Black Carbon and UV component concentrations are given below.

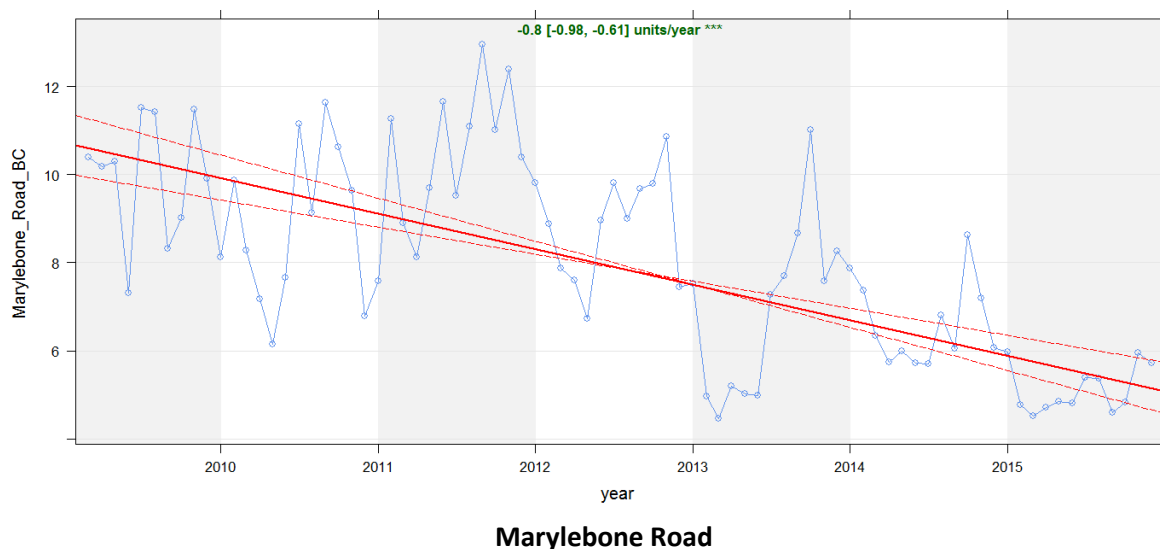
### 5.5.1 Short-Term Trends by Site

Figures 35 to 38 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 2015. The Theil-Sen method in OpenAir<sup>4,5</sup> was used to calculate the regression parameters including slope and uncertainty in the slope.

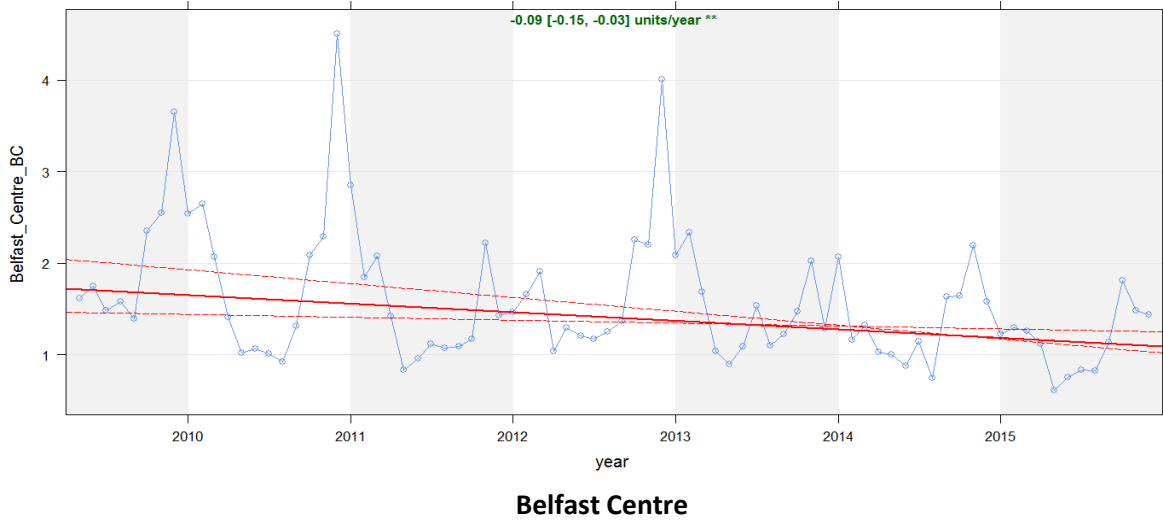
The Theil-Sen method chooses the median slope among all lines through pairs of two-dimensional sample points. The Theil-Sen estimator tends to yield accurate confidence intervals even with non-normal data and heteroscedasticity (non-constant error variance). It is also resistant to outliers.

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

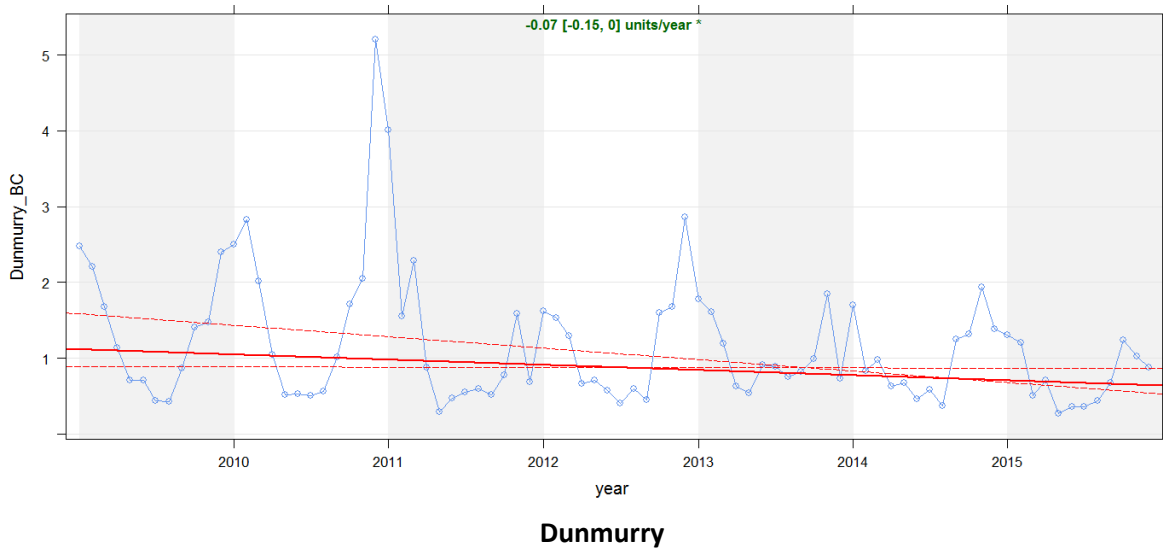
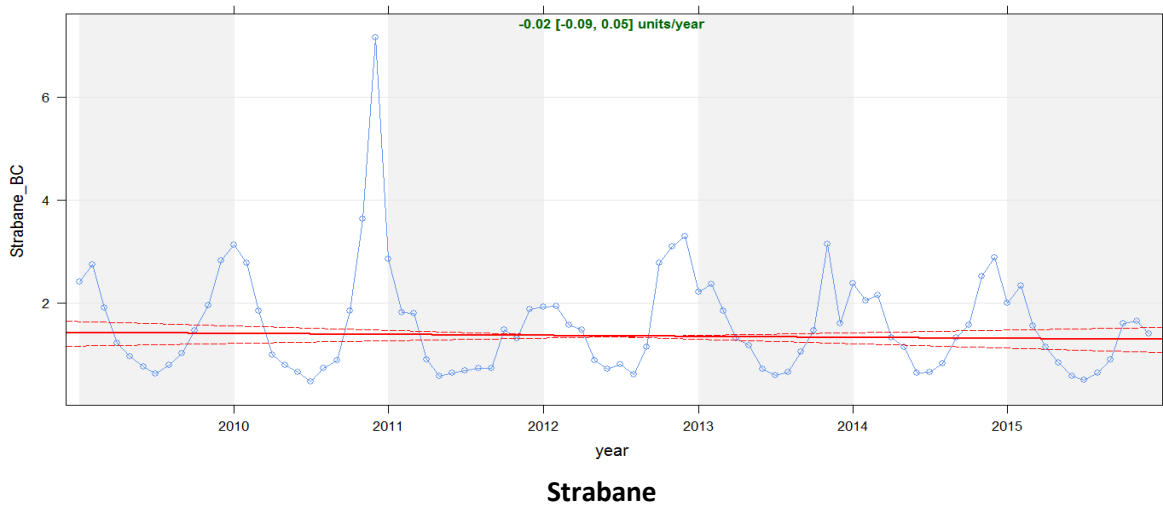
#### 5.5.1.1 Black Carbon

