

NPL REPORT ENV 21

2016 ANNUAL REPORT FOR THE UK BLACK CARBON NETWORK

I HESSEY D BUTTERFIELD S BECCACECI P QUINCEY B SWEENEY A LILLEY C BRADSHAW G FULLER D GREEN A FONT

JUNE 2017

2016 ANNUAL REPORT FOR THE UK BLACK CARBON NETWORK

I Hessey, D Butterfield, S Beccaceci, P Quincey, B Sweeney, A Lilley and C Bradshaw Chemical, Medical and Environmental Sciences Department, NPL

G Fuller, D Green and A Font Font Environmental Research Group, King's College London NPL Report ENV 21

© NPL Management Ltd, 2017

ISSN: 2059-6030

National Physical Laboratory Hampton Road, Teddington, Middlesex, TW11 0LW

Extracts from this report may be reproduced provided the source is acknowledged and the extract is not taken out of context.

Approved on behalf of NPLML by Richard Brown, Knowledge Leader

EXECUTIVE SUMMARY

This report covers the operation of the UK Black Carbon Network and the data collected by the Network in 2016. 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 site closed in June 2014. In March 2015 a new site at Cardiff Centre was installed and also a site at Glasgow High Street which allows urban and roadside increments to be calculated for London, Birmingham and Glasgow. During 2016 a new site opened at Chilbolton to replace Harwell, and Birmingham Tyburn Roadside was moved to Birmingham Tyburn Roadside and Birmingham Tyburn Roadside and Birmingham Tyburn Roadside was movel to Birmingham Tyburn Roadside and Birmingham Tyburn Roadside was movel to Birmingham Tyburn Roadside and Birmingham Tyburn Roadside was movel to Birmingham Tyburn Roadside and Birmingham Tyburn R

The 2016 data capture for Aethalometer measurements was 93%. This figure was strongly influenced by a single breakdown at Strabane. This is a very 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.

2016 urban annual mean Black Carbon concentrations on the Network (with the corresponding 2015 concentrations in brackets) ranged from 0.8 (0.8) μ g.m⁻³ at Dunmurry Kilmakee to 4.9 (5.1) μ g.m⁻³ at Marylebone Road. Values between 0.1 (0.2) μ g.m⁻³ and 0.5 (0.4) μ g.m⁻³ were seen at the rural background sites Chilbolton, Detling and Auchencorth Moss. The network mean for Black Carbon concentration was 1.3 (1.3) μ g.m⁻³.

The annual mean UV component concentrations ranged from 0.1 (0.1) μ g.m⁻³ at Auchencorth Moss to 0.5 (0.6) μ g.m⁻³ at Ballymena. Strabane had the highest UV component concentration and showed a mean of 1.6 (0.9) μ g.m⁻³, but this is skewed as the site only measured in the winter months during 2016. Roadside sites showed many negative spikes in the UV component concentration, thought to be measurement artefacts. These could be caused by volatile components in fresh vehicle exhaust plumes, the internal timing of the Aethalometer measurement or use of an assumed Ångström Coefficient. This effect was most prevalent at Marylebone Road. The network mean for UV component concentrations for 2015.

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

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

Comparisons between Black Carbon concentrations and Elemental Carbon concentrations showed good linear relationships between the measurements at North Kensington, Chilbolton and Marylebone Road, with R^2 values of 0.96, 0.89 and 0.95 respectively. For North Kensington, Chilbolton and Marylebone Road the slopes were 1.08, 1.31, & 1.25 with intercepts of +0.03, +0.03 & +0.26 μ g/m³respectively. 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 made up a significant proportion of the particulate mass concentration at roadside sites. At Marylebone Road the Black Carbon concentration comprised 19% of the PM_{10} concentration and 31% of the $PM_{2.5}$ concentration, while at Birmingham Tyburn roadside Black Carbon formed 12% and 18% of PM_{10} and $PM_{2.5}$ respectively. Glasgow High Street showed 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. None of the sites with a full year average were inferred to exceed the EU target value of 1.0 ng.m⁻³ for BaP based on this method. Strabane was inferred to exceed the target value, and although the 2016 mean was biased high because of the absence of data in the summer months, it would be expected to exceed as it has in 6 of the years between 2009-2015.

Monthly means of Black Carbon concentrations were examined over the period 2009 to 2016 to evaluate trends. Marylebone Road, Belfast Centre, Birmingham Tyburn, North Kensington and Dunmurry have shown a significant downward trend in Black Carbon concentrations. At the four non-roadside sites this trend was likely to have been influenced by a significantly wetter and warmer winter over 2015-2016 which lowered the Black Carbon concentrations in 2015. 2015 was the sixth wettest year since 1910. Although the average concentration increased slightly at these sites in 2016 it was not enough to affect the downward trend. However, Marylebone Road has been showing reduced Black Carbon concentrations year on year since 2011, with 2016 annual mean concentration less than half that of 2011. This drop in concentration was likely to be due to the increased number of low emission (hybrid) buses in the London bus fleet and stricter emission controls on London taxis, HGVs, lorries and vans (Euro III to Euro IV). Low emission buses now make up 19% of the fleet.

The Marylebone Road UV component concentration showed a significant upward trend over the period 2009 to 2016. 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 4 years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which were not dominated by traffic, have remained stable. There was also a significant downward trend in the Dunmurry data which was most likely to be caused by the warmer and wetter 2015-2016 winter than by a significant change in emission sources.

CEN (TC 264 WG 35) have formulated a standard for the measurement of Elemental Carbon and Organic Carbon deposited on filters which was published in March 2017. This CEN working group is now working to bring automatic Black Carbon analysis such as the Aethalometer within the standardisation process.

