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REPORT

NPL REPORT AS 51

CPEA 28: Airborne Particulate Concentrations and Numbers in the United Kingdom (phase 2)

Annual Report - 2009

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NOT RESTRICTED

JUNE 2010

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ISSN 1754-2928

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Annual Report for 2009 on the UK Airborne Particulate Concentrations and Numbers Network

Executive Summary

This report was prepared by NPL as part of the UK Airborne Particulate Concentrations and Numbers contract (CPEA 28) let by the Department for the Environment, Food and Rural Affairs and the Devolved Administrations: the Scottish Executive; the Welsh Assembly Government; and the Department of the Environment in Northern Ireland. This is the Annual Summary Report for 2009 and contains:

- A summary of network operation and quality procedures
- A graphical presentation of all ratified network data from 2009
- Data capture per instrument per month
- Comparison of 2009 particle numbers and concentrations of sulphate, nitrate and carbon with levels in recent years
- Update on relevant policy areas
- Update of the context of the project research and of equipment in the field

All equipment was audited during the annual audit round in November/December 2009, and all instruments have been serviced and calibrated by the instrument manufacturer or Equipment Service Unit or NPL.

In addition to the measurement programme, short-term research projects have been commissioned on specific topics related to the measurement programme. The measurements have been and are being used by the University of Birmingham to gain further understanding of particulate matter, its sources, composition and possible control options. King's College London has concluded its volatile correction model and assessed methods to determine the oxidative potential of ambient samples.
These research projects are reported through separate Topic Reports and are not discussed further here. A list of the completed Topic Reports is, however, given.

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1 Introduction

This report was prepared by NPL as part of the UK Airborne Particulate Concentrations and Numbers contract (CPEA 28) with the Department for the Environment, Food and Rural Affairs and the Devolved Administrations: the Scottish Executive; the Welsh Assembly Government; and the Department of the Environment in Northern Ireland.

This is the Annual Summary Report for the UK Airborne Particulate Concentrations and Numbers Network for 2009 and contains:

- Summary of network operation and quality procedures.
- Data capture per instrument per month.
- Graphical presentation of all validated and ratified network data from 2009.
- Comparison of 2009 particle numbers and concentrations of sulphate, nitrate and carbon with levels in recent years.
- Policy update on relevant areas.
- Update of context of this project research and inventory of equipment in the field.
- Network-related publications.

In addition to the measurement programme, short-term research projects have been commissioned on specific topics. The measurements have been and are being used by the University of Birmingham to gain further understanding of particulate matter, its sources and possible control options. King's College London has concluded its volatile correction model and assessed methods to determine the oxidative potential of ambient samples. These research projects are reported through separate Topic Reports and are not discussed further in this report. A list of the completed Topic Reports is, however, given.

2 Network Operation

2.1 Overview

The operation of the network in 2009 was structured in the same way as the previous year. King's College London (KCL) has continued in its role as the Central Management and Control Unit (CMCU). It has carried out activities including routine collection of data from sites, 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 were performed by NPL and included site audits, instrument calibrations and data ratification.

2.2 Improvement to Network Operation

The EUSAAR (European Supersites for Atmospheric Aerosol Research) project is seeking to harmonise the measurement of particle number and size distributions across Europe. A EUSAAR representative audited the Harwell site in November 2007 and a number of recommendations were made. Following these recommendations, in October/November 2009 upgrade inlet systems were supplied for each SMPS and CPC instruments, which address the recommendations on the sampling inlet and regulating the humidity. The EUSAAR recommendations are likely to be incorporated in a CEN

standard for particles number concentration and size distribution measurements, as discussed in Section 5. Further detail is given in Section 4.4.4.

2.3 New instruments

An aethalometer from Magee Scientific was installed at Harwell on the 11th of November 2009. A description of the operation of the instrument, which is the same as those operated in the Black Smoke Network, can be found in Section 2.4.5.

