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Annual report for 2015 on the UK PAH Monitoring and Analysis Network

Report to the Environment Agency, Department for Environment Food and Rural Affairs the Department of Environment Northern Ireland, the Welsh Government and the Scottish Government

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EXECUTIVE SUMMARY

This annual report for 2015 for the UK PAH Monitoring and Analysis Network was prepared by the National Physical Laboratory (NPL) for the Environment Agency, The Department for Environment, Food and Rural Affairs, the Department of Environment Northern Ireland, the Welsh Government and the Scottish Government. The report contains:

- The measured annual (and where applicable, monthly or quarterly) mean concentrations of benzo[a]pyrene (B[a]P) at each Network station.
- Selected results from other PAHs at selected Network stations.
- A comparison of the B[a]P annual mean concentrations against the target value and assessment thresholds in the EC Fourth Daughter Directive.
- A comparison of the performance of Network operation against the relevant data quality objectives in the Fourth Daughter Directive.
- An overview of the PAH Network, including a summary of Network operation, a description of notable station maintenance issues and changes to the Network during 2015, and data capture statistics for 2015.
- A review of the sources of PAHs in the UK.
- A discussion of PAH concentration trends across the Network and the effect of gaseous oxidants on the annual measured concentrations.
- A summary of scientific research, international representation and other activities related to the Network.

In summary, during 2015:

- The EC target value for B[a]P (an annual mean concentration of 1 $ng.m^{-3}$) was exceeded at two of the 31 stations where PM_{10} particulate samples were taken.
- The EC upper assessment threshold for B[a]P (an annual mean concentration of 0.6 $ng.m^{-3}$) was exceeded at a total of five of these 31 stations.
- The EC lower assessment threshold for B[a]P (an annual mean concentration of 0.4 $ng.m^{-3}$) was exceeded at a total of six of these 31 stations.
- Ten stations have a measured mean annual B[a]P concentration above the UK air quality objective of 0.25 $ng.m^{-3}$, a decrease from eleven stations in 2014.
- The annual mean B[a]P concentrations at each site in 2015 did not show a significant change from those measured in 2014.

Contents

1 INTRODUCTION

This report was prepared by the National Physical Laboratory (NPL) as part of the 2010-2015 UK PAH Monitoring and Analysis Network ('the Network' or 'the PAH Network') contract number AQ0636 with the Environment Agency, The Department for Environment, Food and Rural Affairs, the Department of Environment Northern Ireland, the Welsh Government and the Scottish Government.

NPL assumed full operation of the Network in October 2010 following a one month handover period from the previous contractor. This annual report therefore presents and discusses data from NPL's fourth full year as operator of the Network.

This annual report contains:

- An introduction to polycyclic aromatic hydrocarbons (PAHs) and air quality policy, including a review of the sources of PAHs in the UK.
- An overview of the Network, including a summary of Network operation, and a description of notable station maintenance issues and changes to the Network during 2015.
- Network data capture statistics for 2015.
- A comparison of the performance of Network operation against the relevant data quality objectives in the EC Fourth Daughter Directive.
- Measured annual and monthly mean concentrations† of benzo[a]pyrene (B[a]P) at all monitoring stations during 2015.
- A comparison of the B[a]P annual mean concentrations against the target value and upper and lower assessment thresholds in the Fourth Daughter Directive.
- Measured monthly concentrations of selected other PAHs.
- A discussion of events during 2015 that resulted in the measurement of unusual PAH concentrations.
- Results from the analysis of deposition samples at two Network stations.
- A discussion of PAH concentration trends across the Network in the last 6 years.
- A summary of scientific research, international representation and other activities related to the Network.

The annexes of this report presents tables of data for the monthly concentrations of B[a]P at all Network stations. Monthly concentration data for all other PAHs at all Network stations can be found on the PAH Network data page of the UK-AIR website (http://uk-air.defra.gov.uk).

† Throughout this report, the term 'concentration' is used to denote mass concentration (mass per volume, *i.e.* $ng.m^{-3}$).

2 POLYCYCLIC AROMATIC HYDROCARBONS & AIR QUALITY POLICY

2.1 POLYCYCLIC AROMATIC HYDROCARBONS

Polycyclic aromatic hydrocarbons (PAHs) are a group of persistent organic pollutant compounds, some of which are toxic and are possible or proven human carcinogens [1]. These potential health hazards mean the measurement of the concentration of PAHs in ambient air is essential in order to protect the health of the public and the environment.

Benzo[a]pyrene has been identified as a human carcinogen and is the PAH most harmful to human health, and is therefore the PAH which is specified for monitoring by the EC Fourth Daughter Directive (Directive 2004/107/EC) [2]. B[a]P has also been determined to be a suitable 'marker' PAH to assess the concentration of all PAHs in ambient air [3]. Measurements of B[a]P in ambient air are covered by the European standard EN 15549 [4], which has been adopted as the European reference method.

In order to confirm the use of B[a]P as a marker PAH, and to assess the contribution of B[a]P in ambient air, the Fourth Daughter Directive also requires a number of other PAHs to be monitored at a limited number of measurement stations. As a minimum, these PAHs must include: benz[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, indeno[1,2,3-cd]pyrene and dibenz[a,h]anthracene. The structure of these six PAHs and B[a]P is shown in Figure 1. CEN TC264 WG21 is currently developing a Technical Specification [5] for the measurement of these PAHs and benzo[ghi]perylene in the particulate phase.

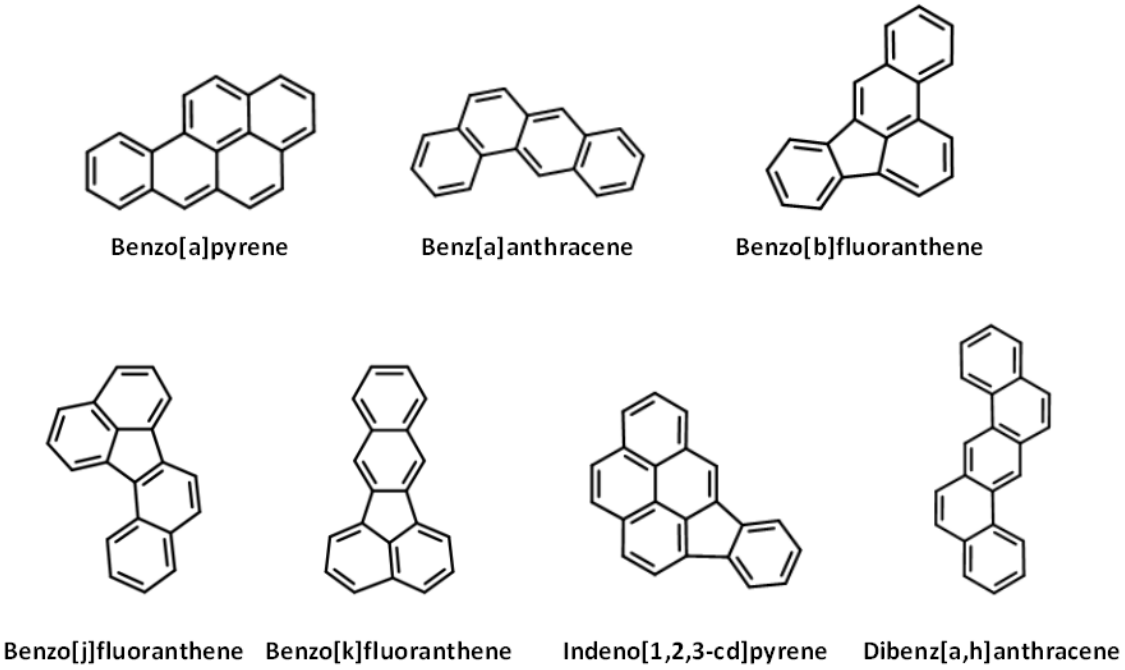

Figure 1 Structure of the seven PAHs specified by the Fourth Daughter Directive

PAHs are produced via incomplete combustion of carbon containing fuels from industrial, commercial, vehicular and residential sources. A detailed discussion of the sources of PAH emissions across the UK is presented in Section 2.3.

In order to demonstrate compliance with legislation that imposes limit and target values relating to ambient air, and to measure human and environmental exposure, the concentration levels of PAHs need to be measured at multiple stations on nationwide air quality monitoring networks. The UK PAH

Network is a regulatory air quality monitoring network that discharges the UK Government's obligation under the EC Air Quality Fourth Daughter Directive to monitor the concentrations of B[a]P in the PM_{10} fraction (particles with an equivalent aerodynamic diameter of 10 μm or less) of ambient air.

2.2 AIR QUALITY POLICY

The EC Air Quality Framework Directive (Directive 96/62/EC) [6] set a strategic framework for tackling air quality in a consistent way by setting Europe-wide limit values for twelve air pollutants in a series of Daughter Directives. The Framework and first three Daughter Directives have been incorporated into the Ambient Air Quality and Cleaner Air for Europe Directive 2008/50/EC.

As discussed above, The Fourth Daughter Directive sets a target value for B[a]P (and, additionally, target values for arsenic, cadmium and nickel, and measurement requirements for mercury). The target values and assessment thresholds set by the Fourth Daughter Directive for B[a]P are:

- Target value (for the total content in the PM_{10} fraction averaged over a calendar year): 1 $ng \cdot m^{-3}$
- Upper assessment threshold: 0.6 $ng \cdot m^{-3}$.
- Lower assessment threshold: 0.4 $ng \cdot m^{-3}$.

The Fourth Daughter Directive also specifies the data quality objectives outlined in Table 1.

Table 1 Data quality objectives as specified by Annex V of the Fourth Daughter Directive. (Indicative measurements are defined as measurements which are performed at reduced regularity but fulfil the other data quality objectives)

Criterion	B[a]P in PM_{10}	Other PAHs in PM_{10}	PAHs in deposition
Expanded uncertainty (fixed and indicative measurements)	50 %	50 %	70 %
Expanded uncertainty (modelling)	60 %	60 %	60 %
Minimum data capture	90 %	90 %	90 %
Minimum time coverage (fixed measurements)	33 %	-	-
Minimum time coverage (indicative measurements)	14 %	14 %	33 %

The Fourth Daughter Directive also specifies requirements for the number and location of monitoring stations, by assessment of the B[a]P concentrations in the relevant zones and agglomerations within each member state.

The UK national air quality objective for B[a]P in ambient air is an annual mean concentration of 0.25 $ng \cdot m^{-3}$. This value was proposed in 1999 by the UK Expert Panel on Air Quality Standards [7] with the intention of reducing any risk to the population from exposure to PAHs to be so small as to be undetectable. The Panel also commented that it does not necessarily follow that all exposure above this standard carries a significant risk, in view of the application of an additional tenfold safety factor in deriving the standard. The annual mean concentration of 0.25 $ng \cdot m^{-3}$ for B[a]P was formally adopted in the UK Air Quality Strategy of 2007 [8].

2.3 SOURCES OF PAHS IN THE UK

Estimates of the quantities of PAHs emitted from the UK have been obtained from The National Atmospheric Emissions Inventory (NAEI; naei.defra.gov.uk), which is based on information in the UK Informative Inventory Report [9]. The NAEI contains data for the annual emissions of the 'EPA priority

16 PAHs'[‡] for every year since 1990. Throughout this section, the reader should be aware that all pollution emission measurements are subject to some uncertainty – in the case of PAHs, the NAEI estimates this uncertainty to be variable, but typically in excess of 60 %.

As discussed in the 2010 PAH Network annual report [10], it should be noted that all of the PAH emission factors used in the NAEI for road transport during the period up to 2009 were significantly revised following a thorough review [11] in early 2011. The PAH emission factors used for the domestic combustion of wood have also been significantly revised. Historical data has been updated to reflect these changes and the effect of this is that the emission estimates presented in this report cannot be compared with the data in the annual reports for any previous years.

As with most pollutants, PAH emissions in the UK have shown dramatic decreases over the last two decades as increasingly stringent emissions regulations and the decline of heavy industry have taken effect. Figure 2 shows that anthropogenic emissions of B[a]P have decreased remarkably over the last 24 years, such that the total B[a]P emissions in 2014 were approximately one-seventh of those in 1991.

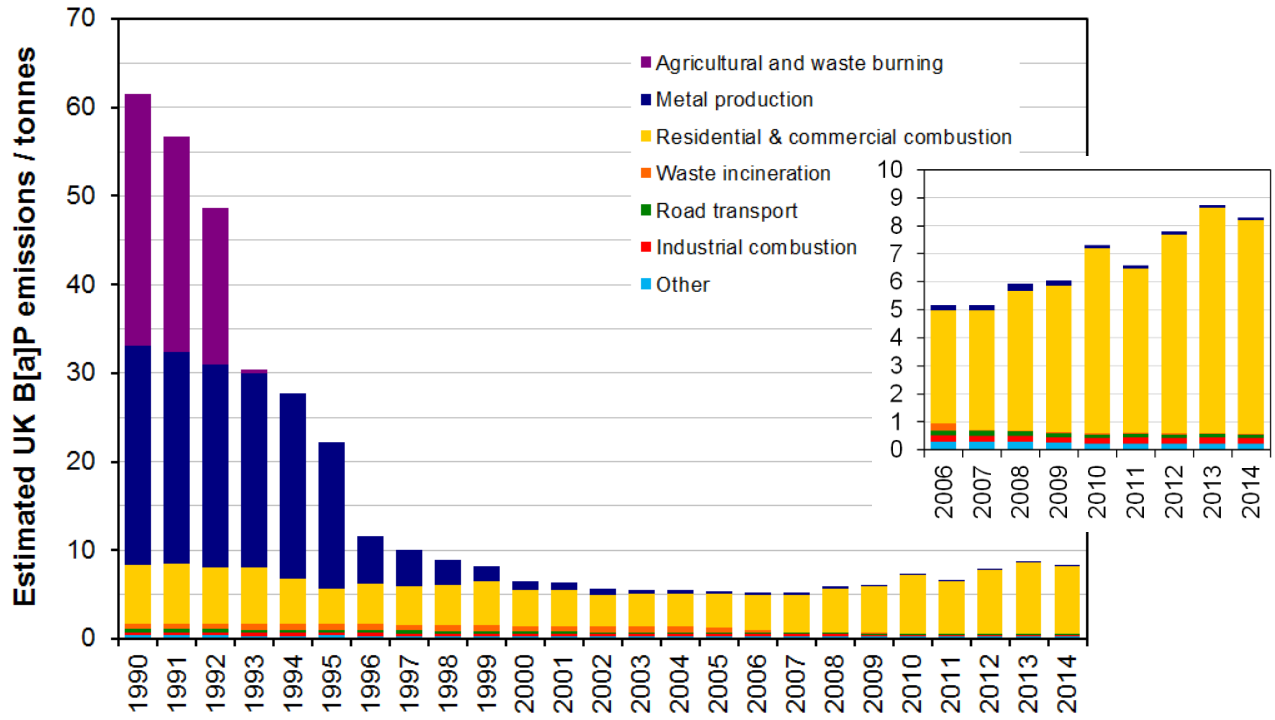

Figure 2 Estimated UK anthropogenic emissions of B[a]P per sector from 1991 to 2014 (main chart). The period from 2006 to 2014 is shown enlarged.