CONTENTS

1.0 INTRO	DUCTION1
1.1 GENE	RAL1
1.2 BLAC	CARBON
1.3 MEAS	UREMENT METHOD
1.3.1	Aethalometer instrument and data processing2
1.3.2	Sampling
2.0 NETWO	DRK INFRASTRUCTURE
2.1 NETV	/ORK SITES AND DESIGN
2.2 NETV	/ORK OPERATION
3.0 QUALI	TY ASSURANCE AND QUALITY CONTROL (QA/QC)9
3.1 SITE	UDITS
3.1.1	Sampler Leak Rate and Calibration of Sample Flow9
3.1.2	Instrument Performance10
4.0 MEASU	IREMENT UNCERTAINTY12
4.0 MEASU 4.1 SAMF	IREMENT UNCERTAINTY
4.0 MEASU 4.1 SAMF 4.2 MEAS	IREMENT UNCERTAINTY
 4.0 MEASU 4.1 SAMF 4.2 MEAS 4.3 CORR 	IREMENT UNCERTAINTY
 4.0 MEASU 4.1 SAMF 4.2 MEAS 4.3 CORR 4.4 INDIC 	JREMENT UNCERTAINTY 12 PLE VOLUME 12 SUREMENT OF ABSORPTION 12 ECTION FOR SPOT DARKENING 12 ATIVE OVERALL MEASUREMENT UNCERTAINTY 13
 4.0 MEASU 4.1 SAMF 4.2 MEAS 4.3 CORR 4.4 INDIC 5.0 RESULT 	JREMENT UNCERTAINTY12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13TS14
 4.0 MEASU 4.1 SAMF 4.2 MEAS 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 	JREMENT UNCERTAINTY12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13TS14SERIES14
 4.0 MEASU 4.1 SAME 4.2 MEAS 4.3 CORE 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 	JREMENT UNCERTAINTY 12 PLE VOLUME 12 SUREMENT OF ABSORPTION 12 ECTION FOR SPOT DARKENING 12 ATIVE OVERALL MEASUREMENT UNCERTAINTY 13 rs 14 SERIES 14 Black Carbon 14
 4.0 MEASU 4.1 SAME 4.2 MEAS 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 5.1.2 	JREMENT UNCERTAINTY12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13FS14SERIES14Black Carbon14UV component17
 4.0 MEASU 4.1 SAME 4.2 MEASU 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 5.1.2 5.2 AVER 	JREMENT UNCERTAINTY12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13TS14SERIES14Black Carbon14UV component17AGES AND DATA CAPTURE20
 4.0 MEASU 4.1 SAME 4.2 MEASU 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 5.1.2 5.2 AVER 5.2.1 	JREMENT UNCERTAINTY.12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13TS.14SERIES14Black Carbon14UV component17AGES AND DATA CAPTURE20Black Carbon21
 4.0 MEASU 4.1 SAME 4.2 MEASU 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 5.1.2 5.2 AVER 5.2.1 5.2.2 	JREMENT UNCERTAINTY.12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13TS.14SERIES.14Black Carbon14UV component17AGES AND DATA CAPTURE20Black Carbon21UV component21UV component25
 4.0 MEASU 4.1 SAME 4.2 MEASU 4.3 CORR 4.4 INDIC 5.0 RESULT 5.1 TIME 5.1.1 5.1.2 5.2 AVER 5.2.1 5.2.1 5.2.3 	JREMENT UNCERTAINTY12PLE VOLUME12SUREMENT OF ABSORPTION12ECTION FOR SPOT DARKENING12ATIVE OVERALL MEASUREMENT UNCERTAINTY13FS14SERIES14Black Carbon14UV component17AGES AND DATA CAPTURE20Black Carbon21UV component25Data Capture27

5.4 COMPAR	SISONS WITH OTHER POLLUTANTS	51
5.4.1 Ele	emental Carbon	51
5.4.2 Pc	olycyclic Aromatic Hydrocarbons (PAH)	55
5.4.3 Pa	articulate Mass Concentration	57
5.5 TRENDS		58
5.5.1 Sh	nort-Term Trends by Site	58
5.5.2 Sh	nort-Term Trends over the Network as a Whole	67
6.0 EUROPEAN	N STANDARDISATION	69
7.0 CONCLUSI	ONS	69

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 was 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 can 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.

The Cardiff site closed in June 2014. In March 2015 a new site at Cardiff Centre was installed and also a site at Glasgow High Street which now allows urban and roadside increments to be calculated for London, Birmingham and Glasgow. During 2016 a new site opened at Chilbolton to replace Harwell, and Birmingham Tyburn Roadside was moved to Birmingham Keeley Street in October 2016.

Therefore for the monitoring year 2016 there were 15 sites which recorded some data (however this is just due to one site being replaced mid year). The year started and concluded with 14 sites overall.

In January 2017 a new contract was issued by the Environment Agency to run the network, continuing with the same monitoring sites and structure.

1.2 BLACK CARBON

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

BC is typically formed through the incomplete combustion of fossil fuels, biofuel, and biomass, and is emitted in both anthropogenic and naturally occurring soot. Black carbon warms the planet by absorbing heat in the atmosphere and by reducing albedo (the ability to reflect sunlight) when deposited on snow and ice. Black Carbon stays in the atmosphere for periods of days to weeks, whereas CO_2 has an atmospheric lifetime of more than 100 years.

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

1.3 MEASUREMENT METHOD

1.3.1 Aethalometer instrument and data processing

Aethalometers quantify Black Carbon on filter samples based on the transmission of light through a sample. The sample is collected onto a quartz tape, and the change in absorption coefficient of the sample is measured by a single pass transmission of light through the sample, measured relative to a clean piece of filter. The system evaluates changes in two optical sensors (sample and reference), with the light source both on and off, such that independent measurements of the change in attenuation of the sample are produced for averaging periods of typically five minutes. The absorption coefficient for material added during the period, σ [m⁻¹], is calculated from the attenuation change, the filter area, and volume of the sample. This is converted to a Black Carbon concentration for the period, as a first approximation, using a mass extinction coefficient [16.6 m².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, 880 nm and 370 nm. The 880 nm wavelength is used to measure the Black Carbon (BC) concentration of the aerosol, while the 370 nm wavelength gives a measure of the "UV component" of the aerosol. At wavelengths shorter than about 400 nm, certain classes of organic compounds (such as polycyclic aromatic hydrocarbons, and also certain compounds present in tobacco smoke and smoke from wood burning) start to show strong UV absorbance. The UV component can therefore in principle be used as an indicator of wood and solid fuel emissions.

The UV component concentration presented in this report is obtained by subtracting the measured BC concentration from the concentration measured by the 370 nm source with a mass extinction coefficient of 39.5 m².g⁻¹, which is the coefficient for this wavelength corresponding to the value used at 880 nm for "ideal" material. The UV component is not a real physical or chemical material, but a parameter based on UV absorption due to the mix of organic compounds measured at this wavelength. This metric termed 'UVPM' is expressed in units of 'BC Equivalent'.

It is well known that the assumption of constant mass extinction coefficient does not hold as the filter spot darkens, leading to nonlinearity in the Aethalometer response. The effect of this nonlinearity

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

means that the Aethalometer has reduced sensitivity to black carbon at high filter tape loadings. To correct for this nonlinearity, the model developed by A Virkkula² has been used to correct for increased attenuation due to spot darkening during sampling. This uses the simple equation:

$$BC_{corrected} = (1 + k.ATN).BC_{uncorrected}$$
 Eqn 1

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

1.3.2 Sampling

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

2.0 NETWORK INFRASTRUCTURE

The following sections present the design of the Network, describe its operation and the changes to the Network in 2016.

2.1 NETWORK SITES AND DESIGN

Figure 1 shows the locations of the Aethalometers during 2016. The sites are located to target the measurement of traffic emissions of Black Carbon in urban areas, and of solid fuel and biomass emissions in Northern Ireland & Cardiff.

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

² 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

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

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

The Birmingham Tyburn Roadside site shut on 7th September 2016 and was relocated to Birmingham Keeley Street which started on 5th October. The Harwell site stopped at the end of 2015 and was replaced by Chilbolton which began measuring on 14th January 2016.