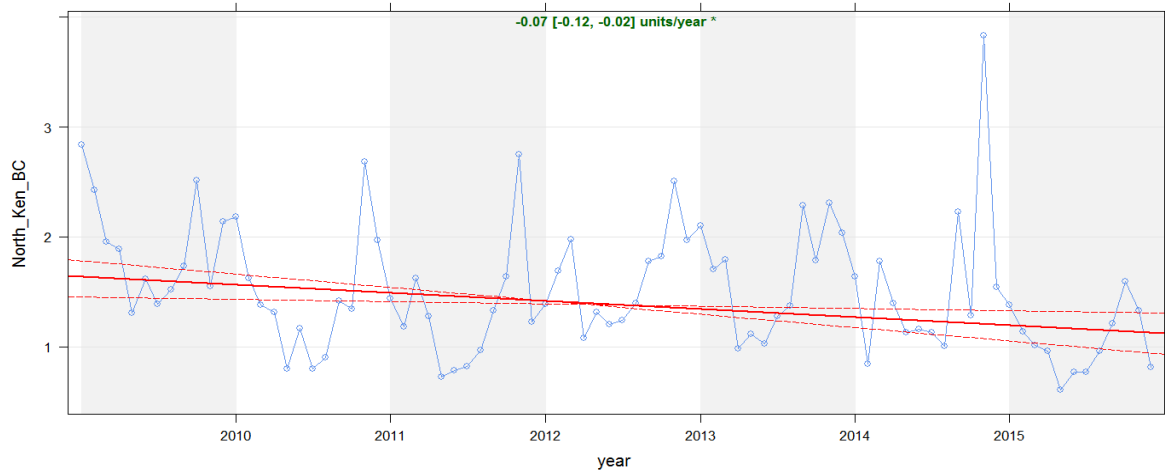


**Figure 35** Black Carbon concentrations measured at roadside sites, 2009 – 2015

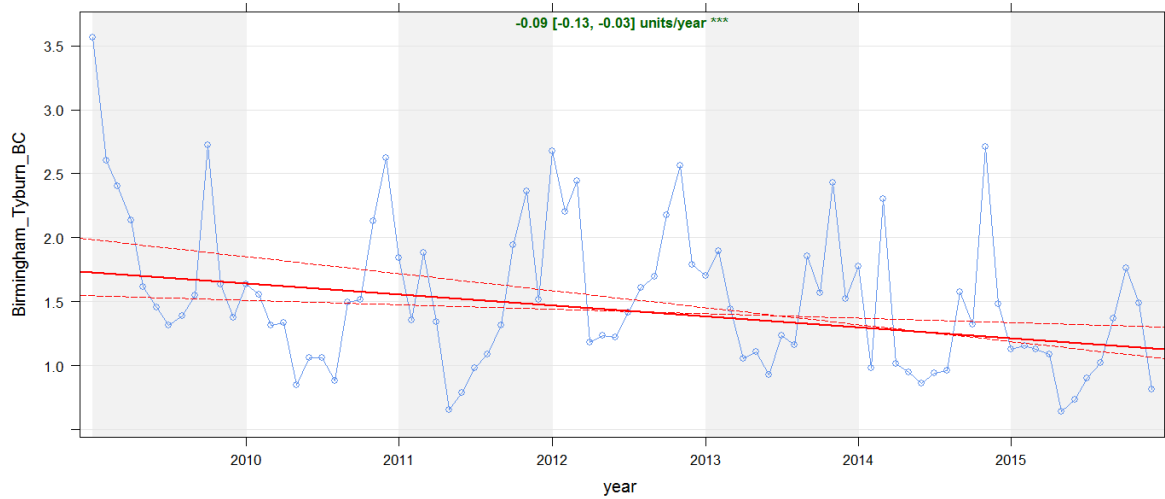


**Figure 36** Black Carbon concentrations measured at urban centre sites, 2009 – 2015



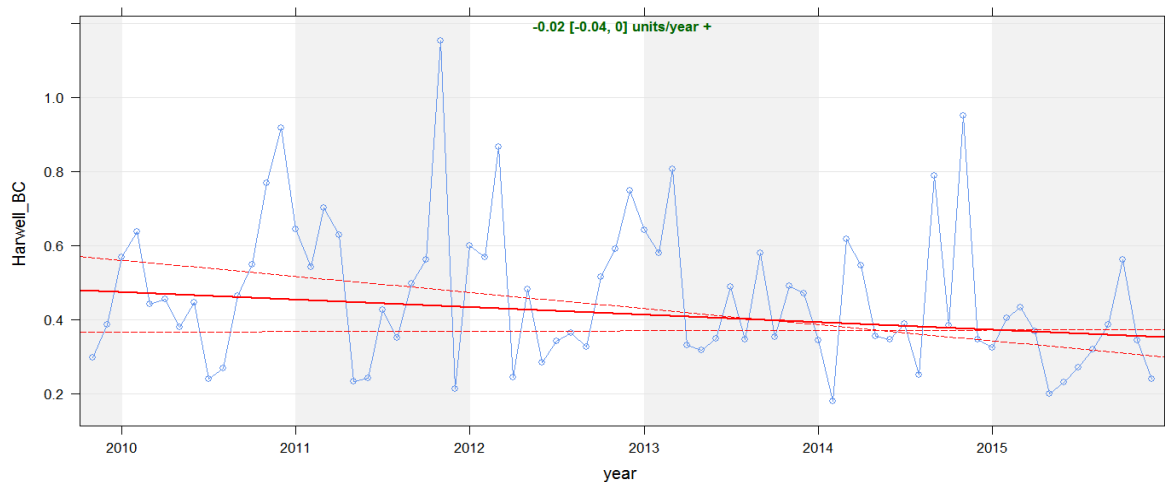


**North Kensington**



**Birmingham Tyburn**

**Figure 37 Black Carbon concentrations measured at urban background sites, 2009 – 2015**



**Harwell**

**Figure 38 Black Carbon concentrations measured at rural background sites, 2009 – 2015**



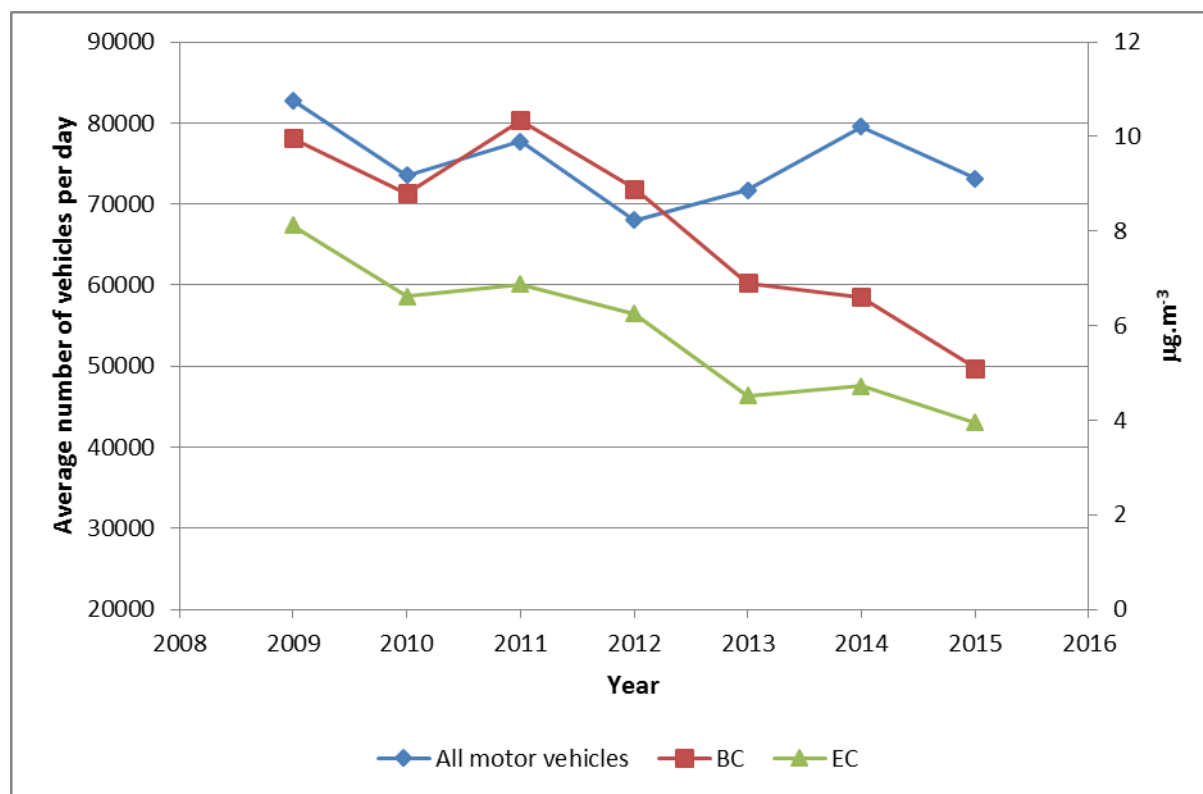
Table 18 summarises the slopes at each site.

| Site                    | Slope<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Lower limit<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Upper limit<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Slope<br>significant |
|-------------------------|--|--|--|----------------------|
| <b>Roadside</b>         |  |  |  |                      |
| Marylebone Road         | -0.80                                    | -0.98  | -0.61  | Y                    |
| <b>Urban Centre</b>     |  |  |  |                      |
| Belfast Centre          | -0.09                                    | -0.15  | -0.03  | Y                    |
| <b>Urban Background</b> |  |  |  |                      |
| Birmingham Tyburn       | -0.09                                    | -0.13  | -0.03  | Y                    |
| North Kensington        | -0.07                                    | -0.12  | -0.02  | Y                    |
| Strabane                | -0.02                                    | -0.09  | 0.05   | N                    |
| Dunmurry                | -0.07                                    | -0.15  | 0.00   | N                    |
| <b>Rural</b>            |  |  |  |                      |
| Harwell                 | -0.02                                    | -0.04  | 0.00   | N                    |