Emissions of B[a]P in the first half of the 1990s were dominated by emissions from metal production, and agricultural and waste burning. Emissions from metal production have declined significantly since the mid-1990s due to the closure of a number of metal processing plants, and the implementation of the Environmental Protection Act 1990, and now account for only 0.10 tonnes, or 1.2 % of the total anthropogenic UK B[a]P emissions. Emissions from agriculture and waste burning were effectively eliminated after 1992 due to the introduction of a ban on burning agricultural stubble.

[‡] The 'EPA (United States Environmental Protection Agency) priority 16 PAHs' are: acenaphthene, cenaphthylene, anthracene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene, chrysene, dibenz[a,h]anthracene, fluoranthene, fluorene, indeno[1,2,3-cd]pyrene, napththalene, phenanthrene and pyrene.

UK residential and commercial emissions of B[a]P dropped from 1991 onwards to a low point of only 3.47 tonnes in 2002, but have since risen again (see enlarged picture in Figure 2). This slight increase in recent years could be due to the use of solid fuel, such as wood for domestic heating. A recent study looked at the current level of wood burning in London and showed a contribution of 1.1 $\mu g \cdot m^{-3}$ to the annual average concentration of PM_{10}[12].

It is also informative to compare the estimated anthropogenic emissions with those from natural sources such forest fires, long-range transport from volcanoes, and other natural combustion events. Natural emissions have been estimated as a constant 2.88 tonnes per year during the time period discussed here (1991-2013). In 1990, natural emissions contributed only 4.5 % to total UK B[a]P emissions, but by 2006 this had increased significantly to 35.8 %. Since 2006, the slight increase in anthropogenic emissions has meant that the percentage of total PAH emissions that result from natural sources had decreased to 25.8 %

A recent report on PAHs in Northern Ireland [13] stated that in 2008, approximately 6.9 % of the UK's anthropogenic emissions of B[a]P originate from Northern Ireland, meaning that PAH emissions are significantly higher per head in Northern Ireland compared to Great Britain. This emphasises the significant contribution of solid fuel use in Northern Ireland to PAH emissions profiles.

The emission profiles of the other PAHs are not shown here, but are very strongly correlated to B[a]P, even though B[a]P only accounts for 0.6 % of the total emissions of the 16 PAHs considered by the NAEI. The total mass of anthropogenic UK PAH emissions decreased from 5,493 tonnes in 1990 to 1,342 tonnes in 2014.

3 THE NETWORK

3.1 NETWORK OBJECTIVES

The objective of the PAH Network is to determine the ambient concentrations of PAHs in ambient air in the UK through monitoring and chemical analysis, and therefore to deliver the following outputs:

- A UK-wide assessment of current concentrations of PAHs for assessment against the Fourth Daughter Directive and the UK Air Quality Strategy objectives, and provide information for future reviews of the Fourth Daughter Directive and UK Air Quality Strategy.
- Measurements and trends of airborne concentrations of PAHs in representative UK industrial, urban and other areas, and to compare and contrast them with those in rural locations.
- Data and metadata to demonstrate the UK's compliance with the Fourth Daughter Directive, the OSPAR convention [14] and the UNECE Convention on Long Range Transboundary Air Pollutants [15].
- Data for submission to the UK-AIR Information Resource (http://uk-air.defra.gov.uk).
- An improvement in the understanding of PAH source and emission estimates, and to examine their agreement with the National Atmospheric Emissions Inventory (www.naei.org.uk). To also input into estimates of atmospheric PAH concentrations, future projections, and the identification of the key areas of uncertainty.

3.2 NETWORK OVERVIEW

3.2.1 Stations

The Network in 2015 comprised of a maximum 31 stations operating at any one time, as specified in Annex A and shown in Figure 3. Twenty of these stations were in England, four in Scotland, four in Wales and three in Northern Ireland. Station activity and Network issues are discussed in more detail in Section 3.3.

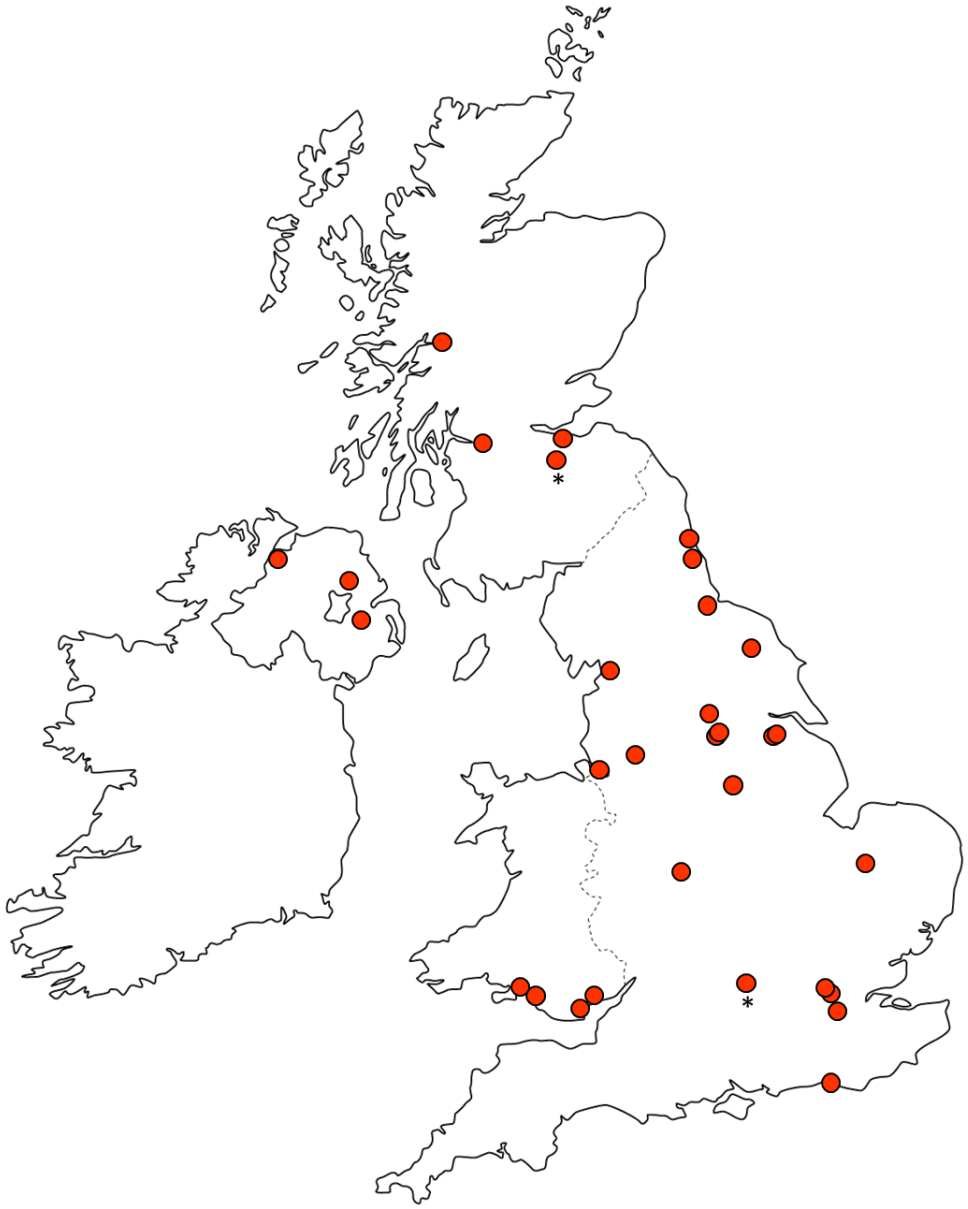

Figure 3 Schematic map showing the location of the Network stations (red circles). The two stations marked with asterisks are Auchencorth Moss and Harwell, where Digitel particulate and deposition samples are also taken (see Section 3.2.2).

3.2.2 Samples

A full description of the sampling equipment is given in Section 3.2.3. The Network requires the sampling and analysis of the following type of samples.

'A' (particulate) samples - samples taken from 'A' (Digitel filter only) stations. Samples of the PM_{10} fraction of ambient air on a filter. 'A' samples are taken daily at all 31 Network stations using Digitel samplers with automatic filter changers. Each sample is taken for 24 h, and a sample is taken every 24 h, with the sample changeover occurring at midnight. The samples are bulked into groups representing calendar months for analysis.

'C' (deposition) samples - samples taken from 'C' (deposition) stations. Deposition samples taken fortnightly at two Network stations (Auchencorth Moss C & Harwell C). Each sample is taken for 14

days, and a sample is taken every 14 days. The samples are bulked into groups representing a monthly time period for analysis.

3.2.3 Sampling equipment

'A' (particulate) samples: Digitel DHA-80 samplers (filter only)

The Digitel DHA-80 samplers (see Figure 4(a)) used throughout the Network are considered to be equivalent to the requirements of the European Standard for sampling PM_{10} matter (EN 12341). Evidence for this is available in a new version of the standard [16] which replaces the Standard published in 1998 [17]. These samplers are therefore valid for use with the European Standard method for the measurement of B[a]P in ambient air (EN 15549). Samples are taken onto quartz fibre filters for a period of 24 h at a flowrate of 30 $m^3 \cdot h^{-1}$.

'C' (deposition samples).

Deposition samples are taken at the Auchencorth Moss C & Harwell C stations using a deposition sampler (Figure 5) that meets the requirement of the European Standard for the measurement of the deposition of PAHs (EN 15980 [18]). The deposition samplers consist of a glass funnel and a four litre brown glass collection bottle, which are located inside a protective tube in order to minimise photochemical reactions and the degradation of PAHs. Spikes have been fitted to the top of the protective tubes to prevent damage and contamination by bird strikes. Deposition samples are taken for a period of two weeks.

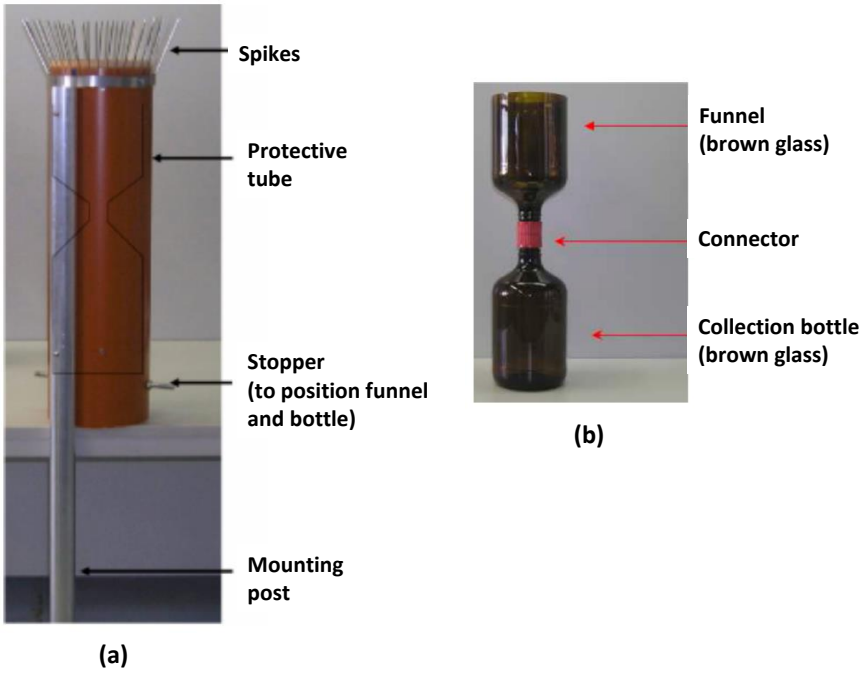

Figure 5 Photographs of (a) a deposition sampler; (b) a funnel and collection bottle (which are housed within the protective tube shown in (a))

3.2.4 Sampling quality control measures

In order to ensure the quality of the sampling procedure, the following quality assurance and quality control measures were undertaken:

- Analysis of sample blanks and filter blanks.
- Despatch and analysis of field blanks.
- On-going communication with and (where necessary) re-training of LSOs following NPL's annual station audit visits.
- Inspection of all sampling media prior to despatch to stations in accordance with the specifications in EN 15549 and EN 15980. For example, filters are inspected for pinholes, loose material and non-uniformity.
- Inspection of all filters when returned from stations. Samples that are damaged (*e.g.* torn or wet filters) or returned un-sampled are rejected and the details of these samples are noted in NPL's Network database.
- Rejection of any unidentifiable samples returned from stations.
- Full investigation of any discrepant or unexpected sampling data (*e.g.* unexpectedly high or low sample volumes).