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

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

Table 2 Sites to measure non-traffic related emission sources

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

Key for Figure 1:

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





2.2 NETWORK OPERATION

The operation of the Network was set up to mirror that of the AURN, to include a Central Management and Control Unit (CMCU) and a Quality Assurance and Quality Control Unit (QA/QC). The Environmental Research Group at King's College London (King's) carries out the 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 μ g m⁻³); the flow data is also checked to ensure it is 4 l/min (±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 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	26/10/2016	862
Ballymena	03/11/2016	849
Belfast	02/11/2016	855
Birmingham Keeley Street	25/10/2016	863
Birmingham Urban background	25/10/2016	859
Cardiff Centre	01/11/2016	869
Detling	21/10/2016	865
Dunmurry Kilmakee	02/11/2016	861
Glasgow High Street	26/10/2016	868
Glasgow Townhead	27/10/2016	856
Chilbolton	19/10/2016	851
Marylebone Road	20/10/2016	866
North Kensington	20/10/2016	867
Strabane	03/11/2016	848

Table 3 Site Audit Visits

3.1.1 Sampler Leak Rate and Calibration of Sample Flow

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

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

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

According to the manufacturer, the maximum acceptable leak rate is 20%. Black Carbon concentrations were not corrected for leak rate, but the leak rate is included in the uncertainty budget.

	Leak Rate,	Indicated Flow,	Inlet Flow,
Site	%	lpm	lpm
Auchencorth Moss	6.7	4.0	4.113
Ballymena	34.6	4.0	2.747
Belfast	5.7	4.0	4.277
Birmingham Roadside	9.4	4.0	4.007
Birmingham Urban Background	9.5	4.0	3.730
Cardiff Centre	6.6	4.0	3.980
Detling	5.2	4.0	3.920
Dunmurry Kilmakee	7.0	4.0	3.953
Glasgow High Street	4.8	4.0	4.370
Glasgow Townhead	8.5	4.0	3.667
Chilbolton	4.5	4.0	4.340
Marylebone Road	7.2	4.0	4.180
North Kensington	7.7	4.0	4.037
Strabane	-	4.0	0.000

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

Note: The leak at Ballymena and fault at Strabane were fixed by the ESU shortly after the audits. See section 5.2.3 for more information on the fault at Strabane.

Table 4 Aethalometer leak rates and sample flows

3.1.2 Instrument Performance

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



Figure 3 Detling Aethalometer sampling HEPA filtered air

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

Site	BC, μg.m ⁻³	UV Channel, µg.m ⁻³	Annual Average BC, μg.m ⁻³
Auchencorth	0.06	0.05	0.1
Ballymena	0.32	0.26	0.8
Belfast	0.47	0.40	1.2
Birmingham Keeley St	0.40	0.37	3.2
Cardiff	0.65	0.38	0.9
Chilbolton	0.06	0.13	0.5
Detling	0.24	0.21	0.5
Dunmurry	0.20	0.16	0.8
Glasgow High St	0.59	1.06	1.7
Glasgow Townhead	1.02	0.60	0.9
Marylebone Road	0.60	0.59	4.9
North Kensington	0.10	0.23	1.1
Strabane	0.29	0.18	1.8

Note: There is no data for Birmingham Tyburn Background as there was a problem with data collection during the audit.

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

The noise value is higher than that expected to be applicable in normal operation due to the short averaging times used for the audit test, which mean that short-term humidity changes caused by air conditioning are more apparent. Values at or below $1.0 \ \mu g.m^{-3}$ are considered acceptable. Some sites

like Glasgow Townhead have been found to have slightly larger noise valuers because they are small enclosures containing a large amount of equipment. 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.

4.0 MEASUREMENT UNCERTAINTY

4.1 SAMPLE VOLUME

From measurements at the site audit the sample volume can be determined with an uncertainty of $\pm 10.1\%$, 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 was not used to correct the results, but was included as an uncertainty if the sampler passed 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 2016 audits was used. As leak rate is considered to be a rectangular distribution, its contribution to the standard uncertainty in sample volume was 4.0%.

4.2 MEASUREMENT OF ABSORPTION

The Aethalometer measurement does not depend on any absolute calibration of the detectors' response signals, but instead relies upon their ability to determine very small relative changes in optical transmission. Determining the zero noise of the system gives relevant information on the instrument's ability to measure small changes in optical transmission. Results from the HEPA filter zero noise tests show that the stability of the optical / electrical system was approximately $\pm 0.11 \,\mu g.m^{-3}$ for hourly means, compared to the network mean of $1.33 \,\mu g.m^{-3}$. Converting this into a standard uncertainty represents a contribution of 8.0%,

4.3 CORRECTION FOR SPOT DARKENING

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

The differences between using individual spot corrections versus seasonal or monthly corrections have been examined, with the conclusion that 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 were combined, the overall measurement uncertainty for hourly Black Carbon concentrations was 23.7%, expressed with a level of confidence of 95%. The only source of uncertainty in the overall measurement uncertainty that reduced 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	18.9%
Monthly	6.3%
Yearly	6.3%

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

This is an indicative measurement uncertainty for the Aethalometer method, using the conventional mass extinction coefficient 16.6 m².g⁻¹ and was calculated from the results of the 2016 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 2016 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 method².

The hourly data set for Black Carbon and UV component concentrations can be downloaded from Defra's UK-AIR: Air Information Resource Web Pages 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 2016. 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 $40 \ \mu g.m^{-3}$ to enable easy comparison between charts, except for Figure 8 which shows concentrations measured at rural locations.



Figure 4 Black Carbon concentrations during 2016 in Northern Ireland



Figure 5 Black Carbon concentrations during 2016 in Scotland



Figure 6 Black Carbon concentrations during 2016 in Wales and the Midlands



Figure 7 Black Carbon concentrations during 2016 in London



Figure 8 Black Carbon concentrations during 2016 at Rural Locations

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

Other concentration peaks related with air pollution events were also seen. A period of smog in London from $19^{\text{th}}-21^{\text{st}}$ January was shown in the North Kensington and Detling site data and was also visible at the Birmingham sites. Other events of high PM₁₀ and PM_{2.5} were visible in the London and Birmingham sites from $10^{\text{th}}-13^{\text{th}}$ and $22^{\text{nd}}-23^{\text{rd}}$ March.

Elevated concentrations towards the end of October were seen in the majority of sites. A pollution event affecting Northern Ireland was seen in these sites from 20-21st October, and a peak from 25th-26th of October was seen both in Northern Ireland and many other sites across the country. Another peak from 29th-30th October was seen in London and Birmingham.

Most UK sites measured elevated black carbon concentrations between 29th November and 6th December which was related to a countrywide air pollution episode. A significant high was also seen at the North Kensington site at the end of December, between the 27th and 30th. This was a PM event focussed on London and is visible at Birmingham but not in Scotland or Northern Ireland.