2.4 Network Structure

The current measurement programme is shown below:

Site	Hourly $PM_{2.5}$ nitrate	Daily PM_{10} anions	Daily PM_{10} OC/EC	CPC	SMPS	Aethalometer
Birmingham Tyburn (Urban background)				x		
Harwell (Rural)	x	x	x	x	x	x
North Kensington (Urban background)	x	x	x	x	x	
Marylebone Road (Roadside)	x	x	x	x	x	

Table 2.1 Network Structure

Wind speed and direction are reported for Harwell and Rochester.
Site details are available through http://aurn.defra.gov.uk/stations/index.htm.

2.4 Instrumentation

A brief summary of the operation of the network instruments is given here. More detailed descriptions of the theory of operation, calibration and the estimated uncertainty in the results are included in the NPL measurement uncertainty report [NPL, 2007].

2.4.1 Particle Counting and Sizing Analysers

Particle number concentrations are measured using a Condensation Particle Counter (CPC). This works by saturating the sample with butanol in a heated tube, and then cooling the sample to set up supersaturated conditions. The butanol vapour then condenses on any particles present, causing them to grow large enough to be counted optically. CPCs are sensitive to particles from 7 nm up to several microns in size, and have a concentration range from zero to10^7 cm^{-3}. At lower concentrations, each particle is individually counted, and at higher concentrations an optical integrating mode is used.

Particle size distributions are measured using a Scanning Mobility Particle Sizer (SMPS). This consists of a CPC combined with an electrostatic classifier. The electrostatic classifier consists of a charge neutraliser (incorporating a Kr-85 radioactive source) and a Differential Mobility Analyser (DMA). The former brings the particles in the sample to a known steady state charge distribution and the latter allows particles of a single electrical mobility (a quantity related to particle diameter) to pass to the CPC. By varying the operating voltage of the DMA, the size of particles sent to the CPC can be varied and a size distribution obtained.

2.4.2 Automated Nitrate Analyser ($PM_{2.5}$)

Ambient samples are pulled through a $PM_{2.5}$ cyclone operated at 5.5 L/min. Particles are collected by humidification and impaction, and assayed in place by flash heating and chemiluminescent analysis of the evolved nitrogen oxide vapors.

The automatic nitrate analysers have been discontinued and are no longer supported by the manufacturer. In order to replace these instruments, an evaluation of two candidate replacements was carried at NPL. The replacement instruments were:

- The Particle into Liquid sampler (PILS), manufactured by Applikon [UK Agent – Metrohm
- The Ambient Ion Monitor (AIM), manufactured by URG [UK Agent - ET]

2.4.3 Inorganic Anions (PM_{10} Sulphate, Nitrate and Chloride)

Daily measurements of the inorganic components of PM_{10} (sulphate, nitrate and chloride) were made using a Thermo Partisol 2025 sequential air sampler. Ultrapure quartz filters (Pallflex Tissuquartz) were used to allow for the analysis of EC/OC and the inorganic components.

The Partisol sampler provides uninterrupted sampling of ambient air and automatic exchange of filters for up to 16 days. The instrument used airflow of 16.7 litres per minute through a PM_{10} inlet and the filter temperature was maintained to within ± 5 °C of ambient temperature. The exposed filters were stored in small polypropylene filter bags and kept in a cold room until analysis to prevent further loss of volatile components. Extracts from the filters were dissolved in an eluent of 3.5 mM sodium carbonate and 1 mM sodium hydrogen carbonate and analysed in the laboratory by ion chromatography, for sulphate, nitrate and chloride content. Ambient concentrations were derived from the mass measured on the filter and the airflow during the sampling period.