3.3 NETWORK ACTIVITIES DURING 2015

3.3.1 Station infrastructure and Network re-organisation

The following network infrastructure changes took place in 2015:

- Scunthorpe Town was closed for a site upgrade (new cabin) from 18th August 2015 to 18th September 2015.

- Newcastle Centre was closed for a site upgrade (new cabin) from 30th July 2014 to 1st March 2015.
- Edinburgh St Leonards was reopened on 1st January 2015 after a site upgrade (new cabin) in 2014

3.3.2 Station audits and calibrations

The following station audit and calibration activities were carried out during 2015:

- Audits were carried out at all Network stations. As part of these visits, the local site operators (LSOs) were audited (and, where necessary, retrained).
- Routine servicing of the samplers was carried out at all Network stations.
- Quarterly calibrations of the flow rate of the samplers at all stations using NPL-designed calibration kits were carried out.
- Portable appliance testing, periodic (five-year) electrical testing and the updating of risk assessments were also performed at the stations as necessary.

3.3.3 Equipment servicing, breakdowns, and station maintenance

Full details of the breakdown and maintenance issues at each Network station during 2015 are given in Annex B. The main issues (data loss of more than 3 days) can be summarised as:

- Motors were replaced at six stations in total (Auchencorth Moss, Edinburgh St Leonards, High Muffles, Middlesbrough, Scunthorpe, South Hiendley).
- Data loss of 12 days at Edinburgh St Leonards in January due to a loading error.
- Data loss of 9 days at Lynemouth 2 in January due to filter changer jamming.
- Data loss of 6 days at Newport in February due to filter changer jamming.
- Data loss of 6 days at Newcastle Centre in March due to LSO programming error.
- Data loss of 9 days at Middlesbrough in May due to a motor failure.
- Data loss of 6 days at High Muffles in May/June due to a motor failure.
- Data loss of 7 days at Harwell in June due to a power failure.
- Data loss of 16 days at South Hiendley in August/September due to motor failure.
- Data loss of 11 days at Salford Eccles in September due to LSO absence.
- Data loss of 6 days at Kilmakee Leisure Centre in November due to a power supply issue.
- Data loss of 5 days at Newcastle Centre in November due to power failure.
- Data loss of 7 days at Derry Brandywell in December due to a wet filter blockage.

4 DATA CAPTURE

4.1 INTRODUCTION AND METHODOLOGY

The data capture results discussed in this section have been calculated assuming a target time coverage of 100 % (entire calendar year). The data capture is therefore the percentage of the year during which samples that ultimately contributed to the monthly average concentration data were taken. Samples that were rejected on return from the stations as un-sampled or damaged are given a data capture value of 0 %.

4.2 DATA CAPTURE VALUES

The annual average data capture values for 2015 are shown in Tables 2(a) – 2(b). The monthly data capture values (which are determined in the same manner as described above) are used to weight the monthly concentration data in order to calculate the annual mean concentrations.

Table 2(a) 2015 data capture values for "A" (particulate) samples

Station	Data capture / %	Station	Data capture / %
Auchencorth Moss A	99	Liverpool Speke	100
Ballymena Ballykeel	100	London Marylebone Road	100
Birmingham Tyburn	100	London Brent	100
Bolsover	100	Lynemouth 2	98
Cardiff Lakeside	100	Middlesbrough	98
Derry Brandywell	98	Newcastle Centre*	97 (81)
London Crystal Palace Parade	100	Newport	99
Edinburgh St Leonards	96	Port Talbot Margam	100
Glasgow Townhead	100	Royston	100
Harwell A	98	Salford Eccles	97
Hazelrigg	99	Scunthorpe Low Santon	99
High Muffles	98	Scunthorpe Town*	100 (92)
Hove	100	South Hiendley	95
Kinlochleven	100	Stoke Ferry	100
Leeds Millshaw	99	Swansea Cwm Level Park	100
Kilmakee Leisure Centre	98		
*station did not run for the entire year so the data capture is relative to the period of operation, not including planned closures for sampler relocation, and is excluded from the 'A-stations average'. The figure in brackets gives the data capture with respect to the whole year.		***'A' stations average:***	***99***

Table 2(b) 2015 data capture values for 'C' (deposition) samples

Station	Data capture / %
Auchencorth Moss C	96
Harwell C	100
'C' stations average	***100***

In 2015, all Network stations reported an annual data capture of more than 90% of the target data capture (excluding site upgrade at Newcastle Centre). The total annual average was 99%, compared to 97% in 2014 and 96% in 2013.

The largest contributor to data losses are hardware issues (mainly related to sampler motor failures, or jamming of the sampler automatic mechanism).

Historic data capture information, for the period 2010-2015 ('A' sites), is shown in Figure 7. The network consistently exhibits high data captures above 90% since 2011, with values as high as 98% achieved for the last seven quarters during 2014 and 2015.

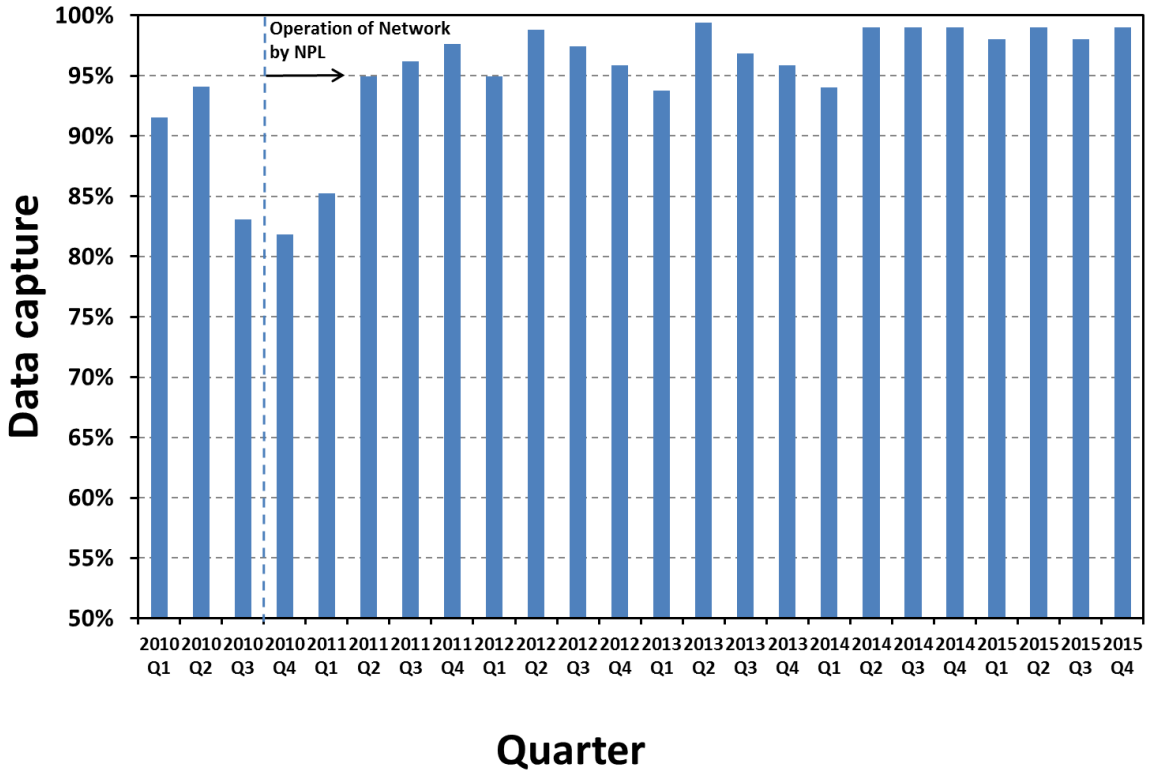

Figure 7 Average 2010-2015 quarterly data capture values for 'A' stations. NPL took over operation of the Network at the end of 2010 Q3.

5 ANALYSIS

5.1 PAHS MEASURED

The PAHs measured by the Network in 2015 for each sample type are indicated in red in Figure 8. The assessment of which PAHs to measure in each sample type was made following a study of co-located Digitel and Andersen samplers in 2007 [19].

PAH	Sample	
	A	C
Naphthalene		
2-Methyl napthalene		
1-Methyl napthalene		
Biphenyl		
Acenaphthylene		
Acenaphthene		
Fluorene		
Phenanthrene		
Anthracene		
2-Methyl phenanthrene		
2-Methyl anthracene		
1-Methyl anthracene		
1-Methyl phenanthrene		
9-Methyl anthracene		
4.5-Methylene phenanthrene		
Fluoranthene		
Pyrene		
Retene *		
Benzo[c]phenanthrene		
Benzo[a]anthracene		

PAH	Sample	
	A	C
Chrysene		
Cyclopenta[c,d]pyrene		
Benzo[b]naph[2,1-d]thiophene		
5-Methyl chrysene		
Benzo[b+j]fluoranthene		
Benzo[k]fluoranthene		
Benzo[e]pyrene		
Benzo[a]pyrene		
Perylene		
Indenopyrene		
Dibenzo[ah+ac]anthracene		
Benzo[ghi]perylene		
Anthanthrene		
Dibenzo[al]pyrene		
Dibenzo [ae]pyrene		
Dibenzo[ai]pyrene		
Dibenzo[ah]pyrene		
Coronene		
Cholanthrene		

* Retene is also known as 1-methyl-7-isopropylphenenthrene

Key
A = Particulate samples (Digitel)
C = Deposition samples

Figure 8 PAHs measured by the Network in 2015 (shown in red)

Note: By following the in-house NPL analyses for A samples, individual determination of Benzo(b)fluoranthene, Benzo(j)fluoranthene and Dibenzo[ah]anthracene, Dibenzo[ac]anthracene was achieved, compared to the combined Benzo[b+j]fluoranthene and Dibenzo[ah+ac]anthracene reported previously. Deposition 'C' samples were analysed externally using the same subcontracted service as in 2014, so results for Benzo[b+j]fluoranthene and Dibenzo[ah+ac]anthracene are given in this case.

5.2 ANALYTICAL TECHNIQUES

The analysis of Network 'A' (particulate) samples for the whole of 2015 was carried out at NPL in accordance with EN 15549 and accredited to ISO 17025. Samples were bulked into monthly batches (using only a specific portion/stamp of each filter) and spiked with a known quantity of surrogate standard solution. The samples were then Soxhlet extracted using a 1:1 v/v hexane/acetone solution as the extraction solvent. The extracts were reduced in volume to 0.3 ml, before undergoing a clean-up procedure using silica column solid phase extraction (SPE) chromatography. After clean-up, the sample was again reduced in volume to approximately 1 ml and analysed using gas chromatography – mass spectrometry (GC-MS). The GC-MS was calibrated using a series of calibration standards containing all the PAHs shown in Figure 8 at a range of concentrations covering the concentrations expected in the samples. The relative expanded uncertainty in a typical analytical result from a Network sample was estimated to be 25 % - well within the data quality objective of an expanded uncertainty of 50 %.

The limits of detection of the GC-MS method were determined by running a series of filter blanks every six months, and a solvent blank with every analytical run.

Finally, analysis of 'C' (deposition) samples was carried out in accordance with EN 15980, but this time using a sub-contracted service from a commercial analytical laboratory accredited to ISO 17025, similarly to 2014. In summary, the deposition samples were bulked into four weekly batches and extracted using liquid-liquid extraction. The resulting extract was dried, reduced to a volume of approximately 1 ml, and analysed using GC-MS as described above.

5.3 ANALYTICAL QUALITY ASSURANCE & QUALITY CONTROL MEASURES

In order to ensure the quality of the analytical data, the following QA and QC measures were undertaken:

- Regular assessment of the limit of detection of the GC-MS method.
- Regular extraction and analysis of an appropriate certified reference material (NIST SRM 1649b/ERM CZ100/NIST 2787) in order to check the recovery of the extraction methods. Recoveries must be within in the limits specified by EN 15549.
- Use of a range of deuterated PAH analogue species in order to assess (and correct for) the recovery of *each sample*. Following the criterion in EN 15549, if the analytical recovery of d_{12}-B[a]P for any sample was determined to be less than 50 %, the result was highlighted. For the in-house analysis, the extraction procedure was then repeated using a different portion of the respective filters, until satisfactory recovery was achieved.
- Use of an internal standard (d_{12}-Perylene), added to all samples and calibration standards to correct for short-term signal drift.
- The Network analysis laboratory (NPL, from 2012 onwards) participates in annual proficiency testing schemes to demonstrate ongoing competence and maintains accreditation for the analysis to ISO 17025.
- All data was assessed for outliers by analysis of the monthly and annual trends observed for each PAH at each station.
- The data was ratified by an NPL quality circle of senior scientific experts independent of the scientists responsible for processing the analytical data.

6 RESULTS & DISCUSSION

6.1 'A' (PARTICULATE) SAMPLES

This section presents and discusses the results from the 'A' (Digitel filter only) stations. The discussion focuses on B[a]P, but some data for other PAHs are also presented. Data for all PAHs for all stations are made available on the UK-AIR website (http://uk-air.defra.gov.uk/).

6.1.1 B[a]P annual concentrations and comparison against target values and air quality objectives

The annual mean B[a]P concentration measured at all 'A' stations is shown in Figure 9.