5.1.2 UV component

The following charts show the UV component concentrations measured by the UK Black Carbon Network for 2016. 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 2016 in Northern Ireland



Glasgow High Street	Glasgow Townhead	Auchencorth Moss

Figure 10 UV component concentrations during 2016 in Scotland



Figure 11 UV component concentrations during 2016 in Wales and the Midlands



Figure 12 UV component concentrations during 2016 in London



Figure 13 UV component concentrations during 2016 at Rural Locations

The Northern Irish sites measured increased UV component concentrations during the cold periods in January, February, March, October November and December. Due to a fault in the instrument at Strabane, data between 4th April and 7th November there is no data available for these months which

means the heating season and any elevations through the summer months can't be identified. In Ballymena, the heating season ran until early April and then started again late-September until the end of the year. Based on the evidence at Ballymena the heating season was slightly shorter in 2016 compared to 2015 and the overall concentrations were lower across the year.

Many of the sites showed the same elevated concentrations in UV component as BC concentrations related to the countrywide air pollution event in the first week of December. The large, positive UV spikes in the data for Detling were likely due to localised bonfires.

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 370mm 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*³ 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 was also found at Birmingham Tyburn Roadside, which still has high traffic flows but less diesel based exhaust emissions than Marylebone Road. This effect was also seen at Glasgow High Street but was 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.

³ 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 2016.

Site	Mean concentration
Site	µg.m⁻³
Auchencorth Moss	0.1
Ballymena	0.8
Belfast Centre	1.2
Birmingham Tyburn BK	1.2
Birmingham Tyburn RS	2.1
Bimringham Keeley Street	3.2
Cardiff Centre	0.9
Detling	0.5
Dunmurry Kilmakee	0.8
Glasgow High Street	1.7
Glasgow Townhead	0.9
Chilbolton	0.5
Marylebone Road	4.9
North Kensington	1.1
Strabane	1.8

Note: Birmingham Tyburn Roadside, Birmingham Keeley Street and Chilbolton are not full calendar years.

Table 6Annual Mean Black Carbon Concentrations for 2016

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 2016

It should be noted that due to the fault at Strabane there is no data between 4th April and 7th November. This has resulted in a skewed yearly average as only the winter months, which were expected to have much higher concentrations, were included.

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.

	Increment, μg.m ⁻³	
Conurbation	Urban	Roadside
London	0.6	3.7
Birmingham- Tyburn	0.6	1.2
Birmingham- Keeley Street	-	1.3
Glasgow	0.8	0.8

Table 7Increments in Black Carbon concentrations between rural, background and roadside
sites in 2016

It can be seen that the urban increment for all three areas was similar, while the roadside increment for London was much larger than that for Birmingham and Glasgow. The roadside site at Birmingham was moved part way through the year and so the data were separated for these calculations. The new site at Keeley Street appeared to measure higher concentrations than were seen at Tyburn Road, but the roadside increment is still well below the one seen in London. This is due to the much larger traffic flow and different vehicle profile of the Marylebone Road site compared with the Birmingham Tyburn and Glasgow High Street roadside sites. Highways Agency traffic count data for 2016 for the three roads passing the monitoring stations are given in Table 8.

	Motor	Cars	Buses	All	All Motor
Road (Count Point ID)	cycles	Taxis	Coaches	HGVs	Vehicles
Marylebone Road (27236)	4719	55207	2774	2851	74932
Tyburn Road (56399)	145	23762	250	1601	31487
Keeley Street (27736)	330	36992	97	2697	46516
Glasgow High Street (10821)	40	12784	210	411	15780
Ratio London to Birmingham					
(Tyburn Road)	33	2.3	11	1.8	2.4
Ratio London to Birmingham					
(Keeley Street)	14	1.5	29	1.1	1.6
Ratio London to Glasgow	118	4.3	13	6.9	4.7
Ratio Birmingham (Tyburn					
Road) to Glasgow	3.6	1.9	1.2	3.9	2.0
Ratio Birmingham (Keeley					
Street) to Glasgow	8.3	2.9	0.5	6.6	2.9

Table 82016 Average daily traffic count data for Marylebone, Tyburn Road and Glasgow
High Street Roadside sites

As the Tyburn Road site was operating for the majority of the year and there was limited data from Keeley Street, Tyburn Road has been used in the following calculations. The Marylebone Road

roadside increment in Black Carbon concentration in 2016 was a factor of 3.1 higher than the Tyburn Road increment. This is somewhat higher than the ratio of numbers of cars / taxis and HGVs between the sites at 2.3 and 1.8 respectively. This means there was a significant input from buses / coaches and motorcycles to result in the larger increase in increment. There were 11 times more buses / coaches and 33 times more motor cycles passing the Marylebone Road site compared with the Birmingham site, which indicates these are a predominant source of Black Carbon emissions at Marylebone Road. However, although an increased increment is seen this is not as large as might be expected from the ratio of buses / coaches between the sites, which reflects the cleaner fleet in London. Changes in emissions from London buses and taxis are discussed further in section 5.5.1.

The Birmingham roadside increment was 1.5 times that of Glasgow. It had twice the number of motor vehicles, the category with the biggest increase at Birmingham compared to Glasgow is HGVs. This supports the hypothesis that HGVs are not a predominant source of black carbon, as a large increase in these vehicles did not also result in a large increase in the Black Carbon increment.

Figure 15 shows how the urban and roadside increments in London and Birmingham have changed over the period 2012 to 2016. The average urban background at both locations is roughly stable with increases during the heating season indicating the contribution from domestic heating. This effect appears reduced in the winter 2015-2016 in both areas which could be a sign of warmer winter temperatures than previous years. The roadside increments for London and Birmingham have clearly dropped over the whole period.



Figure 15 Urban and roadside increments for London and Birmingham for the period 2012 to 2016

The change of London roadside increment in Black Carbon was also correlated with a reduction of the London roadside increment in PM_{2.5} concentration⁴ from 2010-2014 as shown in Figure 16 where a 1:1 correspondence was valid.

⁴ 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.



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

Figure 16 Relationship between trend in London roadside Black Carbon and PM_{2.5} increments between 2010 and the end of 2014.

This indicates similar emission sources (road transport) for Black Carbon and PM_{2.5} roadside increments, as expected.

5.2.2 UV component

Cit -	Mean concentration		
Site	µg.m⁻³		
Auchencorth Moss	0.1		
Ballymena	0.5		
Belfast Centre	0.3		
Birmingham Tyburn BK	0.2		
Birmingham Tyburn RS	0.2		
Birmingham Keeley Street	0.4		
Cardiff Centre	0.2		
Detling	0.2		
Dunmurry Kilmakee	0.4		
Glasgow High Street	0.2		
Glasgow Townhead	0.1		
Chilbolton	0.3		
Marylebone Road	0.3		
North Kensington	0.3		
Strabane	1.6		

Table 9 gives the annual average for each site for 2016.

Note: Birmingham Tyburn Roadside, Birmingham Keeley Street and Chilbolton are not full calendar years.

Table 9 Annual Mean UV component Concentrations for 2016

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





It should be noted that due to the fault at Strabane there was no data between 4th April and 7th November. This has resulted in a skewed yearly average as only the winter months, which were expected to have much higher concentrations, have been included.