2.4.4 Elemental and Organic Carbon (PM_{10})

In the laboratory, a punch is taken from each filter and analysed for elemental and organic carbon in a procedure in which the measurand is method-defined. It involves heating the sample to remove the PM from the filter, conversion to methane, followed by detection by flame ionisation. In a helium atmosphere, the sample is gradually heated to 700°C to remove organic carbon on the filter. During this first phase there are usually some organic compounds that are pyrolitically converted to elemental carbon. Measuring the transmission and reflection of a laser beam through the filter continuously monitors this pyrolitic conversion and allows a correction to be made for it. Elemental carbon is detected in the same way after heating to 870°C in the presence

of oxygen and helium. The protocol used is termed Quartz, a close variation of the NIOSH protocol, with correction made using the transmitted signal.

2.4.5 Aethalometer ($PM_{2.5}$)

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 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 absorption coefficient α [m^{-1}] is calculated from the transmission, area and volume of the sample, and converted to a black carbon concentration, as a first approximation, using a mass extinction coefficient.

The aethalometer run on the Network operates at 2 wavelengths, 880nm and 370 nm. The 880nm wavelength is used to measure the Black Carbon (BC) concentration of the aerosol, while the 370nm wavelength gives a measure of the "UV component" of the aerosol. At wavelengths shorter than about 400 nm, certain classes of organic compounds (such as polycyclic aromatic hydrocarbons, and also certain compounds present in tobacco smoke and fresh diesel exhaust) start to show strong UV absorbance.

The UV component concentration is obtained by subtracting the measured BC concentration from the concentration measured by the 370nm source. 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 fictional material 'UVPM' is expressed in units of 'BC Equivalent'.

In this Network ambient air is drawn into the sampling system through a standard stainless steel rain cap mounted on the end of a vertical stainless steel tube. Size selection of the sampled aerosol is made by a $PM_{2.5}$ cyclone placed close to the inlet of the aethalometer. All of the tubing before the cyclone is constructed from stainless steel.

3 Data Quality

A summary of the principal quality-assurance and quality control procedures used during the measurement and ratification process is given below:

- Continued training of and regular communication with Local Site Operators (LSOs).
- The KCL Duty Officer is available to advise LSOs 365 days per year.
- Scheduled instrument services and calibrations are ongoing.
- An annual audit of all sites and instruments has been conducted by NPL.
- Calibration data produced at audit by the Equipment Support Unit (ESU), and regular calibrations carried out automatically or by the LSOs, are all used to produce an appropriate scaling factor to apply to the data.
- Field blank filters have been analysed to control the contamination due to the transport of the filters to the sites and back to the laboratory. However, no subtraction has been applied to the sampled filters.
- Routine maintenance is carried out on all instruments according to manufacturers' instructions.

- The Thermo 8400N nitrate analysers is calibrated with NO, which is certified at NPL and traceable to primary standards.
- The ESU is contracted to respond to breakdowns within 48 hours.
- Data collection is automated by the MONNET system at KCL.
- Automatic and manual data validation is followed by rigorous ratification procedures.
- Research into particulate mass, chemical composition and speciation continues at NPL under the Department for Business Innovation and Skills (BIS) Chem-Bio Programme.

Data quality circle meetings are held at least annually to review the data. This may lead to tracking back through the measurements and analytical procedures to confirm the validity of specific measurements. Other measurements made in this monitoring programme and in other Defra monitoring programmes will also be used to check the validity of the measurements.

3.1 Scheduled Instrument Services and Calibrations

The automatic nitrate and the Partisols are serviced twice yearly by the ESU, Air Monitors. The service procedure includes replacing old or worn parts, calibration of the NOx analyser, temperature and flow calibrations, leak tests and pump refurbishment. The services completed during 2009 are indicated in Table 3-1.

Site	Instruments	Service 1	Service 2
Harwell	Partisol Nitrate	18/11/2008	25/05/2009
Marylebone Road	Partisol Nitrate	27/02/2009	02/09/2009
North Kensington	Partisol Nitrate	27/02/2009	02/09/2009

Table 3-1: Equipment Services completed during 2008 for Partisol and Nitrate Analysers

Since January 2009, the CPCs have been serviced and calibrated at NPL. NPL received ISO 17025 accreditation for this calibration in 2008.