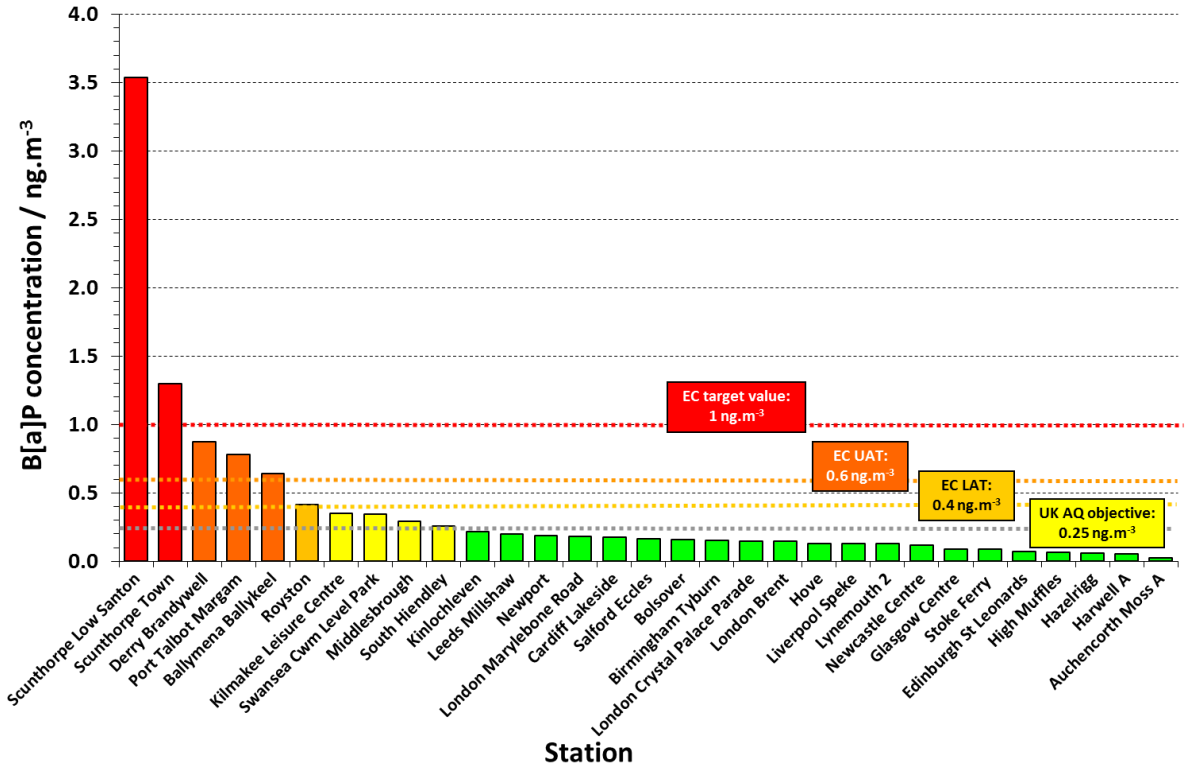

Figure 9 Annual mean B[a]P concentrations recorded at 'A' stations during 2015. The colour coding of the data from each station indicates that the mean concentration is:

Red: in excess of the EC target value (1 ng.m^{-3});

Dark orange: in excess of EC upper assessment threshold (UAT; 0.6 ng.m^{-3}), but less than the target value;

Light orange: in excess of EC lower assessment threshold (LAT; 0.4 ng.m^{-3}), but less than the UAT;

Yellow: in excess of the UK air quality objective (0.25 ng.m^{-3}), but less than the LAT;

Green: below the UK air quality objective.

From Figure 9 it can been seen that:

- Two stations have a measured annual mean B[a]P concentration above the EC target value of 1 ng.m^{-3}, this the same number of stations as in 2014.

- A total of five stations have a measured annual mean B[a]P concentration above the EC upper assessment threshold of 0.6 ng.m^{-3}, which is a decrease of one stationcompared to 2014.
- A total of six stations have a measured annual mean B[a]P concentration above the EC lower assessment threshold of 0.4 ng.m^{-3}, a decrease from nine stations in 2014.
- Ten stations have a measured mean annual B[a]P concentration above the UK air quality objective of 0.25 ng.m^{-3}, a decrease from eleven stations in 2014.

Table 3 details the measured annual mean B[a]P concentrations at the stations where the EC lower assessment threshold of 0.4 ng.m^{-3} was exceeded in 2015 or 2014.

Station	B[a]P conc. / ng.m^{-3}		Station / area type [notes]
	2015	2014	
Scunthorpe Low Santon	3.54	3.62	Industrial / Urban
Scunthorpe Town	1.30	3.45	Industrial / Urban
Derry Brandywell	0.87	0.85	Background / Suburban [N. Ireland]
Port Talbot Margam	0.78	0.60	Industrial / Urban
Ballymena Ballykeel	0.64	0.79	Background / Urban [N. Ireland]
Royston	0.41	0.92	Industrial / Urban
Kilmakee Leisure Centre	0.35	0.41	Background / Suburban [N. Ireland]
Middlesbrough	0.29	0.49	Industrial / Urban
South Hiendley	0.26	0.44	Industrial / Urban

Table 3: Stations that recorded an annual mean B[a]P concentration greater than the EC lower assessment threshold of 0.4 ng.m^{-3} during either 2015 or 2014. The colours indicate the same exceedances as in Figure 9.

The six Network stations that exceeded the EC lower assessment threshold of 0.4 ng.m^{-3} during 2015 are all either industrial / urban stations in Great Britain, or background / urban or suburban stations in Northern Ireland with significant solid fuel use:

- *Scunthorpe Santon & Scunthorpe Town*: Downwind and upwind from steel works.
- *Royston* : Upwind from coke works.
- *Port Talbot Margam*: Next to steel works.
- *Ballymena Ballykeel and Derry Brandywell*: Stations in Northern Ireland with domestic solid fuel use.

6.1.2 B[a]P: Monthly concentrations

The full dataset showing the mean B[a]P concentration for each month at each 'A' Network station is shown in Annex C. The data are shown graphically in Figure 10.

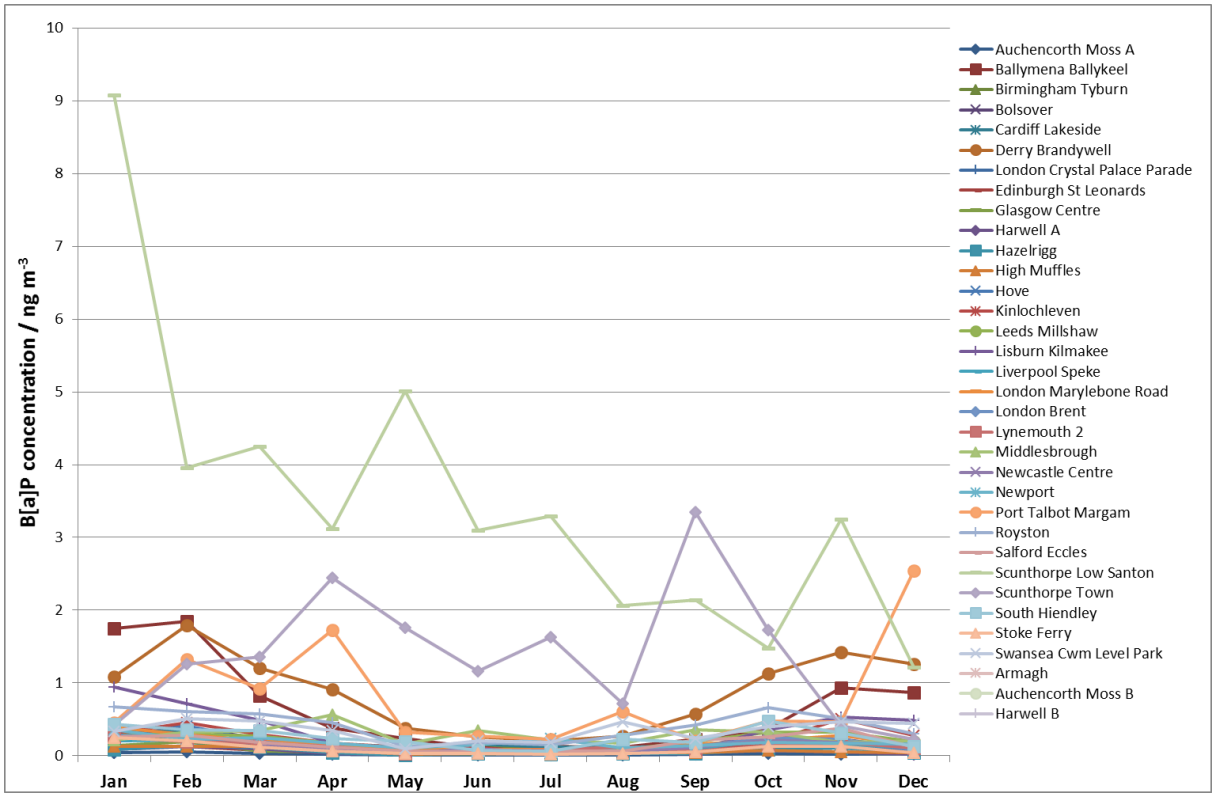

Figure 10 Monthly mean B[a]P concentrations recorded at 'A' stations during 2015

In general, PAHs are expected to show seasonal characteristics, with the highest concentrations being recorded at the beginning and the end of each year as a result of combustion processes peaking in winter. It is clear from Figure 10 that the two major industrial sites, Scunthorpe Low Santon and Scunthorpe Town, deviate from this pattern, with the second highest monthly mean B[a]P concentration (5.0 ng·m^{-3}) during 2015 observed at Scunthorpe Low Santon in May.

The annual variation in B[a]P concentrations at all stations can be observed and analysed in more detail if the data from stations with similar characteristics are plotted together. These plots are shown in Figures 11(a) to (d).

Figure 11(a) shows the monthly concentration of B[a]P measured at each of the Northern Irish monitoring stations in 2015, along with the monthly average temperature. It is evident that there is a very clear correlation of B[a]P emissions to temperature; one of the main contributory factors to this is thought to be the increase in domestic fuel heating during the winter months.

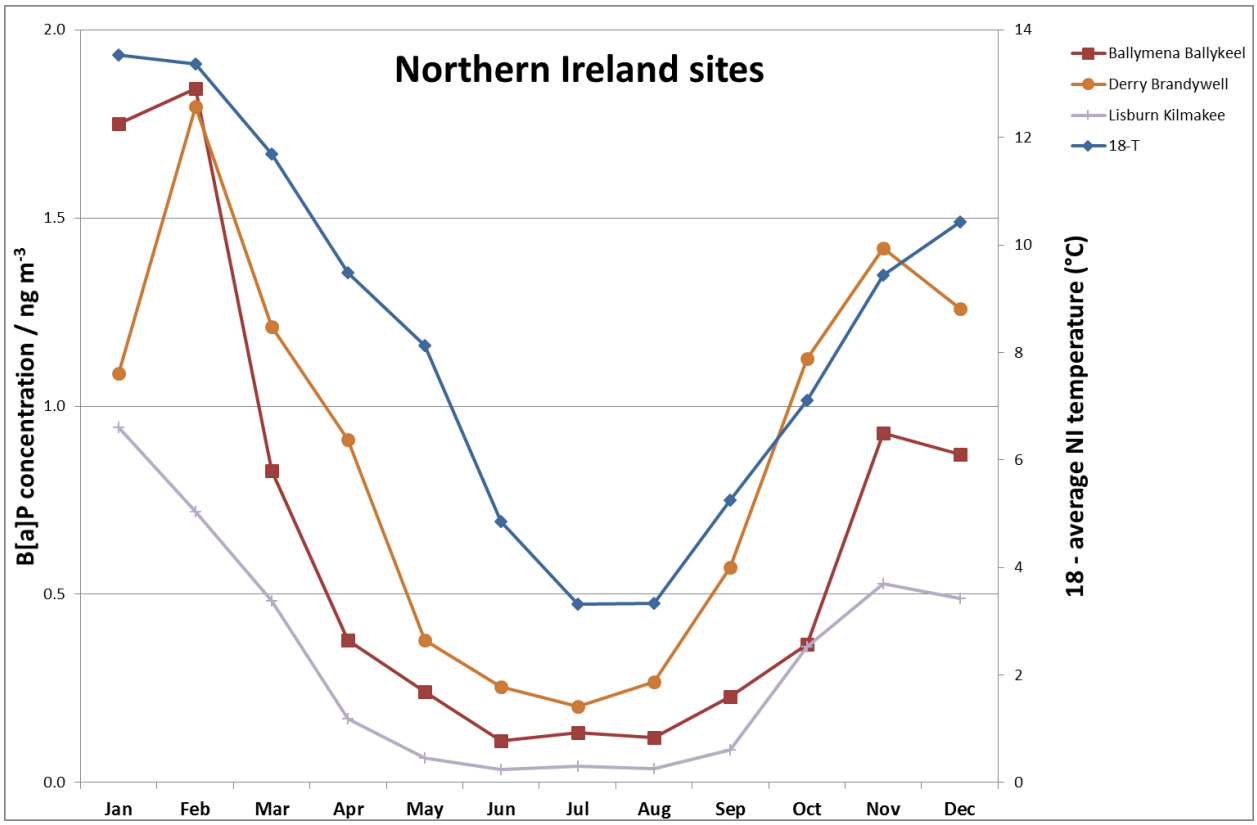

Figure 11(a) Monthly mean B[a]P concentrations recorded at background / urban stations in Northern Ireland during 2015, plotted together with the 2015 monthly average temperature (plotted as (18 - T) °C)

This monthly variability of B[a]P concentrations across the year (concentrations at their highest during winter months) is also exhibited at the background / urban and traffic / urban stations in Great Britain not influenced by major industrial processes (see Figure 11(b)) and the background / rural stations (see Figure 11(c)).

The B[a]P concentrations in November, however, were higher than the concentrations in October and December at most stations. One possible cause of higher emissions in November could be the emissions from bonfires and fireworks around the 5th November (Bonfire Night).