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.

	Increment, µg.m ⁻³		
Conurbation	Urban	Roadside	
London	0.0	0.1	
Birmingham	0.0	0.0	
Scotland	0.0	0.1	

Table 10Increments in UV component concentrations between rural, background and
roadside sites in 2016

There was no significant difference in increments between 2015 and 2016.

It can be seen that the urban and roadside increments at all sites were similar, as domestic emissions in the three areas were of a similar magnitude due to similar fuel types and that road traffic was not a significant source for the UV component. Although the roadside site at Birmingham moved part way through the year, the data from both has been used to calculate the single increment as there was no noticeable different in UV component concentrations between the two.

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,	Increment compared to Belfast,
	μg.m⁻³	%
Dunmurry	0.1	22
Ballymena	0.1	33
Strabane	1.3	305

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. As discussed previously, there was only data for Strabane until April 4th and then from November 7th to the end of the year. These were the major heating months of the year, and the very large increment compared to Belfast from this data confirms that this was due to heating sources. Ballymena is supplied by natural gas, but the benzo[a]pyrene (BaP) concentrations measured by the PAH Network were higher than might be expected. A similar increase in the UV component was 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 was 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 PM₁₀.
Compared to 2015, the Ballymena increment has decreased and the Dunmurry and Strabane have increased. The increase at Strabane from 0.6 μ g.m⁻³ to 1.3 μ g.m⁻³ was due to the data being skewed by an instrument fault in the summer. At Dunmurry the increase was small, from 0.0 μ g.m⁻³ to 0.1 μ g.m⁻³ which was also the value seen in 2014. This suggests that this increase is not a significant trend but just small fluctuations between years. The decrease at Ballymena does appear to be a trend as it fell from 0.5 μ g.m⁻³ in 2014 to 0.3 μ g.m⁻³ in 2015 and this has continued in 2016 when 0.1 μ g.m⁻³ was recorded. This decrease may be due to the warmer weather in the winter of 2015-2016, or reduced use of solid fuel for domestic heating due to reduced fuel poverty.

5.2.3 Data Capture

Cit-	Data Capture	Time Coverage %	
Site	%		
Auchencorth Moss	97	97	
Ballymena	97	97	
Belfast Centre	95	95	
Birmingham Tyburn BK	100	100	
Birmingham Tyburn RS	98	67	
Birmingham Keeley Street	100	24	
Cardiff Centre	91	91	
Detling	96	96	
Dunmurry Kilmakee	100	100	
Glasgow High Street	94	94	
Glasgow Townhead	96	96	
Chilbolton	99	96	
Marylebone Road	98	98	
North Ken	97	97	
Strabane	41	41	

Table 12 gives the data capture for each site for 2016. Due to the Network changes during 2016 the time coverage for the complete calendar year for each site has also been given.

Table 12Data capture rates of the Aethalometers for 2016

The average data capture for the Network was 93% but this figure was significantly affected by one instrument fault at Strabane and the majority of sites obtained a data capture above 95%. The reduced time coverage at Birmingham Tyburn Roadside and Birmingham Keeley Street was due to the site move, Tyburn Roadside was operational until September and Keeley Street started in October. Chilbolton also had a lower time coverage as it didn't open until January 14th. The low data capture seen at Strabane was due to a problem with the flow at the inlet, which resulted in data from April to November being rejected as it was invalid. The fault was found at the scheduled audit in November (shown in Table 4) and was difficult to spot earlier because the instrument was still recording a normal flow and reading fluctuating, low concentrations. This was as would be expected during the summer months and without comparison of these values in a time series over the year they were not obviously incorrect. The problem was traced back to the instrument service in April, even though the postservice flow and leak test showed the instrument in good working order. From our experience of operating the network over many years this appears to have been a one-off error which is unlikely to happen again.

5.3 TEMPORAL VARIATIONS

The following section presents analysis of the 2016 Black Carbon and UV component concentrations with respect to the hour of the day. Charts of variations over the day of the week and the month in the year were made using the data from 2009 - 2016, 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 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 μ g.m⁻³ for Black Carbon and equivalent μ 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 2016 data are presented in Figures 18 to 20.

The 2009-2016 data are presented in Figures 21 to 22. These long term plots only include those sites which have been operating for the whole of this time period. The site at Chilbolton was seen to show significantly different concentrations from that at Harwell, so this site has been removed from the long term time series plots. Strabane is also not included in this report because the 2016 data is skewed by an instrument fault.

<u>Chart Key</u>

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 21-22 is for display purposes only, to allow the uncertainty in the mean value to be seen more clearly.

<u>Acknowledgement</u>

Figures 18 to 22 are generated using the Open-Air Tools run on the R software platform^{5,6}.

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 (2016). OpenAir: Open-source tools for the analysis of air pollution data, R package version 1.1-5

2016 Data



Marylebone Road

NPL Report ENV 21



Birmingham Keeley Street



Birmingham Tyburn Roadside



Figure 18 Roadside Sites



Strabane

NPL Report ENV 21







Dunmurry Kilmakee

35



Belfast Centre



North Kensington

NPL Report ENV 21



Birmingham Tyburn Background



Cardiff Centre



Figure 19 Urban Background Sites



Chilbolton

NPL Report ENV 21



Detling



Figure 20 Rural Sites



2009 – 2016 Data



Figure 21 Roadside Sites



Dunmurry Kilmakee

NPL Report ENV 21



North Kensington



Figure 22 Urban Background Sites

Roadside sites

It can be seen that in general the Black Carbon concentrations at the roadside sites followed the expected profile for traffic movements through the day, with raised concentrations in the morning and evening rush hours. This was particularly prominent at Glasgow High Street and the new site at Birmingham Keeley Street which had a significant evening rush hour peak. The site at Tyburn Road which was replaced showed a morning peak followed by a drop in concentrations during the day, with a smaller peak for the evening rush hour. Concentrations at Marylebone Road remained high throughout the day.

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

Weekend concentrations were also much lower at Birmingham Tyburn and Keeley Street with less obvious peaks. Keeley Street still showed variability throughout the weekend days however, particularly with a peak seen on Sunday evening which could be caused by return travel at the end of the weekend. As with Marylebone Road there was little UV component signature, however Birmingham showed a slight increase in the evenings of the weekends, indicating possible local solid fuel / wood burning secondary heating.

Glasgow High Street also showed lower, more stable concentrations of Black Carbon at the weekend than on weekdays.

Urban Background sites

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

Concentrations measured at Birmingham Tyburn, Cardiff Centre, North Kensington and Glasgow Townhead showed expected peaks in Black Carbon concentrations during the morning rush hour, with little increase in UV component concentrations. There was also an evening rush hour peak but the high Black Carbon concentrations from this continued into the evening. At the weekend there were elevated Black Carbon concentrations late into Saturday evening / early Sunday morning which could be due to evening leisure journeys. However there was often also a similar increase in UV component concentration which indicates domestic emission sources, probably from secondary heating. This effect was particularly visible at Cardiff Centre and Belfast Centre.