In January 2010 the SMPS instruments were also calibrated at NPL to allow the ratification of the 2009 data.

Site	05/06 service	06/07 service	07/08 service	**09 service**	**10 service**
Birmingham	July 05	August 06	October 07	**March 09**	**January 10**
North Kensington	April 06	March 07	April 08	**March 09**	**February 10**
Marylebone Road	September 05	October 06	November 07	**January 09(*)**	**February 10**
Harwell	-	-	January 08	**March 09**	**January 10**

Table 3-2: Annual CPC Service and Calibration Dates. In bold calibrations carried out at NPL. (*) Only 'as found' calibration

4 Network data

4.1 Instrument Performance And Concentration Data

The following sections discuss the different measurements made in the monitoring programme. The concentration data is also presented in a graphical format and unusual or interesting occurrences are noted and discussed.

Full, ratified data from 2009 will be provided to Defra's Air Quality Information Archive (www.airquality.co.uk) and will be used to form the basis of future topic reports, produced in collaboration with the University of Birmingham.

4.2 Inorganic Anions

4.2.1 Partisol 2025 Measurements (PM_{10})

Daily measurements of particulate sulphate, nitrate and chloride in the PM_{10} fraction were made at 3 sites during 2009 (Harwell, North Kensington and Marylebone Road). The extracts from the filters exposed were analysed by ion chromatography at NPL. The filter extracts are analysed for sulphate, as required by Defra, and also for nitrate and chloride.
The measurements of particulate sulphate, chloride and nitrate concentrations made in 2009 are displayed in Figure 4-1. All the concentrations are blank corrected.
Monthly data capture rates for the Partisol 2025 instruments in 2009 are given in Table 4-1. The data capture in September was low at North Kensington as the sampler had loss of data memory due to a fault that was then cleared by the ESU.
High chloride concentrations were found on December 19th and 20th at Marylebone Rd. Those days were characterised by snow and very cold temperature that caused formation of ice on the road. Resuspension of salt from the ground may have caused high concentrations of chloride in the air. However, this effect was not observed at North Kensington.

Site	Harwell	North Kensington	Marylebone Rd
January	52%	68%	97%
February	57%	96%	86%
March	100%	90%	100%
April	87%	97%	97%
May	77%	100%	90%
June	80%	93%	93%
July	71%	90%	100%
August	97%	71%	71%
September	97%	27%	87%
October	87%	100%	97%
November	90%	93%	87%
December	97%	94%	84%
Average	**83%**	**85%**	**91%**