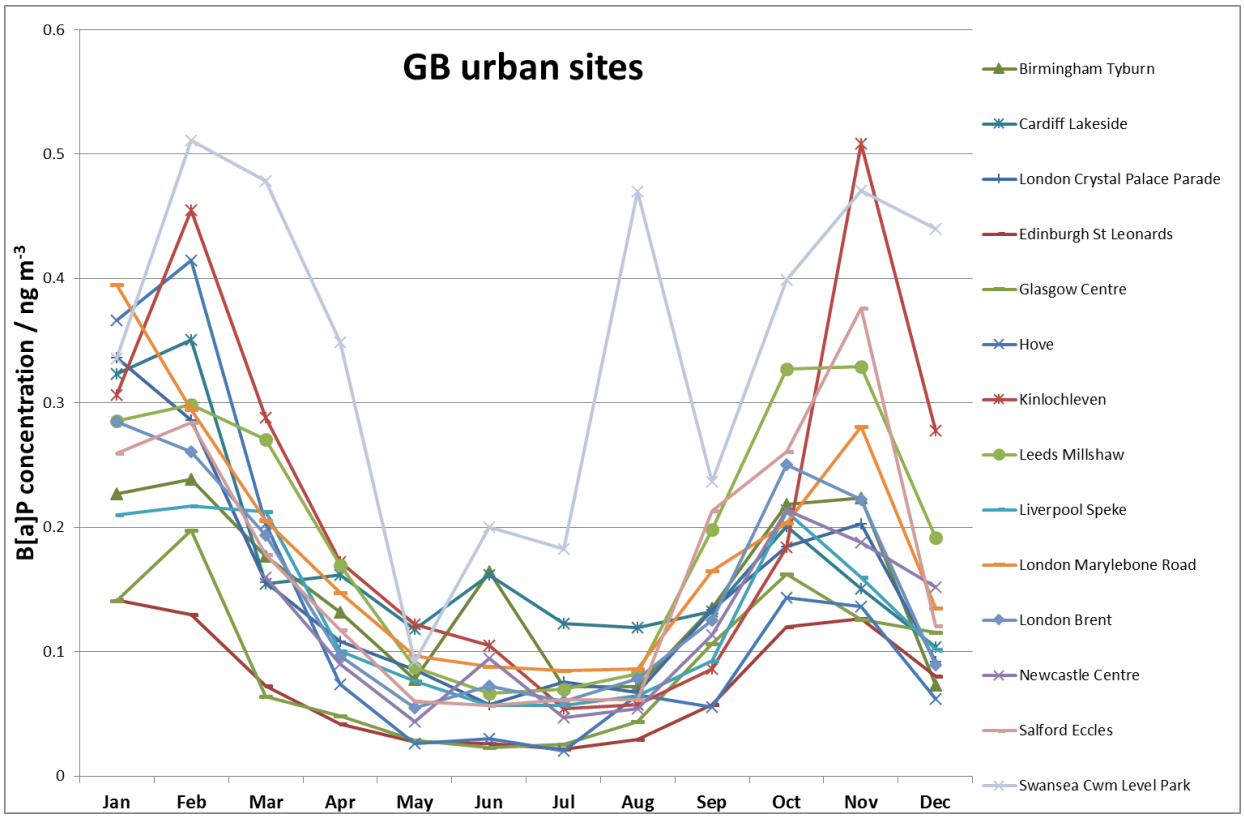

Figure 11(b) Monthly mean B[a]P concentrations recorded during 2015 at the traffic / urban (London Crystal Palace Parade and London Marylebone Road) and background / urban stations in Great Britain not influenced by major industrial processes (all others).

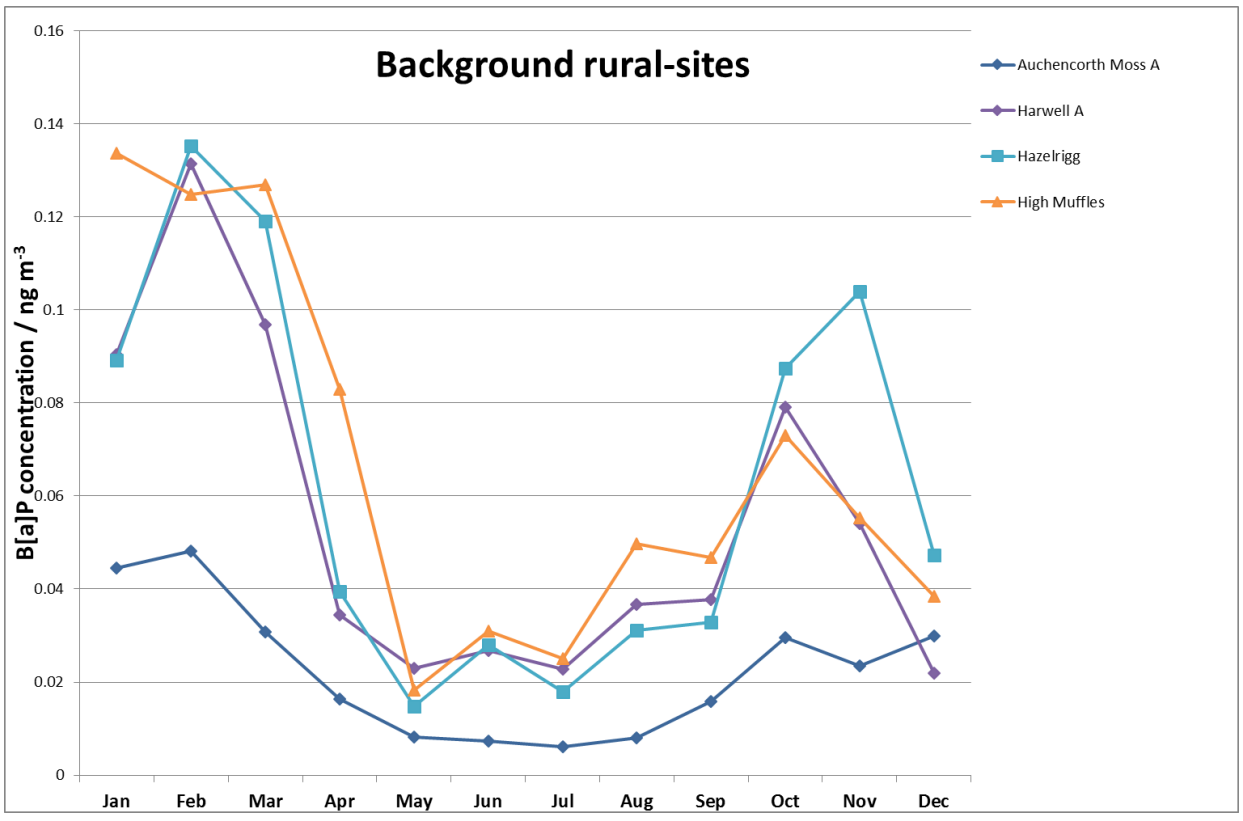

Figure 11(c) Monthly mean B[a]P concentrations recorded during 2015 at background / rural stations

In comparison, at the Network stations that are influenced by industrial processes (Figure 11(d)), very little seasonality is seen, showing that these measurements are dominated by industrial processes that are relatively invariant throughout the year. An intermediate level of seasonality is observed at the other stations in Figure 11(c).

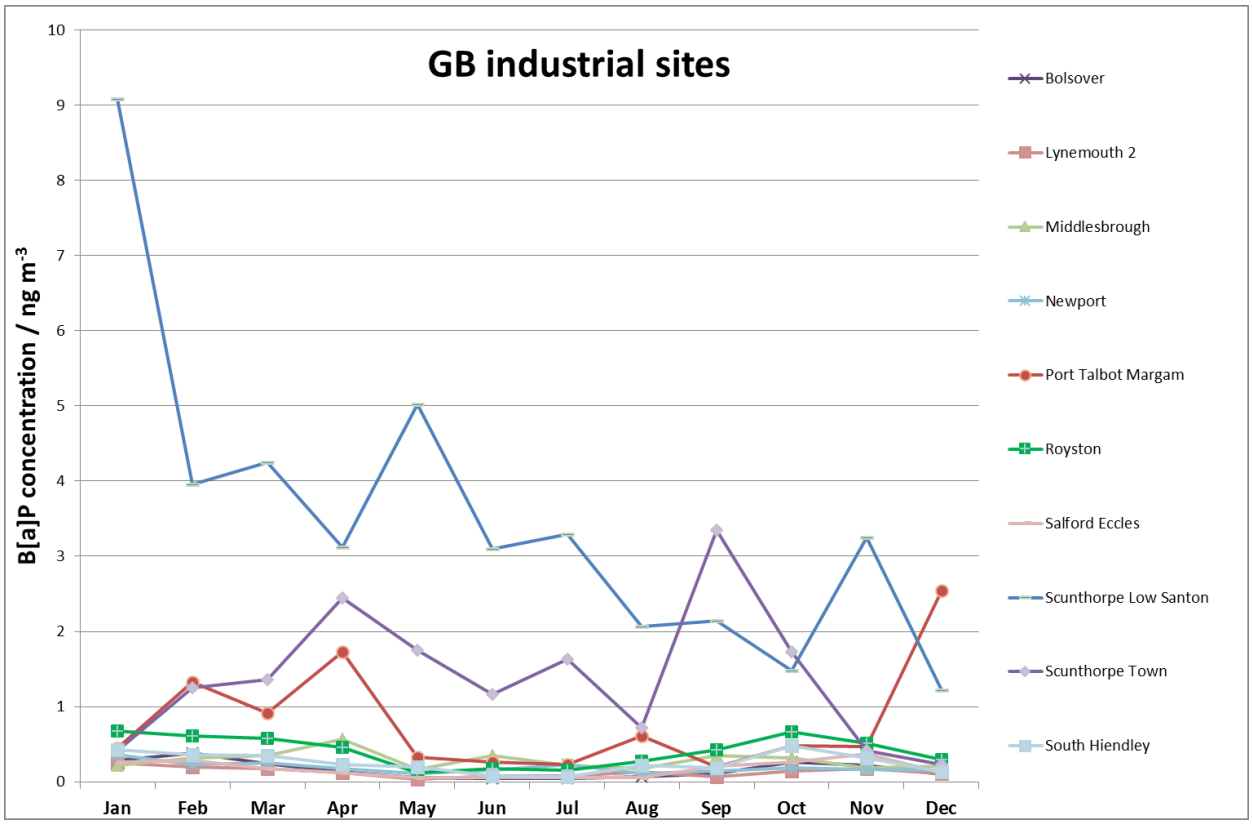

Figure 11(d) Monthly mean B[a]P concentrations recorded during 2015 at Network stations influenced by major industrial processes. The stations are: five industrial / urban stations (Midlesbrough, Port Talbot Margam, Royston, Scunthorpe Low Santon, Scunthorpe Town & South Hiendley), one industrial / suburban station (Lynemouth 2) and three background / urban stations in Great Britain that are influenced by industrial plants (Bolsover, Salford Eccles & Newport)

6.1.3 Other PAHs: Monthly concentrations

As discussed in Section 2.2, the Fourth Daughter Directive also specifies that (at least) the following six PAHs should be monitored at a limited number of measurement stations: benz[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene. CEN TC264 WG21 has developed Technical Specification for the measurement of the these PAHs and benzo[ghi]perylene in the particulate phase [5]. The Network measures all of these PAHs covered by the Technical Specification at all stations, as indicated below:

- Benz[a]anthracene: measured individually
- Benzo[b]fluoranthene: measured individually (previously measured as Benzo[b+j]fluoranthene)
- Benzo[j]fluoranthene: measured individually (previously measured as Benzo[b+j]fluoranthene)
- Benzo[k]fluoranthene: measured individually
- Indeno[1,2,3-cd]pyrene: measured individually
- Dibenz[a,h]anthracene: measured individually (previously measured as Dibenz[a,h+a,c]anthracene)
- Benzo[ghi]perylene: measured individually

The monthly mean concentration of each of these PAHs (or groups of PAHs) measured at all 'A' stations is shown in Figures 12(a)-12(g). It is worth noting that all these compounds follow a trend similar to the 'marker' B[a]P, giving high confidence in the respective analytical results.

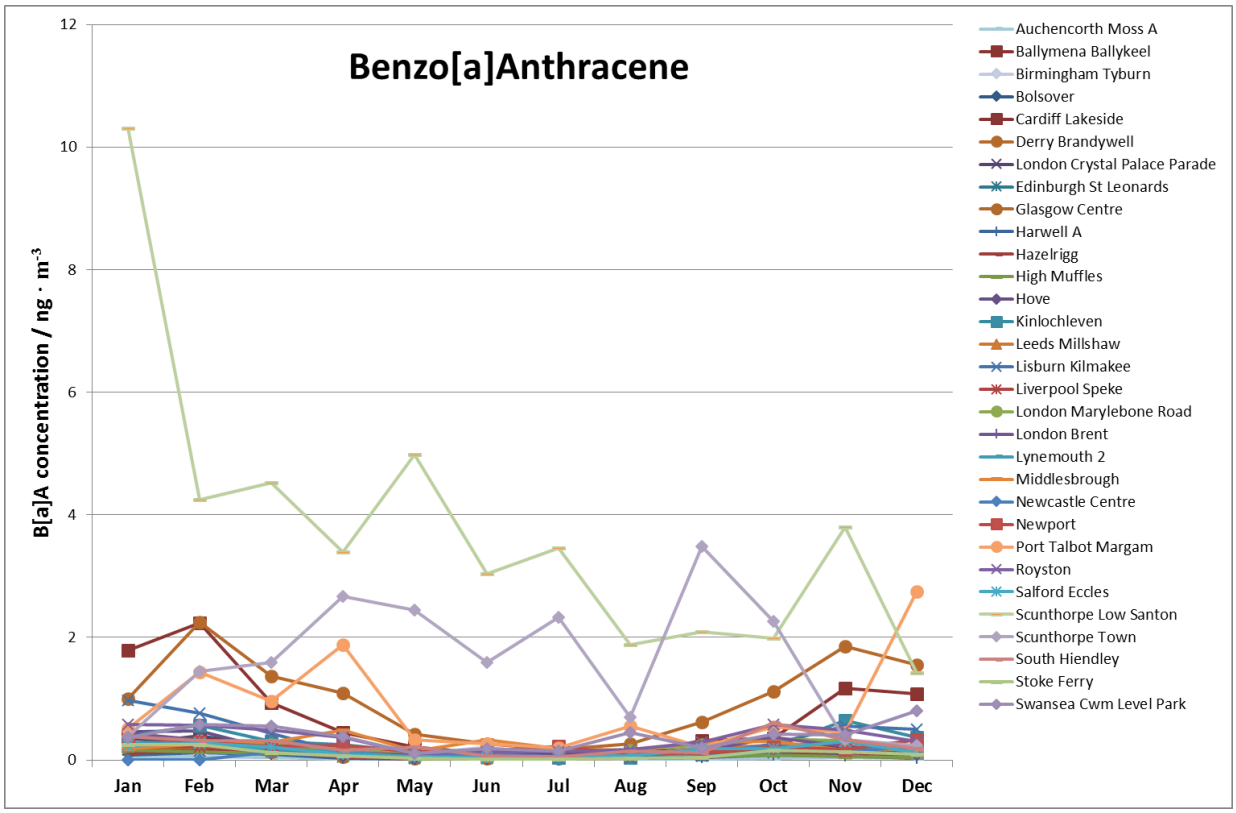

Figure 12(a) Monthly mean benzo[a]anthracene concentrations recorded at 'A' stations during 2015