Black Carbon concentrations were generally lower at the weekends compared with working days. Both Black Carbon and UV component concentrations showed 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 were 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 23, using Belfast Centre as an example.



Figure 23 Normalised monthly variability at Belfast for the period 2009 - 2016

Concentrations measured at Strabane, Ballymena and Dunmurry Kilmakee in Northern Ireland were dominated by emissions from domestic heating. UV component and Black Carbon concentrations followed similar hourly, daily and seasonal trends and had similar absolute concentrations. The high concentration bias on the Strabane data for 2016 must be taken into account. Strabane is not on the natural gas supply and domestic heating mainly comes from oil. Strabane is in a smokeless zone, however it appears that solid fuel burning may be occurring in residential areas. Due to the large emission factors of PAHs from smoky coal⁷ compared to oil and gas, it does not take many houses burning this coal to have a big influence on ambient concentrations. At Strabane there was 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, but the evening rush hour was masked by domestic emissions which were also seen in the UV component. Figure 24 gives the normalised monthly variability and Figure 25 gives the hourly variability.

⁷ UK National Atmospheric Emissions Inventory



Figure 24 Normalised monthly variability at Strabane and Dunmurry for the period 2009 - 2016



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 25 Seasonal Black Carbon and UV component concentrations measured at Strabane and Dunmurry for the period 2009 - 2016

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

<u>Rural sites</u>

The rural background site concentrations were lower than the other site classifications, as expected, although the Chilbolton site did show higher concentrations than the site it replaced at Harwell. Chilbolton and Detling displayed the morning rush hour to a small extent, but this effect was less visible at Auchencorth Moss. Detling showed some evidence of the evening rush hour, but at Chilbolton and Auchencorth Moss the higher concentrations in the evening were later than would be expected for a traffic signal. All three remained relatively high into the evening and this was also seen in the UV component suggesting a domestic heating source. All three rural sites showed lower and more stable concentrations at the weekends with a heating trend seen but no visible effects from traffic.

5.4 COMPARISONS WITH OTHER POLLUTANTS

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

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

5.4.1 Elemental Carbon

Daily Elemental Carbon (EC) measurements are made at the North Kensington, Marylebone Road and Chilbolton sites by the Particle Concentration and Number Network⁸. Aethalometer concentrations (BC) at these sites have been averaged into daily measurements and plotted as scatter plots against the elemental carbon (EC) concentrations in Figures 26 to 28. The regression is calculated according to the Reduced Major Axis (RMA) method⁹, which is based on minimising the product of the *x* and *y* deviations between the data values and "fitted values" instead of the least squares method, which minimises the sum of the squared deviations between the dependent variable (y) and the "fitted values". RMA is better suited to air quality measurements as pollutant concentrations are often 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.

⁸ J Tompkins et al, Draft NPL REPORT, 2016 Annual Report for Airborne Particulate Concentrations and Numbers in the United Kingdom (phase 3), June 2017

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



Figure 26 2016 EC and BC Measurements at North Kensington



Figure 27 2016 EC and BC Measurements at Marylebone Road



Figure 28 2016 EC and BC Measurements at Chilbolton



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

Figure 29 2016 EC and BC Measurements for all 3 sites. The inset graph shows points close to the origin.

It can be seen that there were good linear relationships ($R^2 > 0.8$) between the Elemental Carbon and Black Carbon concentrations measured at all three sites. The regression parameters between Black Carbon and Elemental Carbon were 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 29. In all cases the intercept value was relatively small, indicating that there was no significant zero offset between the two methods, which are based on entirely independent principles. The annual regression results (all RMA) are shown in Table 13.

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

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

Table 13Relationships between Black Carbon and Elemental Carbon over the period 2009 –
2016

It can be seen that the slopes, i.e. the measured amounts of Black Carbon relative to Elemental Carbon, had been roughly constant at Marylebone Road from 2009 to 2014, at 1.4 ± 0.1 , but this dropped to around 1.2 in 2015 and remained at a similar value in 2016. The parameters at Harwell and North Kensington have been more variable, as would be expected because of the lower concentrations.

5.4.2 Polycyclic Aromatic Hydrocarbons (PAH)

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

$$BaP = a. UV^2 + b. UV + c$$
 Eqn 2

Where

BaP = predicted BaP concentration in ng.m⁻³;

UV = measured UV component concentration in μ g.m⁻³

Site type	Class	а	b	С
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 gives the coefficients a, b & c for the different site types.

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

Using this relationship, Table 15 shows the measured and predicted annual 2016 BaP concentration at each Aethalometer site based on the measured annual UV component concentration.

¹⁰ https://uk-air.defra.gov.uk

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.

			Predicted BaP,	Measured
	1		ng.m °	ВаР
		UV		
		component,		
Site	Class	µg.m⁻³	site specific	ng.m⁻³
Auchencorth Moss	R	0.08	0.0	0.0
Ballymena	NI	0.49	0.5	0.5
Belfast Centre	NI	0.31	0.3	
Birmingham Tyburn BK	UR	0.24	0.2	0.2
Birmingham Tyburn RS	UR	0.19	0.1	
Cardiff Centre	UR	0.23	0.1	
Detling	R	0.21	0.1	
Dunmurry Kilmakee	NI	0.38	0.4	0.5
Glasgow High Street	UR	0.21	0.1	
Glasgow Townhead	UR	0.12	0.0	0.1
Chilbolton	R	0.27	0.1	0.1
Marylebone Road	MY	0.34	0.4	0.2
North Ken	UR	0.27	0.2	
Strabane	NI	1.60	2.2	

Note: Birmingham Tyburn Roadside has been included instead of Birmingham Keeley Street because it ran for the majority of the year.

Table 15Predicted 2016 annual BaP concentration based on measured UV component
concentrations

It can be seen that the predicted is within 0.2 ng.m⁻³ at all of the sites where BaP is measured. As the 2016 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 (as this site has the highest measured UV components and is therefore the most likely of the sites to be close to the BaP target values). The 2016 data has not been added to this because the incomplete data gives an average that is not representative.

Year	UV component concentration	Predicted BaP concentration ng.m ⁻³
2009	0.9	1.1
2010	1.3	1.7
2011	0.8	0.9
2012	1.1	1.4
2013	1.2	1.5
2014	1.1	1.4
2015	0.9	1.1

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

Six out of these seven years have had predicted BaP concentrations above the 1.0 ng.m⁻³ target value in the EC Directive 2004/107/EC¹² relating to ambient BaP concentrations. The average concentration over these 7 years is predicted to be 1.3 ng.m⁻³.