**Table 18 Summary of Black Carbon trends**

Over the period 2009 to 2015 Marylebone Road, Belfast Centre, Birmingham Tyburn and North Kensington have shown significant downward trend in Black Carbon concentrations. At the three non-roadsite sites this trend is likely to be due to significantly lower Black Carbon concentrations measured in 2015. This is probably due to meteorological conditions with the last quarter being significantly wetter and warmer than in previous years. 2015 was the sixth wettest year since 1910, or the introduction of Euro 6/VI vehicles into the fleet.

However, Marylebone Road has been showing reduced black carbon concentrations year on year since 2011, with 2015 annual mean concentration roughly half that of 2011. Similarly the concentrations of Elemental Carbon made at Marylebone Road have also followed the trend in Black Carbon concentrations. Figure 39 shows the annual Black Carbon and Elemental Carbon concentrations along with the average daily traffic flow past the site.



**Figure 39 Annual Average Black Carbon, Elemental Carbon and Motor Vehicles per Day at Marylebone Road for the period 2009 – 2015**

It can be seen that the changes in Black Carbon and Elemental Carbon concentrations follow changes in the total traffic flow for the years 2009 to 2012 & 2015, but not for 2013 to 2014, which would indicate that Black Carbon emissions per vehicle have changed over the last 3 years. The drop in emissions per vehicle type may be linked to the increased proportion of low emission buses (hybrid and fuel cell / hybrid) in the London bus fleet<sup>14</sup>. Table 19 shows the composition of the London bus fleet over the period 2010 to 2015. The bottom row of the table shows the percentage of low emission buses, which is a combination of the hybrid 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.

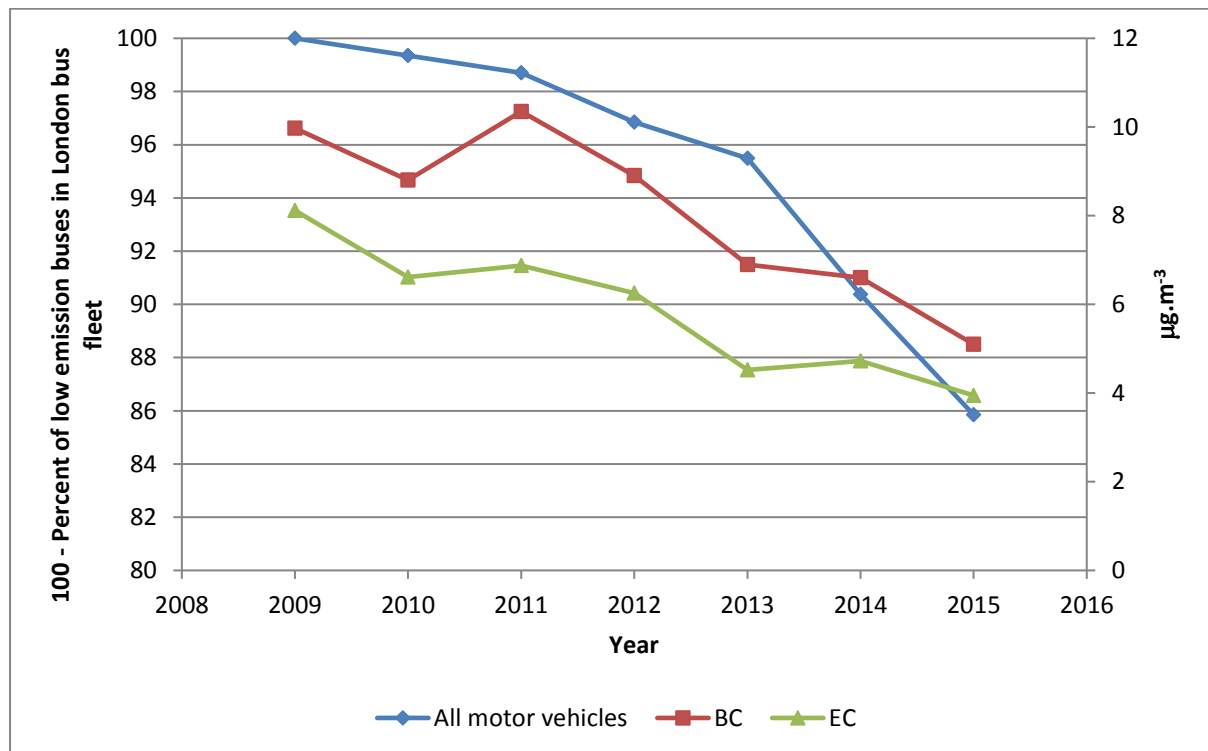
Also in 2012 the vehicles types affected by the London Low Emission Zone (LEZ) was increased to include: large vans, minibuses and other specialist diesel vehicles. Vehicles entering the LEZ must be Euro III or higher to be compliant with the requirements. In addition the requirements for Lorries, buses, 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.