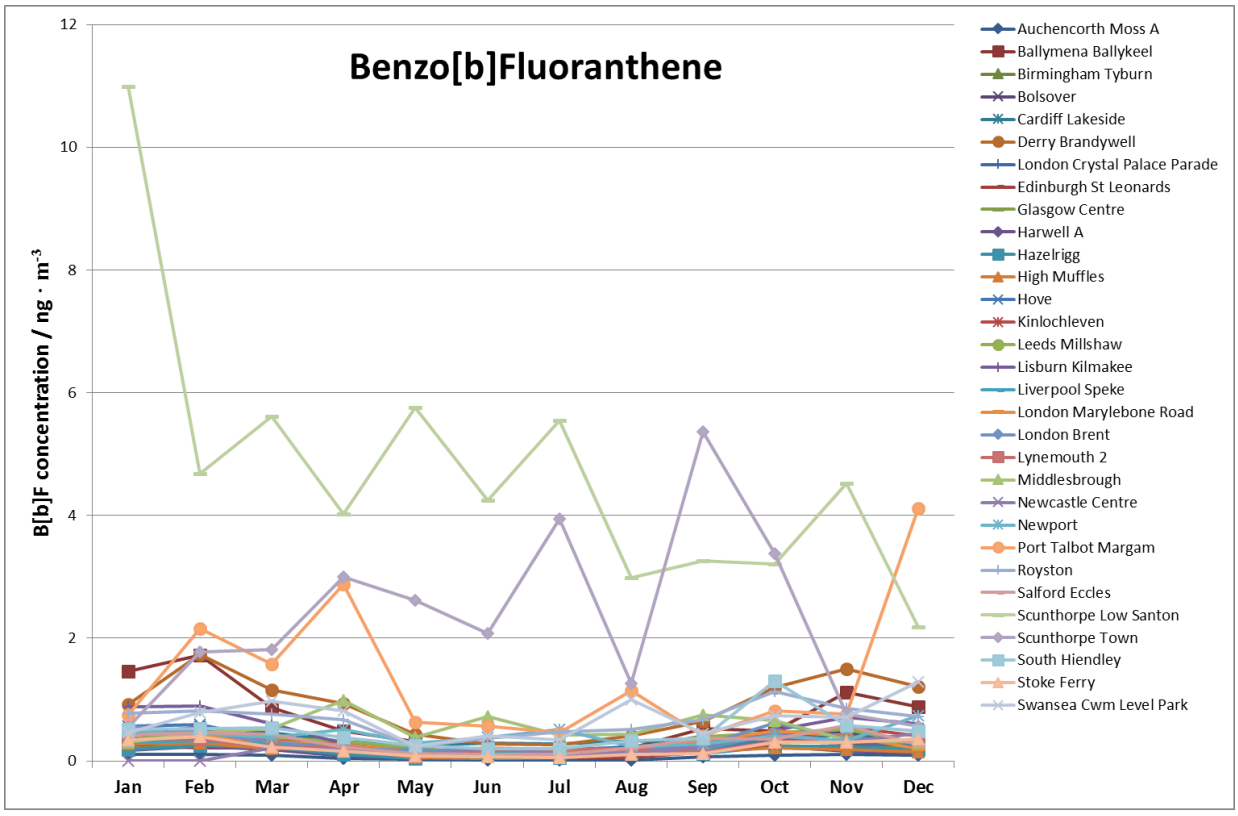

Figure 12(b) Monthly mean benzo[b]fluoranthene concentrations recorded at 'A' stations during 2015

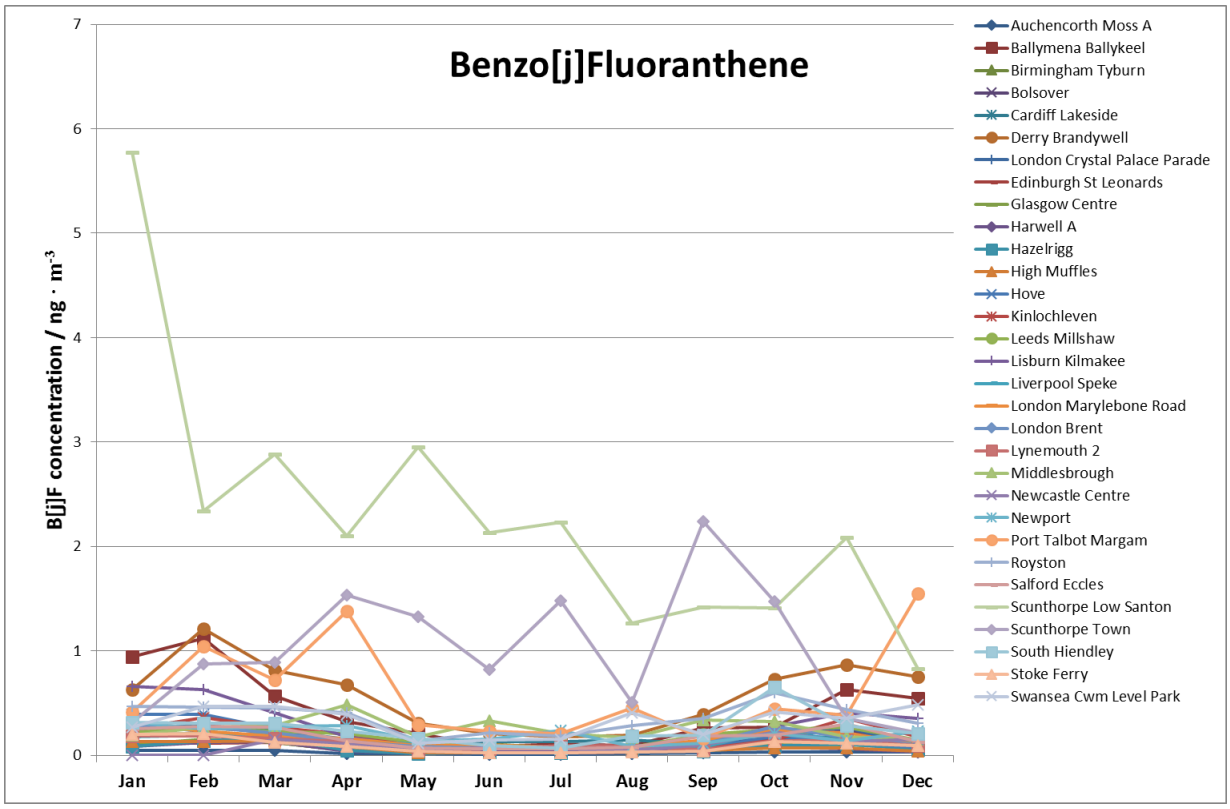

Figure 12(c) Monthly mean benzo[j]fluoranthene concentrations recorded at 'A' stations during 2015

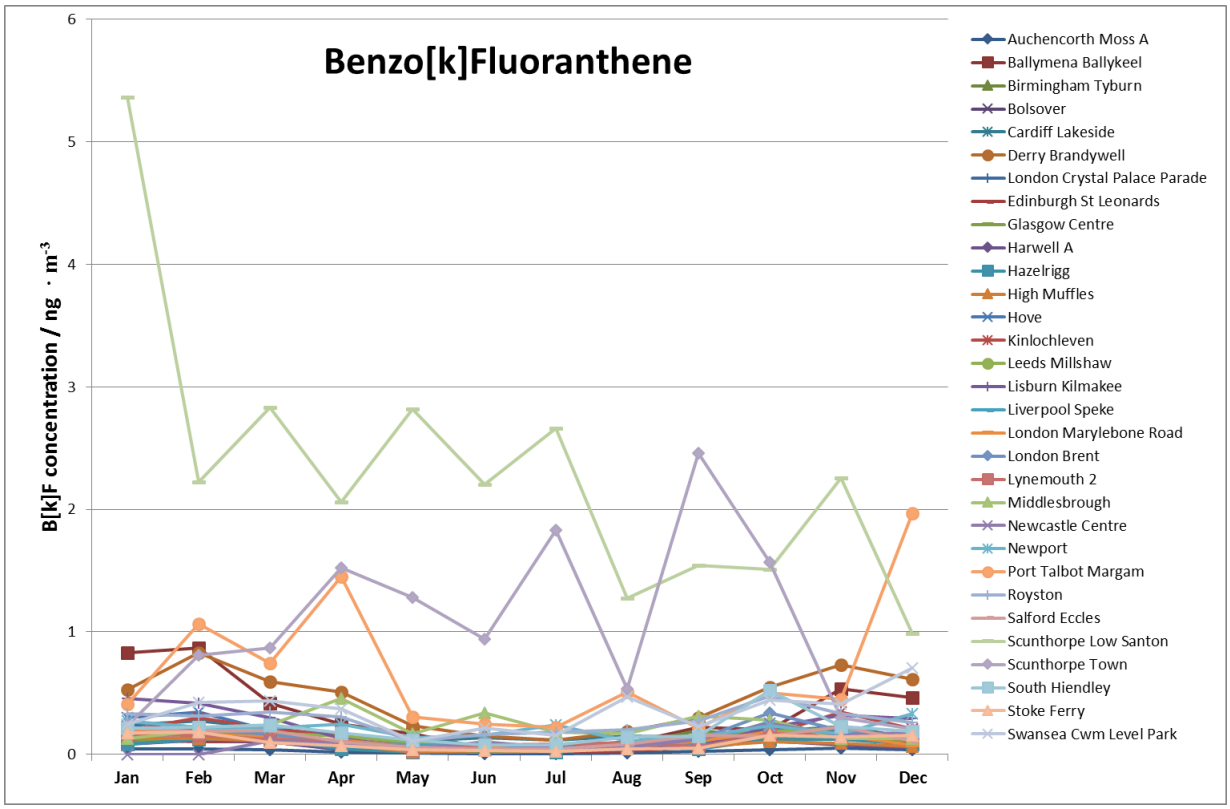

Figure 12(d) Monthly mean benzo[k]fluoranthene concentrations recorded at 'A' stations during 2015

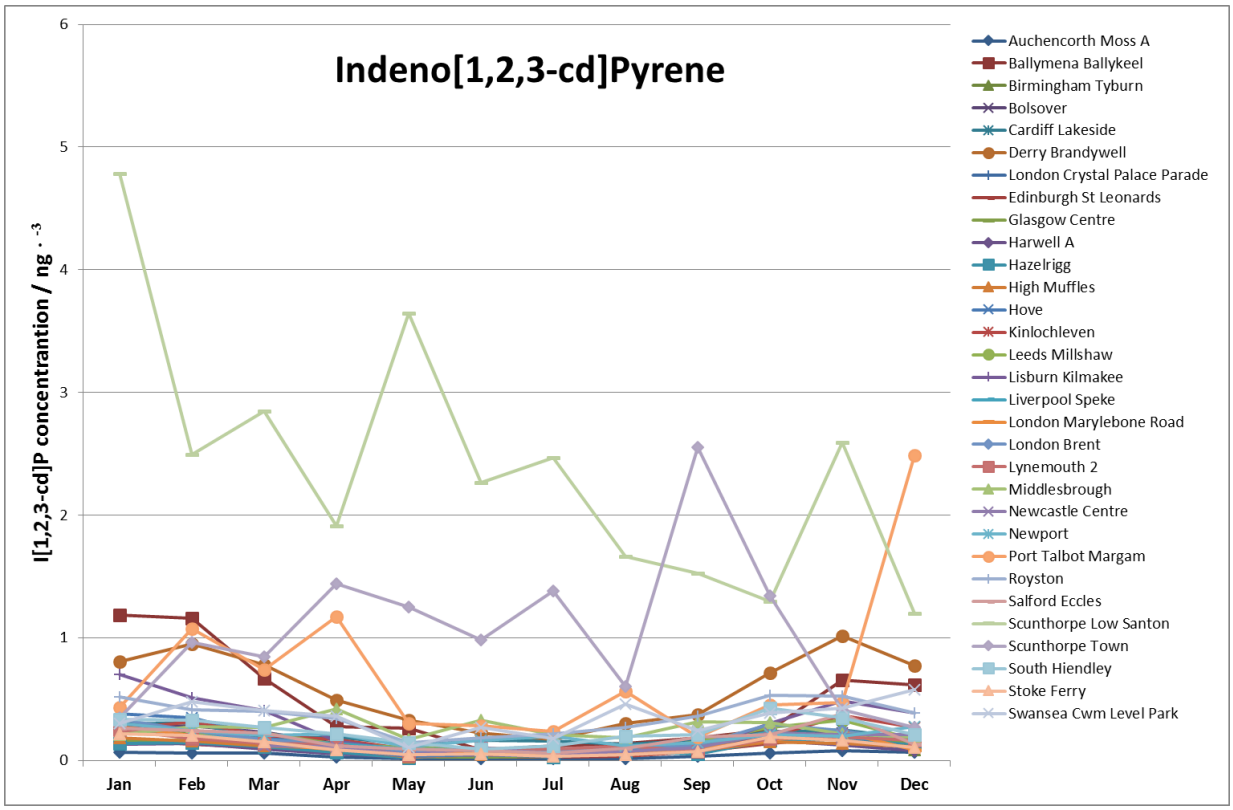

Figure 12(e) Monthly mean indeno[1,2,3-cd]pyrene concentrations recorded at 'A' stations during 2015