5.4.3 Particulate Mass Concentration

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

		TEOM FDMS	TEOM FDMS	Percent BC	Percent BC
	BC	PM ₁₀	PM _{2.5}	PM ₁₀	PM _{2.5}
Site	µg.m⁻³	µg.m⁻³	µg.m⁻³	%	%
Marylebone Road	4.9	26	16	19	31
Birmingham Tyburn Roadside	2.1	18	12	12	18
Glasgow High Street	1.7	13	8	13	21
Birmingham Tyburn Background	1.2	16	11	8	11
North Kensington	1.1	20	12	6	9
Glasgow Townhead	0.9	12	7	8	13
Cardiff	0.9	19	10	5	9
Belfast Centre	0.8	16	10	5	8
Chilbolton	0.5	15	9	3	6
Detling [*]	0.5	20		3	
Auchencorth Moss	0.1	7	3	1	3

Note: * Local Authority run site; may not have identical QAQC procedures to AURN datasets.

Grey shaded cells indicate no measurements were made.

Table 17 Comparison of Annual Black Carbon and Particulate Mass Concentrations

It can be seen that the PM_{10} and $PM_{2.5}$ mass concentration measured at Marylebone Road, Birmingham Tyburn roadside and Glasgow High Street sites had a much higher percentage of Black Carbon than the other sites. Black Carbon represented a large proportion of the total particulate mass 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_{10} and $PM_{2.5}$ mass concentrations as found by $Font^4 et al$ (Figure 16).

At the rural background sites Black Carbon made up 3% or less of the PM₁₀ mass. This represents the regional background contribution.

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

5.5 TRENDS

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

5.5.1 Trends by Site

Figures 30 to 31 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 2016. Once again Harwell and Strabane have been removed from this analysis, Harwell permanently because the site has been moved to one which is not directly comparable, and Strabane for this year due to only having data for a small part of it. The Theil-Sen method in OpenAir^{4,5} was used to calculate the regression parameters including slope and uncertainty in the slope.

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

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



5.5.1.1 Black Carbon





Dunmurry



Belfast



North Kensington



Birmingham Tyburn



Site	Slope µg.m ⁻³ .year ⁻¹	Lower limit µg.m ⁻³ .year ⁻¹	Upper limit µg.m ⁻³ .year ⁻¹	Slope significant
Roadside				
Marylebone Road	-0.80	-0.94	-0.67	Y
Urban Background				
Belfast Centre	-0.09	-0.14	-0.04	Y
Birmingham Tyburn	-0.08	-0.12	-0.04	Y
North Kensington	-0.09	-0.13	-0.04	Y
Dunmurry	-0.07	-0.13	-0.01	Y

Table 18 summarises the slopes at each site.

Table 18 Summary of Black Carbon trends

Over the period 2009 to 2016 all of the long running sites in the network have shown a significant downward trend in Black Carbon concentrations. At the non-roadside sites this trend was likely to be due to significantly lower Black Carbon concentrations measured during 2015. This was probably due to meteorological conditions as the last quarter was significantly wetter and warmer than in previous years. 2015 was the sixth wettest year since 1910. These conditions continued into the first two months of 2016 and relatively low concentrations were still seen for this time. Towards the end of 2016 concentrations appeared to rise again at all of these but this was not enough to affect the downward trend. The other factor which was likely to have contributed to the overall decrease in concentrations was the introduction of Euro 6/VI vehicles into the fleet.

However, the decrease at Marylebone Road was much larger and the black carbon concentrations have been reducing year on year since 2011. The 2016 annual mean concentration was less than half that of 2011. The concentrations of Elemental Carbon made at Marylebone Road have followed a similar trend. Figure 32 shows the annual Black Carbon and Elemental Carbon concentrations along with the average daily traffic flow past the site.



Figure 32 Annual Average Black Carbon, Elemental Carbon and Motor Vehicles per Day at Marylebone Road for the period 2009 – 2016

It can be seen that the changes in Black Carbon and Elemental Carbon concentrations followed changes in the total traffic flow for the years 2009 to 2012 but they do not correlate so well from 2013 onwards. This would indicate that Black Carbon emissions per vehicle have changed over the last 4 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 fleet¹³. Table 19 shows the composition of the London bus fleet over the period 2010 to 2016. The bottom row of the table shows the percentage of low emission buses, which is a combination of the hybrid, electric and fuel cell / hybrid bus numbers. In addition all of London's Euro II and III diesel buses were retro-fitted with engine exhaust particulate filters by the end of 2011, which would have also reduced Black Carbon emissions.

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

¹³ 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

		Number of buses						
Bus Type	Drive train	2010	2011	2012	2013	2014	2015	2016
	type							
New Routemaster	Hybrid	0	0	5	8	168	432	736
Routemaster	Diesel	18	18	19	20	19	19	10
Artic	Diesel	320	260	0	0	0	0	0
	Diesel	2,676	2,670	2,661	2,608	2,606	2,662	2,617
Single deck	Fuel	0	5	5	5	8	8	8
	Cell/Hybrid							
	Hybrid	27	27	33	28	23	23	18
	Electric	0	0	0	0	2	8	17
Double dook	Diesel	5,554	5,487	5,787	5,696	5,296	5,026	4,794
	Hybrid	29	79	233	352	643	799	981
TOTAL		8,624	8,546	8,743	8,717	8,765	8977	9186
% low emission		0.65	1.30	3.16	4.51	9.63	14.15	19.16

Table 19Composition of London bus fleet, 2010 to 2016

Figure 33 is Figure 32 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 33 Annual Average Black Carbon, Elemental Carbon and 100 – percentage of low emission buses in the London bus fleet for the period 2009 – 2016
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 34 to 35 show the UV component trends.



Marylebone Road





Dunmurry



Belfast



North Kensington



Birmingham Tyburn



UV component concentrations measured at urban background sites, 2009 – 2016

Site Slope Lower limit Upper limit Slope µg.m⁻³.year⁻¹ µg.m⁻³.year⁻¹ µg.m⁻³.year⁻¹ significant Roadside Marylebone Road 0.04 0.03 0.06 Υ **Urban Background** -0.04 -0.08 -0.01 Υ Dunmurry **Belfast Centre** 0.00 -0.02 0.02 Ν North Kensington 0.00 0.01 Ν -0.02 Birmingham Tyburn -0.01 -0.02 0.00 Ν

Table 20 summarises the slopes at each site.

Table 20Summary of UV component trends

The Marylebone Road UV component concentration showed a significant upward trend over the period 2009 to 2016, this was 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 from relatively high values, it is to be expected that the calculation of the small UV component will be affected. It is unlikely that the UV component emissions across London have risen in the last two years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which were not dominated by traffic, have fallen. This trend should be treated with caution due to the low concentrations involved.

There was also a significant downward trend in the Dunmurry data which was most likely to be caused by the warmer and wetter weather over the 2015-2016 winter 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 were strongly affected by domestic solid fuel use, are plotted in Figure 36, along with average temperature for same period. Strabane data from 2016 haven't been included due to the instrument fault during this year. Temperature measurements from Armagh have been used as this is the nearest Met Office site with a long time series.