14 Number of Buses by Type of Bus in London, tfl-buses-type.xls, London Datastore, <https://londondatastore-upload.s3.amazonaws.com/tfl-buses-type.xls>

| Bus Type        | Drive train type | Number of buses |       |       |       |       |       |
|-----------------|------------------|-----------------|-------|-------|-------|-------|-------|
|                 |                  | 2010            | 2011  | 2012  | 2013  | 2014  | 2015  |
| New Routemaster | Hybrid           | 0               | 0     | 5     | 8     | 168   | 432   |
| Routemaster     | Diesel           | 18              | 18    | 19    | 20    | 19    | 19    |
| Artic           | Diesel           | 320             | 260   | 0     | 0     | 0     | 0     |
| Single deck     | Diesel           | 2,676           | 2,670 | 2,661 | 2,608 | 2,606 | 2,662 |
|                 | Fuel Cell/Hybrid | 0               | 5     | 5     | 5     | 8     | 8     |
|                 | Hybrid           | 27              | 27    | 33    | 28    | 23    | 23    |
|                 | Electric         | 0               | 0     | 0     | 0     | 2     | 8     |
| Double deck     | Diesel           | 5,554           | 5,487 | 5,787 | 5,696 | 5,296 | 5,026 |
|                 | Hybrid           | 29              | 79    | 233   | 352   | 643   | 799   |
| TOTAL           |                  | 8,624           | 8,546 | 8,743 | 8,717 | 8,765 | 8977  |
| % low emission  |                  | 0.65            | 1.30  | 3.16  | 4.51  | 9.63  | 14.15 |

**Table 19 Composition of London bus fleet, 2010 to 2015<sup>[14]</sup>**

Figure 40 is Figure 39 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].



**Figure 40 Annual Average Black Carbon, Elemental Carbon and 100 – percentage of low emission buses in the London bus fleet for the period 2009 – 2015**

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.

5.5.1.2 UV Component

Figures 41 to 44 show the UV component trends.