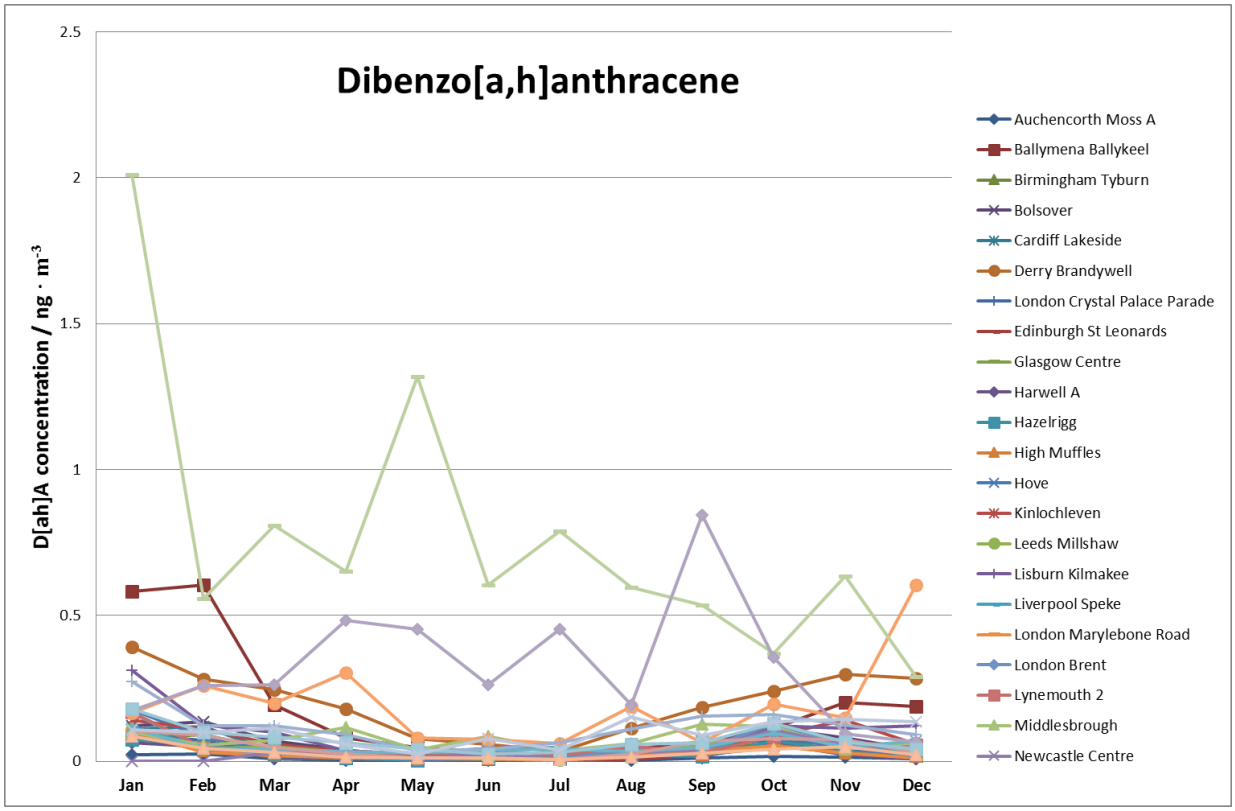

Figure 12(f) Monthly mean dibenzo[a,h]anthracene concentrations recorded at 'A' and 'B' stations during 2015

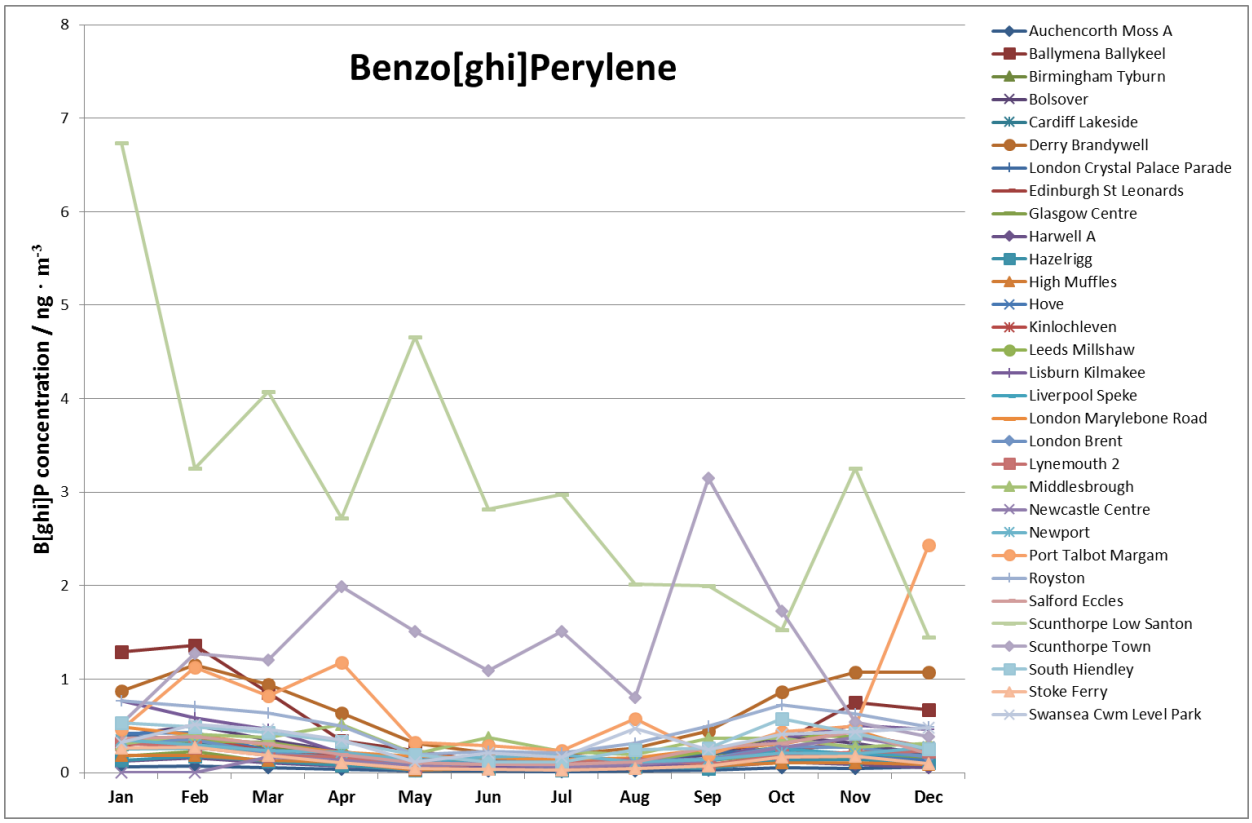

Figure 12(g) Monthly mean benzo[ghi]perylene concentrations recorded at 'A' and 'B' stations during 2015

6.2 DEPOSITION ('C') SAMPLES

The monthly and annual mean B[a]P concentrations measured at each deposition Network station are presented in Annex D.

The data for B[a]P, and for the other five PAHs (or groups of PAHs) covered by the European standard EN 15980 [18] show that, in general, background levels of PAHs in deposition have remained very low at both the Auchencorth Moss C and Harwell C stations during 2015, with the majority of results (more than 50 % at both sites) being less than the analytical limit of detection.

7 TRENDS IN MEASURED DATA

7.1 UK TRENDS IN B[A]P CONCENTRATION DATA (2007-2015)

Figure 17(a) shows the annual mean B[a]P concentrations measured at all 'A' Network stations since the installation of Digitel samplers (in 2007 or 2008). Figure 17(b) shows the same data for all stations (except all stations in Northern Ireland and those stations in Great Britain influenced by major industrial processes) using an expanded concentration scale on the y-axis.

7.2

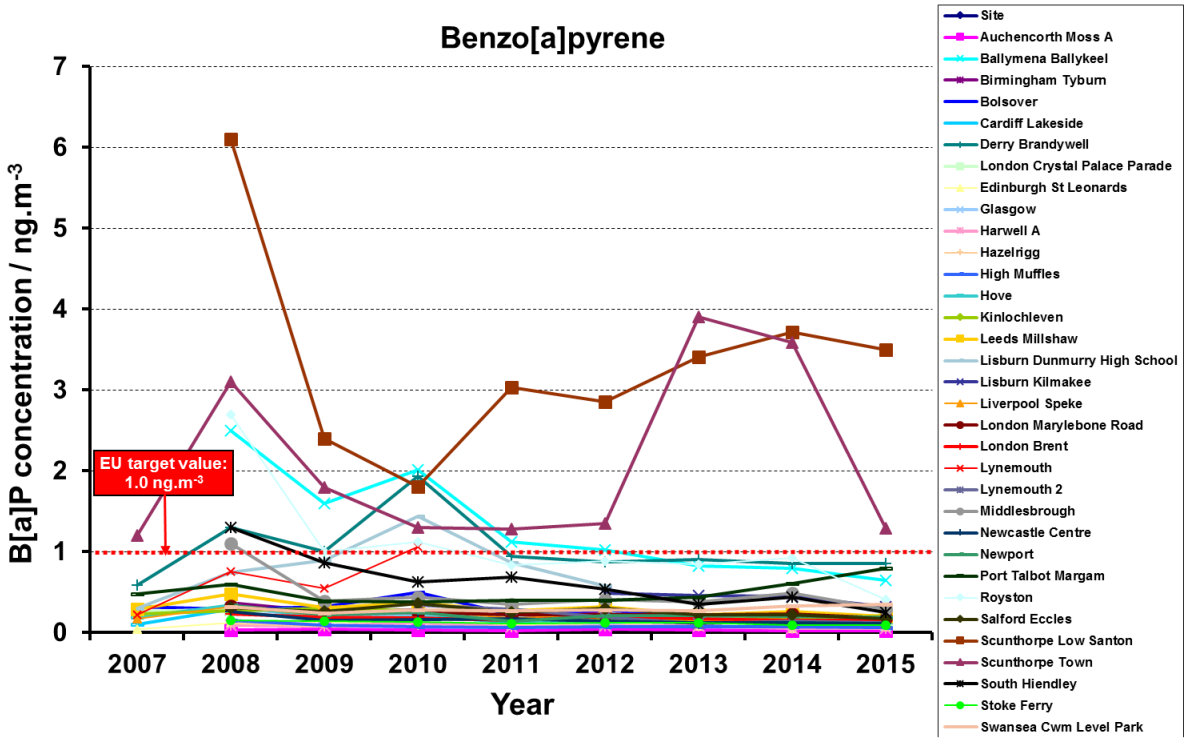

Figure 17(a) Annual mean B[a]P concentration measured at all 'A' stations from 2007-2015 inclusive

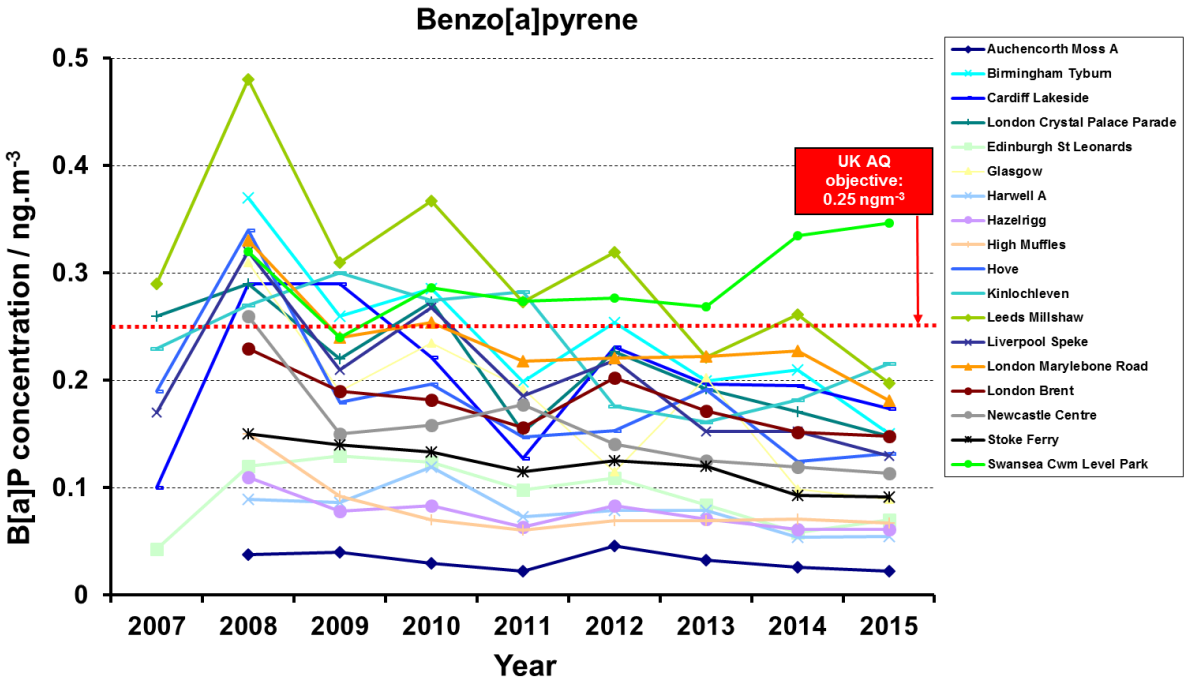

Figure 17(b) Annual mean B[a]P concentration measured at all 'A' stations (except all stations in Northern Ireland, and those stations in Great Britain influenced by major industrial processes) from 2007-2015 inclusive

It is clear from Figure 17(a) that with the exception of the Scunthorpe Town and Scunthorpe Low Santon sites, that the B[a]P annual average concentrations have in general decreased. With all other sites now falling below the EU target value of 1.0 ng m^{-3}. When the sites in Northern Ireland and those influenced by major industrial processes are removed in Figure 17(b) it can be seen that with the exception of Swansea Cwm Level Park all other sites have seen their B[a]P concentrations drop below the UK air quality objective of 0.25 ng m^{-3}. Swansea Cwm Level Park has an industrial component which can be seen by the peak in August 2015 in Figure 11(b). This may be due to PAH transport effects from the steelworks in Port Talbot.