Figure 36 Strabane monthly UV component concentration and average ambient temperature for 2009 – 2015

It can be seen that the UV component concentration was inversely related to the average ambient temperature. This was due to the main source of UV component emissions being local domestic heating in Strabane. This was evident in both the winter and the summer indicating that there were still solid fuel emissions in the summertime. The relationship is shown in Figure 37 as a scatter plot.



Note: Valid Strabane 2016 data are included.

Figure 37 Scatter plot of monthly UV component concentration versus ambient temperature at Strabane over the period 2009 -2016

It can be seen that there was a clear linear relationship between increased UV component concentrations with a drop in ambient temperature, due to the increase in fuel usage in cold weather periods. It can also be seen that there was a significant UV source when temperatures were below 15° C, linking the UV component to fuels used for domestic heating systems. There was only 1 month where the monthly UV component concentration was less than the detection limit of 0.1 μ g.m⁻³, indicating that there was a UV component emission source all year.

5.5.2 Trends over the Network as a Whole

Figures 38 and 39 show the Network annual mean and median concentrations for Black Carbon and UV component for the subset of sites that have been continuously running since 2009 (Belfast Centre, Birmingham Tyburn UB, Dunmurry, Harwell/Chilbolton, Marylebone Road, North Kensington and Strabane). The median concentration is shown to remove the influence of large changes in a single site that would skew the overall result for the Network. Due to the inclusion of this median concentration, and as the data shown is an average of a large number of sites, Strabane and Chilbolton data are included even though they are not directly comparable to previous years at these sites.







Figure 39 Annual average UV component concentrations for long-term sites

It can be seen that the network annual mean for Black Carbon has been generally decreasing over this time, with significant drops in 2013 and 2015. This drop is mainly driven by the drop in concentrations at Marylebone Road. There was an increase seen in 2016 but this was only a slight change and was likely due to the particularly low concentrations seen at many sites in 2015 rather than an upward trend. The median was relatively stable ($1.35 \ \mu g.m^{-3} \pm 0.25 \ \mu g.m^{-3}$) from 2010-2014 and then also dropped in 2015, likely due to the warmer and wetter weather seen at the end of 2015. The concentration increased slightly again in 2016 as this did not have so much of an effect.

The annual average UV component concentrations varied from year to year with the maximum concentration seen in 2010. 2010 had very cold and snowy periods at the start and end of the year, especially in Northern Ireland, as shown in Figure 45, so elevated concentrations were probably due to domestic emissions. The relatively high value in 2016 was likely due to the partial data from Strabane, as the median value remained constant, very close to the value it has been since 2011.

6.0 EUROPEAN STANDARDISATION

The European standardisation body CEN has formulated a European Standard on the measurement of Elemental Carbon and Organic Carbon deposited on filters. This standard was published in March 2017 as BSEN 16909. The EUSAAR2 thermal protocol combined with transmission OC/EC split point determination has been adopted as the standard analysis protocol for PM_{2.5} particulate matter collected on filters. All filters for 2016 (on the Particles network) were analysed using the CEN standard method. Previously filters were analysed using the NIOSH transmission protocol.

Protocols that have a lower maximum temperature during the inert-gas heating phase, such as the EUSAAR2 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. Parallel analyses within the Particles network have shown that differences in results from the two protocols for these sites are relatively small.

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

7.0 CONCLUSIONS

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

Black Carbon concentrations measured at most sites in 2016 were similar to those in previous years, with Marylebone Road concentrations continuing to drop as they have since 2012. Although at the majority of sites the Black Carbon concentrations in 2016 were higher than 2015, these increases were small and the Network mean increased only slightly to 1.33 μ g.m⁻³ from 1.26 μ g.m⁻³. UV component concentrations in 2016 were very similar to previous years.

From looking at the trend in Black Carbon concentrations for an average of those sites that have been open from 2009 – 2016, the concentration was stable from 2009 to 2012. This was followed by a significant drop of 22% from 2.51 μ g.m⁻³ in 2012 to 2.06 μ g.m⁻³ in 2013. This fell to 1.98 μ g.m⁻³ in 2014

and again by 23% to 1.53 μ g.m⁻³ in 2015. There was an increase of 7% to 1.64 μ g.m⁻³ in 2016. There were some site changes and issues that could have contributed to this, Chilbolton showed higher concentrations that the site at Harwell which it replaced, and an incomplete dataset at Strabane resulted in a skewed average for 2016. The median concentration is much less susceptible to changes at one site. This dropped by 12% from 1.63 μ g.m⁻³ to 1.43 μ g.m⁻³ in 2013, dropped to 1.40 μ g.m⁻³ in 2014 and fell again by 21% to 1.1 μ g.m⁻³ in 2015. In 2016 there was a rise of 8.1% to 1.19 μ g.m⁻³. The drop seen in 2015 was due to the reduction at Marylebone Road and the wetter and warmer weather in the last quarter of 2015. While this continued into the first few months of 2016 the overall effect was not the same and average concentrations in this year were generally higher. A similar drop in Elemental Carbon concentrations was also seen at Marylebone Road.

The average 2016 UV component concentration was 0.5 μ g.m⁻³, a rise from the roughly constant values of 0.3-0.4 μ g.m⁻³ seen from 2009 to 2015. This increase can be attributed to skew in the Strabane data as the median value has remained relatively constant. The exception in 2010 (mean 0.55 μ g.m⁻³, median 0.47 μ g.m⁻³) 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 2016, Marylebone Road, Belfast Centre, Birmingham Tyburn, North Kensington and Dunmurry have shown a significant downward trend in Black Carbon concentrations. This overall trend is not affected by the small increase in concentration seen at most of these sites in 2016. At the four non-roadside sites this trend was likely to be influenced by a significantly wetter and warmer winter over 2015-2016 which lowered the Black Carbon concentrations in 2015. 2015 was the sixth wettest year since 1910. Although the average concentration increased slightly at these sites in 2016 it was not enough to affect the downwards trend. However, Marylebone Road has been showing reduced Black Carbon concentrations year on year since 2011, with 2016 annual mean concentration less than half that of 2011. This drop in concentration was likely to be due to the increased number of low emission (hybrid) buses in the London bus fleet and stricter emission controls on London taxis, HGCs, lorries and vans (Euro III to Euro IV). Low emission buses now make up 19% of the fleet.

The Marylebone Road UV component concentration showed a significant upward trend over the period 2009 to 2016. This trend should be treated with caution due to the low concentrations involved. It was 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 has fallen from relatively high values, it is to be expected that the calculation of the small UV component will be affected. It is unlikely that the UV component emissions across London have risen in the last 2 years due to domestic fuel usage as the Black Carbon concentrations at North Kensington, which were not dominated by traffic, have remained stable. There was also a significant downward trend in the Dunmurry data which was most likely to be caused by the warmer and wetter 2015-2016 winter 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, $0.6 - 0.8 \ \mu g.m^{-3}$, 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 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 showed that the dominant emission sources are road traffic and domestic heating using non-smokeless fuel. The diurnal average concentrations of the UV

component showed that its main source is domestic heating with solid fuels with little influence from traffic.