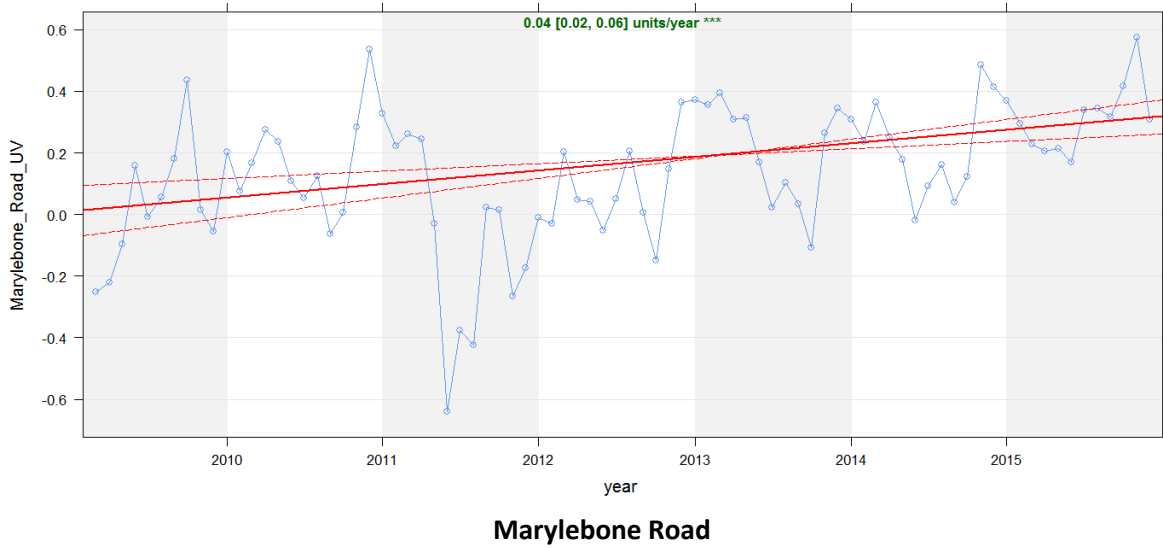


Figure 41 UV component concentrations measured at roadside sites, 2009 – 2015

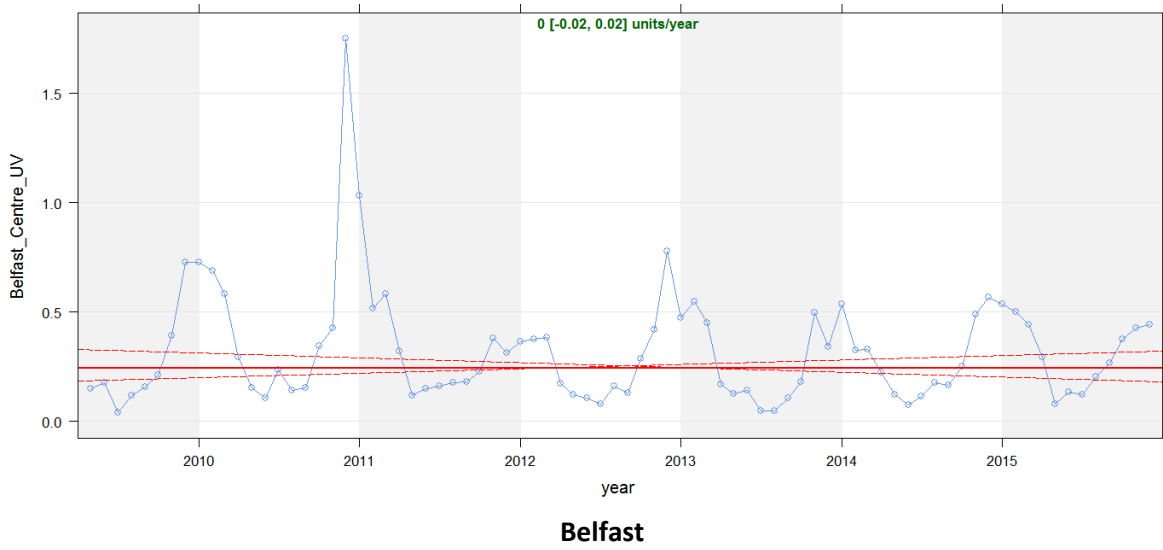
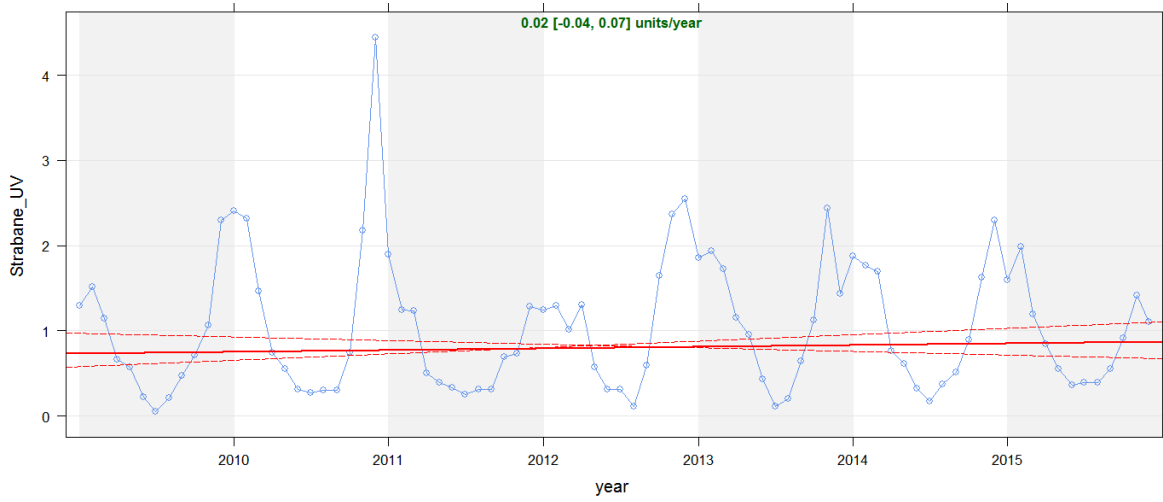
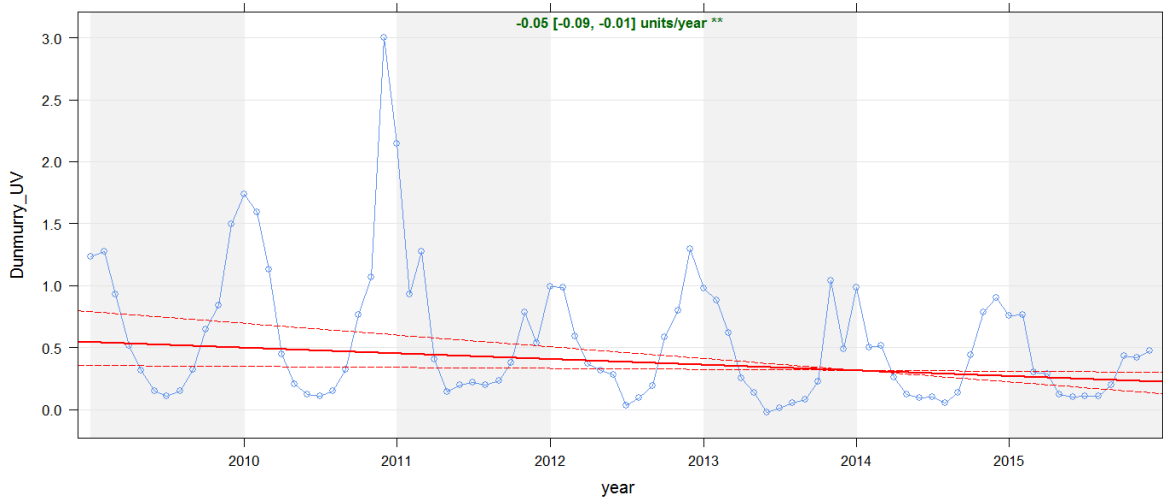


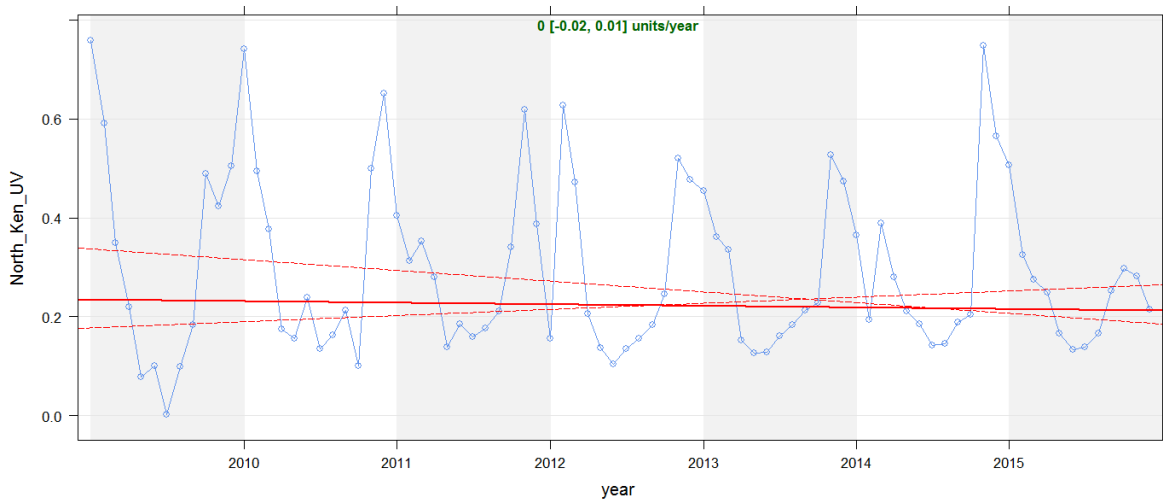
Figure 42 UV component concentrations measured at urban centre sites, 2009 – 2015