8 OTHER ACTIVITIES RELATED TO THE NETWORK

8.1 STANDARDISATION & OTHER INTERNATIONAL ACTIVITIES

NPL continue to represent the UK's interests on CEN TC264 WG21 (measurement method for B[a]P in ambient air). The activities of WG21 during 2015 have included:

- The last draft of technical report for nitro- and oxy-PAHs was reviewed. The final changes will be implemented by the chairman and the secretary, before being sent to the CEN/TC.
- "Combination of EN 15549 and CEN/TS 16645" was discussed. A field evaluation will need to be conducted and costs obtained to discuss with CEN. The field evaluations will include:
    - Laboratory test: standard solutions, extract of real samples, solid SRM, real filters
    - Extraction methods: ASE, Quechers, ultrasonic, soxhlet and thermal desorption
    - Analysis: HPLC, GC-MS
    - Field test: 4 sites (rural north, rural south, urban, industrial)
    - Oxidant denuders: parallel sampling testing denuders like MnO_2 and active carbon
    - Data evaluation

8.2 PAPERS AND PRESENTATIONS

NPL has produced a number of articles in learned journals during 2015 that feature the data, analytical procedures and operation of the Network and research relevant to Network objectives. A particularly important study that was carried out in 2015 and published early in 2016 was:

Wavelength-dependent light absorption as a cost effective, real-time surrogate for ambient concentrations of polycyclic aromatic hydrocarbons
Brown, R J C; Butterfield, D M; Goddard, S L; Hussain, D; Quincey, P G; Fuller, G W
Atmospheric Environment, 2016, **127**, 125-132.

This study used data from a combination of the ultraviolet and infrared channels of aethalometers (referred to as UV BC), operated as part of the UK Black Carbon Network, as a surrogate measurement for PAH concentration. This established a relationship between concentrations benzo[a]pyrene (B[a]P) and the UV BC signal at locations where these measurements have been made together from 2008 to 2014. This relationship was observed to be non-linear. Relationships for individual site types were used to predict measured concentrations with, on average, 1.5% accuracy across all annual averages, and with only 1 in 36 of the predicted annual averages deviating from the measured annual average by more than the B[a]P data quality objective for uncertainty of 50% (at -65%, with the range excluding this value between +38% and -37%). These relationships were then used to predict B[a]P concentrations at stations where UV BC measurement are made, but PAH measurements are not. This process produced results which reflected expectations based on knowledge of the pollution climate at these stations gained from the measurements of other air quality networks, or from nearby stations. The influence of domestic solid fuel heating was clear using this approach which highlighted Strabane in Northern Ireland as a station likely to be in excess of the air quality directive target value for B[a]P.

ANNEX A: NETWORK STATION LOCATIONS

Station	Sample(s)	Longitude	Latitude	Start date	Type of station	Type of area
Auchencorth Moss	A, C	-3.24290	55.79216	Jan 08	Background	Rural
Ballymena Ballykeel	A	-6.25087	54.86160	Jan 07	Background	Urban
Birmingham Tyburn	A	-1.83058	52.51172	Jan 07	Background	Urban
Bolsover	A	-1.29708	53.25637	Dec 06	Background	Urban
Cardiff Lakeside	A	-3.16934	51.51241	Dec 06	Background	Urban
Derry Brandywell	A	-7.33213	54.99234	Dec 06	Background	Urban
London Crystal Palace Parade	A	-0.07553	51.42468	Jan 08	Traffic	Urban
Edinburgh St Leonards	A	-3.18219	55.94559	Mar 07	Background	Urban
Glasgow Townhead	A	-4.25516	55.85773	Dec 07	Background	Urban
Harwell	A, C	-1.32528	51.57108	Nov 07	Background	Rural
Hazelrigg	A	-2.77540	54.01364	Sep 07	Background	Rural
High Muffles	A	-0.80855	54.33494	Sep 07	Background	Rural
Hove	A	-0.18298	50.83659	Mar 07	Background	Urban
Kinlochleven	A	-4.96418	56.71445	Mar 07	Background	Urban
Leeds Millshaw	A	-1.57862	53.76611	Dec 06	Background	Urban
Kilmakee Leisure Centre	A	-6.00834	54.54376	Jul 12	Background	Suburban
Liverpool Speke	A	-2.84433	53.34633	Dec 06	Background	Urban
London Marylebone Road	A	-0.15461	51.52253	Feb 08	Traffic	Urban
London Brent	A	-0.27622	51.58977	Oct 07	Background	Urban
Lynemouth 2	A	-1.53674	55.21136	Sep 10	Industrial	Suburban
Middlesbrough	A	-1.22087	54.56930	Sep 07	Industrial	Urban
Newcastle Centre	A	-1.61053	54.97825	May 07	Background	Urban
Newport	A	-2.97728	51.60120	Apr 07	Background	Urban
Port Talbot Margam	A	-3.77082	51.58395	Oct 07	Industrial	Urban
Royston	A	-1.43945	53.60028	Sep 07	Industrial	Urban
Salford Eccles	A	-2.33414	53.48481	Oct 07	Background	Urban
Scunthorpe Low Santon	A	-0.59724	53.59583	Sep 07	Industrial	Urban
Scunthorpe Town	A	-0.63681	53.58634	Dec 06	Industrial	Urban
South Hiendley	A	-1.40084	53.61194	Oct 07	Industrial	Urban
Stoke Ferry	A	0.50615	52.55985	Sep 07	Background	Rural
Swansea Cwm Level Park	A	-3.93945	51.64584	Nov 07	Background	Urban

Sample codes
A: 'A' (particulate) samples
C: 'C' (deposition) samples

Start dates
The start date given for each station is the date at which sampling commenced using the present type of sampler.

ANNEX B: SUMMARY OF EQUIPMENT SERVICING, BREAKDOWN AND MAINTENANCE ISSUES DURING 2015

Station	Issue(s)
Auchencorth Moss A	• Motor replaced
Ballymena Ballykeel	----
Birmingham Tyburn	----
Bolsover	----
Cardiff Lakeside	----
Derry Brandywell	----
London Crystal Palace Parade	----
Edinburgh St Leonards	• Re-opened after site upgrade; motor replaced
Glasgow Townhead	----
Harwell A	----
Hazelrigg	----
High Muffles	• Motor replaced
Hove	----
Kinlochleven	----
Leeds Millshaw	----
Lisburn Kilmakee	----
Liverpool Speke	----
London Marylebone Road	----
London Brent	----
Lynemouth 2	----
Middlesbrough	• Motor replaced
Newcastle Centre	• Re-opened after site upgrade
Newport	----
Port Talbot Margam	----
Royston	----
Salford Eccles	----
Scunthorpe Low Santon	----
Scunthorpe Town	• Site upgrade; temperature probe replaced; motor replaced
South Hiendley	• Motor replaced
Stoke Ferry	----
Swansea Cwm Level Park	----

ANNEX C: MONTHLY & ANNUAL MEAN B[A]P CONCENTRATIONS – 'A' STATIONS

Station	Mean B[a]P concentration / ng.m^{-3}												
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Annual mean
Auchencorth Moss A	0.044	0.048	0.031	0.016	0.008	0.007	0.006	0.008	0.016	0.030	0.024	0.030	0.022
Ballymena Ballykeel	1.751	1.844	0.829	0.378	0.242	0.111	0.132	0.118	0.228	0.367	0.929	0.872	0.644
Birmingham Tyburn	0.227	0.239	0.177	0.132	0.077	0.164	0.072	0.072	0.135	0.219	0.224	0.073	0.150
Bolsover	0.283	0.385	0.236	0.130	0.052	0.050	0.041	0.063	0.106	0.257	0.223	0.102	0.159
Cardiff Lakeside	0.324	0.351	0.155	0.162	0.118	0.162	0.122	0.120	0.132	0.201	0.151	0.104	0.174
Derry Brandywell	1.088	1.797	1.211	0.911	0.379	0.254	0.202	0.268	0.572	1.126	1.419	1.260	0.860
London Crystal Palace Parade	0.336	0.286	0.156	0.109	0.086	0.058	0.076	0.067	0.134	0.184	0.203	0.092	0.148
Edinburgh St Leonards	0.142	0.130	0.073	0.042	0.027	0.026	0.021	0.029	0.058	0.120	0.127	0.080	0.070
Glasgow Townhead	0.141	0.197	0.064	0.048	0.029	0.023	0.025	0.044	0.106	0.162	0.126	0.115	0.089
Harwell A	0.090	0.131	0.097	0.034	0.023	0.027	0.023	0.037	0.038	0.079	0.054	0.022	0.055
Hazelrigg	0.089	0.135	0.119	0.039	0.015	0.028	0.018	0.031	0.033	0.087	0.104	0.047	0.062
High Muffles	0.134	0.125	0.127	0.083	0.018	0.031	0.025	0.050	0.047	0.073	0.055	0.038	0.068
Hove	0.367	0.415	0.203	0.074	0.026	0.030	0.020	0.065	0.055	0.144	0.137	0.062	0.132
Kinlochleven	0.306	0.455	0.288	0.172	0.122	0.105	0.054	0.058	0.086	0.184	0.508	0.278	0.216
Leeds Millshaw	0.286	0.298	0.271	0.169	0.087	0.066	0.070	0.082	0.198	0.327	0.329	0.191	0.197
Kilmakee Leisure Centre	0.943	0.720	0.484	0.170	0.065	0.034	0.044	0.037	0.087	0.360	0.527	0.488	0.325
Liverpool Speke	0.210	0.217	0.213	0.100	0.076	0.057	0.057	0.065	0.093	0.212	0.160	0.102	0.130
London Marylebone Road	0.395	0.295	0.206	0.147	0.097	0.088	0.085	0.086	0.165	0.203	0.281	0.135	0.181
London Brent	0.285	0.261	0.193	0.096	0.055	0.072	0.060	0.078	0.125	0.250	0.222	0.089	0.148
Lynemouth 2	0.255	0.194	0.174	0.127	0.032	0.074	0.076	0.129	0.064	0.143	0.179	0.111	0.126
Middlesbrough	0.215	0.320	0.348	0.568	0.164	0.351	0.219	0.177	0.352	0.321	0.189	0.207	0.288
Newcastle Centre			0.160	0.090	0.044	0.094	0.047	0.054	0.113	0.214	0.188	0.152	0.113
Newport	0.361	0.226	0.248	0.171	0.110	0.149	0.233	0.109	0.140	0.185	0.170	0.158	0.188
Port Talbot Margam	0.452	1.329	0.918	1.727	0.327	0.267	0.232	0.605	0.203	0.476	0.466	2.542	0.792
Royston	0.670	0.608	0.575	0.453	0.126	0.179	0.154	0.279	0.426	0.664	0.515	0.298	0.411
Salford Eccles	0.259	0.284	0.178	0.117	0.060	0.057	0.061	0.061	0.213	0.261	0.376	0.120	0.168
Scunthorpe Low Santon	9.080	3.953	4.248	3.120	5.016	3.093	3.293	2.064	2.146	1.482	3.252	1.219	3.502
Scunthorpe Town	0.419	1.256	1.357	2.448	1.755	1.167	1.626	0.718	3.345	1.729	0.411	0.232	1.294
South Hiendley	0.430	0.360	0.347	0.233	0.197	0.079	0.069	0.224	0.187	0.476	0.317	0.133	0.256
Stoke Ferry	0.252	0.202	0.119	0.061	0.033	0.026	0.022	0.031	0.051	0.133	0.130	0.045	0.091
Swansea Cwm Level Park	0.336	0.511	0.478	0.349	0.092	0.200	0.182	0.470	0.236	0.399	0.471	0.440	0.346

ANNEX D: MONTHLY & ANNUAL MEAN B[A]P CONCENTRATIONS – 'C' (DEPOSITION) STATIONS

Period	Auchencorth Moss C			Harwell C		
	Start date	End date	Mean B[a]P ($ng.m^{-2}.day^{-1}$)	Start date	End date	Mean B[a]P ($ng.m^{-2}.day^{-1}$)
1	30/12/2014	28/01/2015	< 15	07/01/2015	04/02/2015	< 15
2	28/01/2015	25/02/2015	< 13	04/02/2015	04/03/2015	23
3	25/02/2015	25/03/2015	< 5.8	04/03/2015	01/04/2015	14
4	25/03/2015	22/04/2015	< 5.8	01/04/2015	29/04/2015	< 5.8
5	22/04/2015	20/05/2015	< 4.1	29/04/2015	27/05/2015	< 4.1
6	20/05/2015	13/06/2015	< 26	27/05/2015	24/06/2015	< 26
7	13/06/2015	15/07/2015	< 2	24/06/2015	22/07/2015	< 2
8	15/07/2015	12/08/2015	< 2.3	22/07/2015	19/08/2015	12
9	12/08/2015	09/09/2015	5.5	19/08/2015	15/09/2015	8.2
10	09/09/2015	07/10/2015	< 2	15/09/2015	14/10/2015	< 2
11	21/10/2015	04/11/2015	< 2	14/10/2015	11/11/2015	< 2
12	04/11/2015	02/12/2015	< 2	11/11/2015	09/12/2015	< 2
13	02/12/2015	30/12/2015	< 2	09/12/2015	08/01/2016	< 2
Annual mean	01/01/2015	31/12/2015	7.2	01/01/2015	31/12/2015	12.9

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