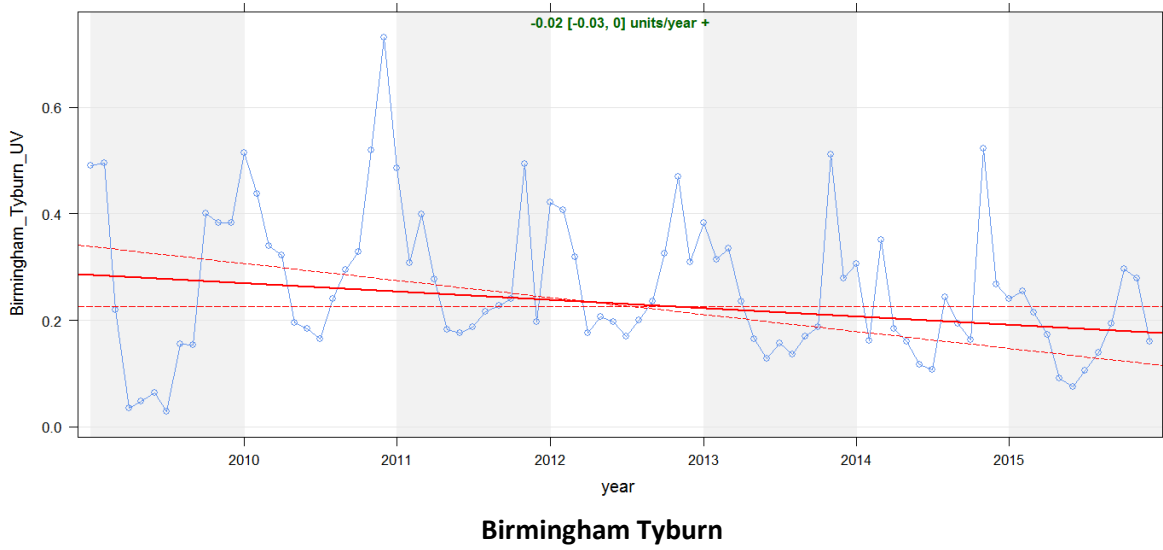
**Strabane**



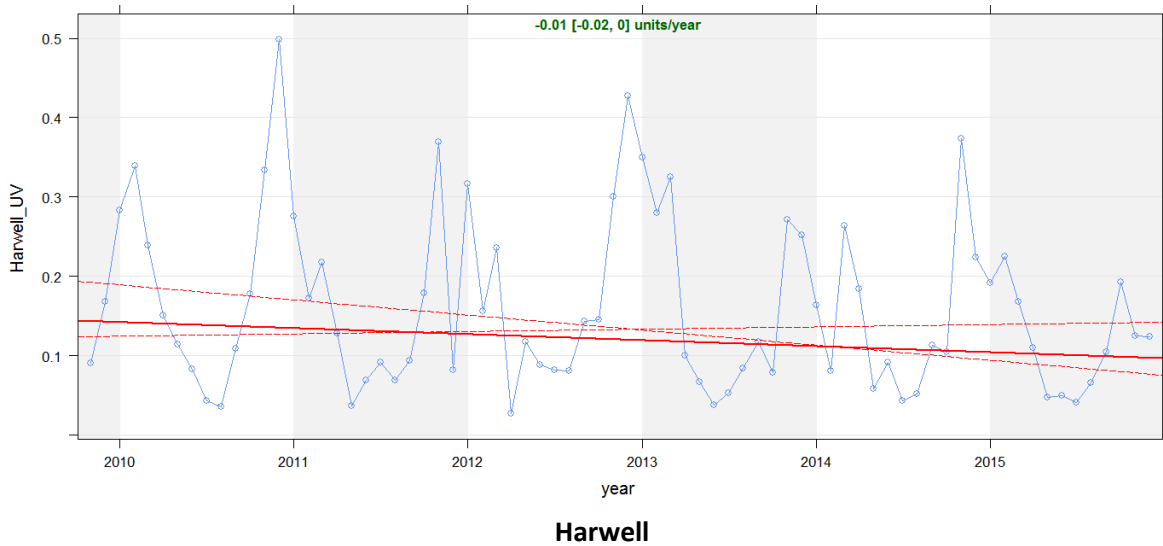
**Dunmurry**



**North Kensington**



**Figure 43** UV component concentrations measured at urban background sites, 2009 – 2015



**Figure 44** UV component concentrations measured at rural background sites, 2009 – 2015

Table 20 summarises the slopes at each site.

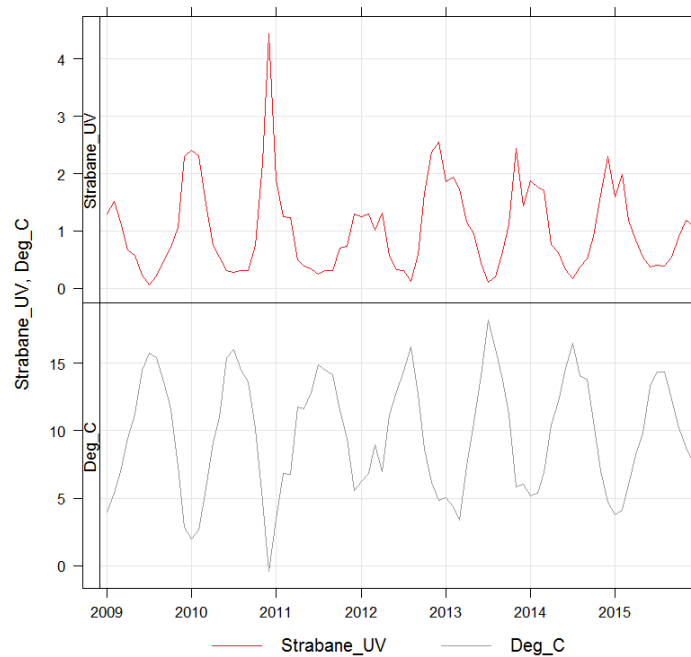
| Site                    | Slope<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Lower limit<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Upper limit<br>$\mu\text{g}\cdot\text{m}^{-3}$ | Slope<br>significant |
|-------------------------|--|--|--|----------------------|
| <b>Roadside</b>         |  |  |  |                      |
| Marylebone Road         | 0.04                                     | 0.02   | 0.06   | Y                    |
| <b>Urban Centre</b>     |  |  |  |                      |
| Belfast Centre          | 0.00                                     | -0.02  | 0.02   | N                    |
| <b>Urban Background</b> |  |  |  |                      |
| Dunmurry                | -0.05                                    | -0.09  | -0.01  | Y                    |
| Strabane                | 0.02                                     | -0.04  | 0.07   | N                    |
| North Kensington        | 0.00                                     | -0.02  | 0.01   | N                    |
| Birmingham Tyburn       | -0.02                                    | -0.03  | 0.00   | N                    |
| <b>Rural</b>            |  |  |  |                      |
| Harwell                 | -0.01                                    | -0.02  | 0.00   | N                    |

**Table 20 Summary of UV component trends**

The Marylebone Road UV component concentration shows a significant upward trend over the period 2009 to 2015, this is probably due to the reduced Black Carbon concentrations over the last four years. The Aethalometer measures the UV component by the difference between the BC and UV channel. As Black Carbon has fallen it has become easier for the Aethalometer to determine the small UV component. It is unlikely that the UV component emissions across London have risen in the last two years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which is not dominated by traffic, have fallen. This trend should be treated with caution due to the low concentrations involved.

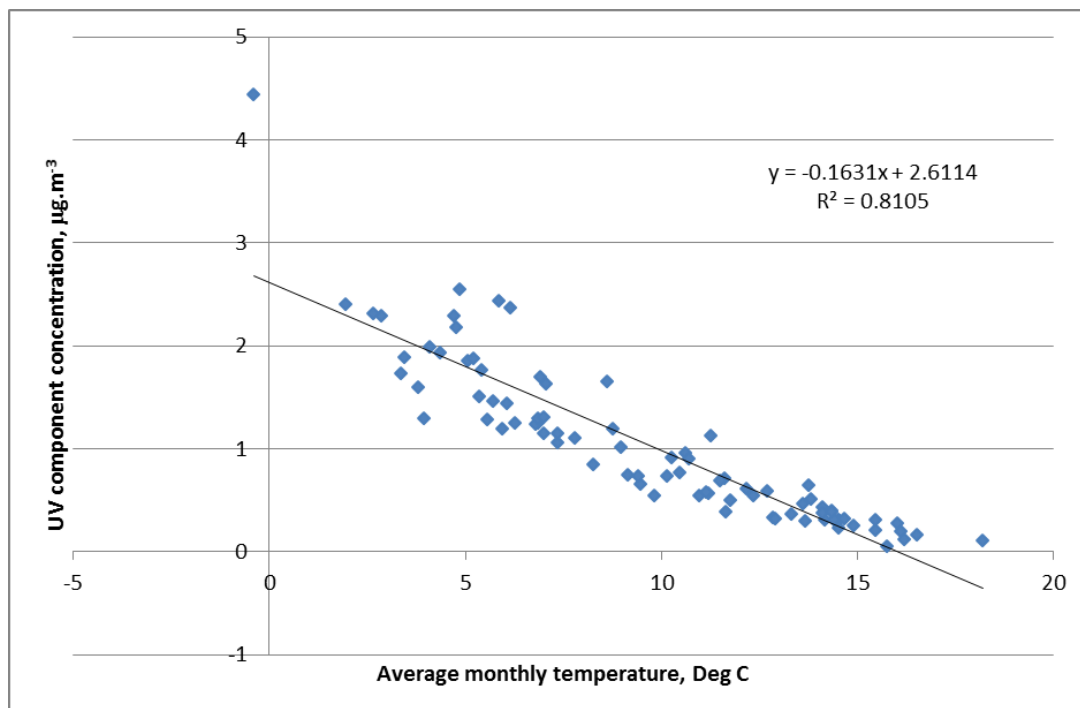
There is also a significant downward trend in the Dunmurry data which is most likely to be caused by the warmer and wetter 2015 weather than by a significant change in emission sources.

To show how pollutant concentrations can depend strongly on the weather, the 2009-2015 UV component concentrations at Strabane, which are strongly affected by domestic solid fuel use, are plotted in Figure 45, along with average temperature for same period. Temperature measurements from Armagh have been used as this is the nearest Met Office site with a long time series.



**Figure 45** Strabane monthly UV component concentration and average ambient temperature for 2009 – 2015

It can be seen that the UV component concentration is inversely related to the average ambient temperature. This is due to the main source of UV component emissions being local domestic heating in Strabane. This is evident in both the winter and the summer indicating that there are still solid fuel emissions in the summertime. The relationship is shown in Figure 46 as a scatter plot.



**Figure 46** Scatter plot of monthly UV component concentration versus ambient temperature at Strabane over the period 2009 -2015



It can be seen that there is 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. It can also be seen that there is a significant UV source when temperatures are below 15°C, linking the UV component to fuels used for domestic heating systems. There is only 1 month where the monthly UV component concentration is less than the detection limit of 0.1 µg.m<sup>-3</sup>, indicating that there is a UV component emission source all year.

### 5.5.2 Short-Term Trends over the Network as a Whole

Figures 47 and 48 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, Marylebone Road, North Kensington and Strabane). The median concentration is shown to remove the influence of large changes in a single site that would skew the overall result for the Network.

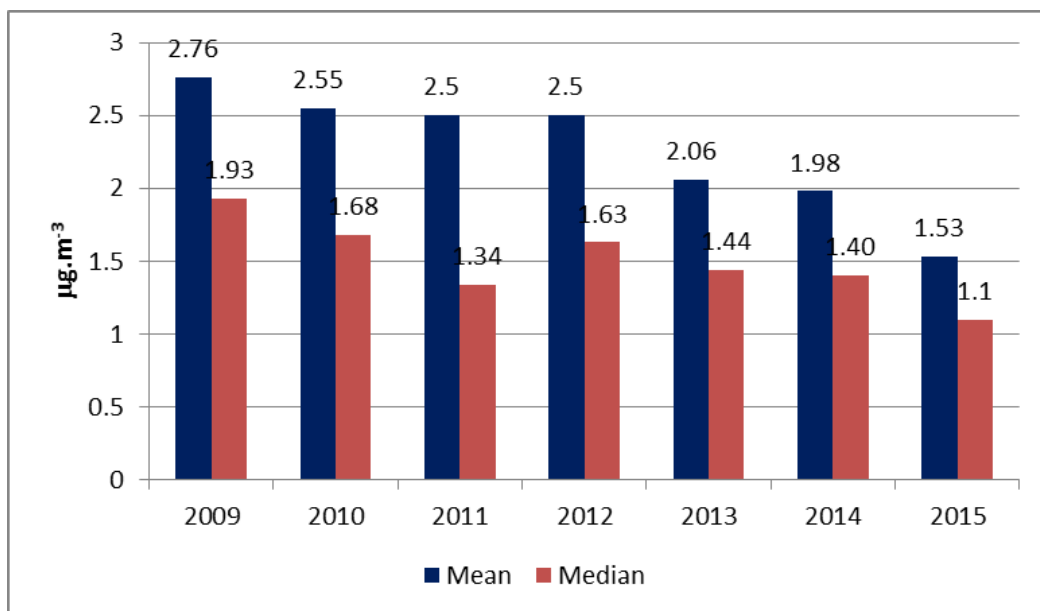
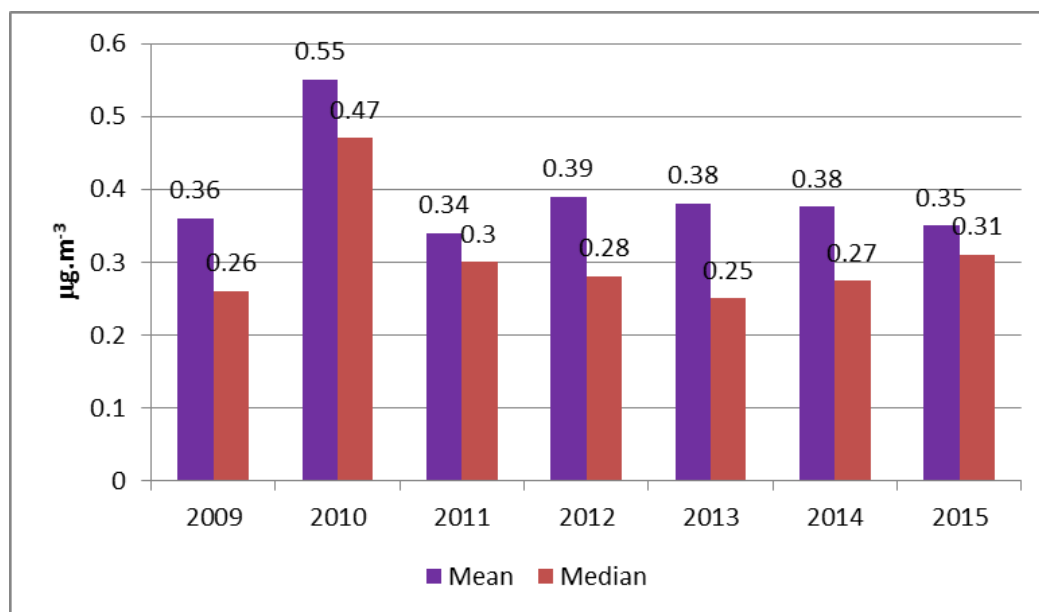


Figure 47 Annual average Black Carbon concentrations for long-term sites



**Figure 48 Annual average UV component concentrations for long-term sites**

It can be seen that there has been a considerable drop in the Network Annual mean for Black Carbon in 2013 & 2015, with a small drop in 2014, while the median concentration has remained roughly similar over the last 5 years ( $1.35 \mu\text{g.m}^{-3} \pm 0.25 \mu\text{g.m}^{-3}$ ). The drop in average concentration is mainly driven by the drop in concentrations at Marylebone Road. The drop in median concentrations in 2015 is due to the warmer and wetter weather.

Both the annual average and median UV component concentrations vary from year to year with maximum concentrations 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 45, so elevated concentrations are probably due to domestic emissions.

## 6.0 EUROPEAN STANDARDISATION

The European standardisation body CEN is formulating a European Standard on the measurement of Elemental Carbon and Organic Carbon deposited on filters. Technical Report EN/TR 16243 has been published<sup>15</sup>. The standardisation working group have completed validation work on the procedures and requirements laid out in this technical report. The work packages performed were:

- WP1: Literature review
- WP2: Lab tests
- WP3: Field tests
- WP4: Statistical evaluation

The current progress is as follows:

<sup>15</sup> CEN/TR 16243: Ambient air quality - Guide for the measurement of elemental carbon (EC) and organic carbon (OC) deposited on filters, August 2011.

WP1 The final draft of the literature review has been published as an internal CEN report and has been converted into a scientific paper for publication. The paper is currently in open for interactive public discussion in Atmospheric Measurement Techniques<sup>16</sup>.

WP2 Two inter-laboratory comparisons have been performed. The first demonstrated the requirement to perform regular instrument calibrations to ensure satisfactory performance from the thermal / optical method. The second intercomparison showed good agreement between the 4 laboratories.

WP3 Sampling has been performed at 6 field locations along with parallel sampling using automatic analysers. The automatic analysers being used are the Magee AE33 dual spot, 7-wavelength Aethalometer, and a Thermo Scientific Multi Angle Absorption Photometer (MAAP). The AE33 differs from the AE22 Aethalometer used on the Network in 2 ways. Firstly the AE22 is a 2 wavelength analyser, while the AE33 uses 7 wavelengths. Secondly the AE33 does an internal correction for tape loading, while for the AE22 this has to be performed post-sampling in the ratification process. Part of the remit of the working group is to comment on the comparability between the proposed reference method and automatic analyser methods.

WP4 The first interim report on the working group's progress has been approved by the working group and sent to CEN. The statistical analysis of the field data has been completed and reported to CEN.

EUSAAR2 thermal protocol with transmission OCEC split point determination has been adopted as the standard analysis protocol for OCEC PM<sub>2.5</sub> particulate matter collected on filters. Currently the Network uses the NIOSH transmission protocol. In 2012 NPL analysed filters using both the EUSAAR2 transmission and NIOSH transmission protocols<sup>17</sup> and found that no significant difference could be identified in TC – a reassuring check of the analysis system. As expected, NIOSH gives somewhat lower EC values than EUSAAR II (with correspondingly higher OC values). These differences are of the order of 20% for EC and 5% for OC.

Protocols that have a lower maximum temperature during the inert-gas heating phase, such as the EUSAAR II protocol with a maximum of 650°C, tend to record significantly higher EC values than protocols such as NIOSH, with a maximum of 870°C, with correspondingly less OC, such that the sum of EC and OC is the same in both cases. More charring of organic material will occur in the NIOSH case, which may be inadequately accounted for by the optical correction. Alternatively, the EUSAAR II protocol may not be removing all material that should be classified as OC. Ultimately the difference is because EC and OC are not objectively defined, but rather defined by the method used.

The final draft of the proposed standard was finalised in May 2015 and sent for CEN enquiry. Suggested changes to the standard were received and have been implemented. The revised standard has been submitted to CEN for formal vote and subsequent publication. Publication is due in late 2016.

NPL will apply the EUSAAR2 transmission protocol for all filters exposed from 1<sup>st</sup> January 2016 onwards.

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16 Karanasiou, A., Minguillón, M. C., Viana, M., Alastuey, A., Putaud, J.-P., Maenhaut, W., Panteliadis, P., Močnik, G., Favez, O., and Kuhlbusch, T. A. J.: Thermal-optical analysis for the measurement of elemental carbon (EC) and organic carbon (OC) in ambient air a literature review, *Atmos. Meas. Tech. Discuss.*, 8, 9649-9712, doi:10.5194/amtd-8-9649-2015, 2015.

17 Airborne Particulate Concentrations and Numbers in the United Kingdom (phase 3) Annual report 2012, NPL Report AS 83, December 2013.

## 7.0 CONCLUSIONS

Black Carbon concentrations measured at most sites in 2015 were similar to those in previous years, with the 2013 & 2014 drop in Marylebone Road concentrations maintained through 2015. In general Black Carbon concentrations in 2015 were lower than 2014 for all sites, the Network means for these years were  $1.60 \mu\text{g.m}^{-3}$  and  $1.26 \mu\text{g.m}^{-3}$ , respectively. UV component concentrations in 2015 were very similar to previous years. From looking at the trend in Black Carbon concentrations for only those sites that have been open from 2009 – 2015, the concentration was stable from 2009 to 2012, with a significant drop in average concentration from  $2.51 \mu\text{g.m}^{-3}$  in 2012 to  $2.06 \mu\text{g.m}^{-3}$  in 2013 (22%),  $1.98 \mu\text{g.m}^{-3}$  in 2014 and  $1.53 \mu\text{g.m}^{-3}$  (23%) in 2015, while the median concentration has dropped from  $1.63 \mu\text{g.m}^{-3}$  to  $1.43 \mu\text{g.m}^{-3}$  in 2013 (12%),  $1.40 \mu\text{g.m}^{-3}$  in 2014 and  $1.1 \mu\text{g.m}^{-3}$  (21%) in 2015. The median concentration is much less susceptible to changes at one site. The drop in 2015 is due to the reduction at Marylebone Road and the wetter and warmer weather in the last quarter of 2015. A similar drop in Elemental Carbon concentrations is also seen at Marylebone Road. The 2015 UV component concentration was  $0.3 \mu\text{g.m}^{-3}$ , which has stayed roughly constant between 2009 and 2015 with the exception of 2010 ( $0.4 \mu\text{g.m}^{-3}$ ). 2010 was an exceptionally cold year, especially in Northern Ireland. The range of concentrations across roadside and rural background sites were also similar.

Over the period 2009 to 2015, Marylebone Road, Belfast Centre, Birmingham Tyburn and North Kensington have shown a significant downward trend in Black Carbon concentrations. At the three non-roadside sites this trend is likely to be influenced by meteorological conditions significantly lowering Black Carbon concentrations measured in 2015; with the last quarter being significantly wetter and warmer than in previous years. 2015 was the sixth wettest year since 1910. However, Marylebone Road has been showing reduced Black Carbon concentrations year on year since 2011, with 2015 annual mean concentration roughly half that of 2011. This drop in concentration is 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, HGCs, lorries and vans (Euro III to Euro IV). Low emission buses now make up 14% of the fleet.

The Marylebone Road UV component concentration shows a significant upward trend over the period 2009 to 2015. This trend should be treated with caution due to the low concentrations involved. It is probably related to the reduced Black Carbon concentrations over the last 4 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 2 years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which is not dominated by traffic, have fallen. There is also a significant downward trend in the Dunmurry data which is most likely to be caused by the warmer and wetter 2015 weather than by a significant change in emission sources.

The new 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. The urban increment for Black Carbon was similar for all locations while the roadside increment was roughly proportional to road traffic volumes. Reductions in the roadside increment for Black Carbon have fallen in line with reductions in roadside increment for  $\text{PM}_{2.5}$  concentrations indicating similar emission sources. There was no significant urban or roadside increment in UV component concentration.

Diurnal average concentrations of Black Carbon show that the dominant emission sources are road traffic and domestic heating using non-smokeless fuel. The diurnal average concentrations of the UV component show that its main source is domestic heating with solid fuels with little influence from traffic.