Table 4-1: Monthly Data Capture for the Partisol 2025 Samplers during 2009

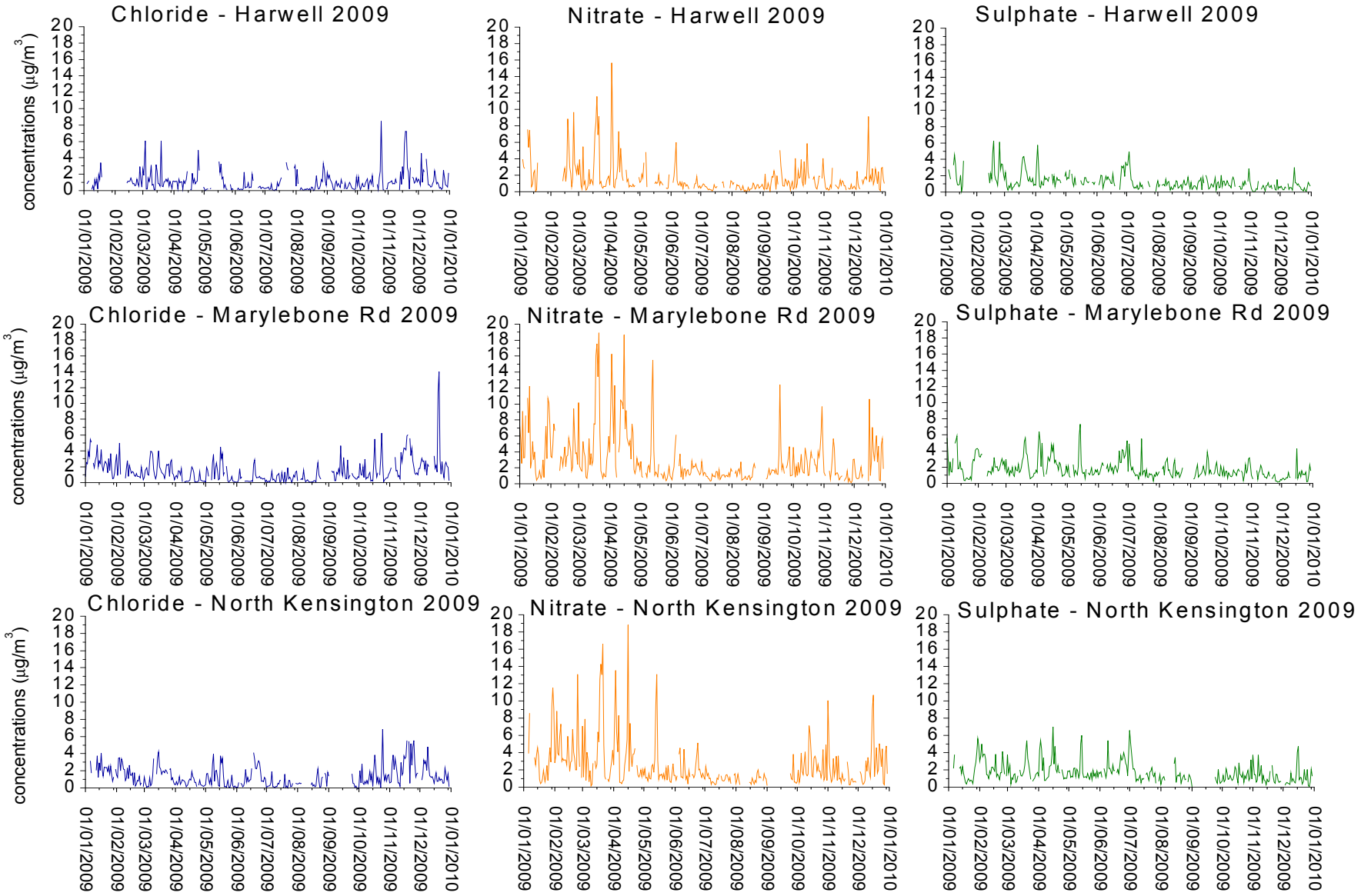


Figure 4-1: Partisol PM_{10} Anions Concentrations at Network Sites during 2009

4.2.2 Automatic Nitrate Analysers ($PM_{2.5}$)

The automatic daily measurements of particulate nitrate made in 2009 are displayed in Figure 4-2 and Figure 4-3 compared with PM_{10} Partisol nitrate concentrations.
Monthly data capture rates for the automatic nitrate instruments are presented in Table 4-2.

The ESU carried out services and instrument calibrations until September 2009 when the instruments were no longer supported by Air Monitors.

Site	Harwell	Marylebone Road	North Kensington
January	85.5%	86.4%	79.8%
February	87.2%	87.0%	84.3%
March	89.8%	98.9%	92.7%
April	49.7%	96.1%	96.2%
May	95.4%	99.3%	79.4%
June	87.2%	96.1%	95.3%
July	85.3%	97.0%	99.2%
August	99.5%	93.4%	94.1%
September	96.2%	93.3%	91.7%
October	99.1%	96.1%	46.5%
November	32.7%	94.9%	86.4%
December	85.5%	99.2%	39.7%
Average	**82.8%**	**94.8%**	**81.9%**

Table 4-2: Monthly Data Capture for the Automatic Nitrate Instruments during 2009

The analysers at North Kensington and Harwell were switched off respectively in December 2009 and January 2010 due to faults not fixable without the service by the ESU.

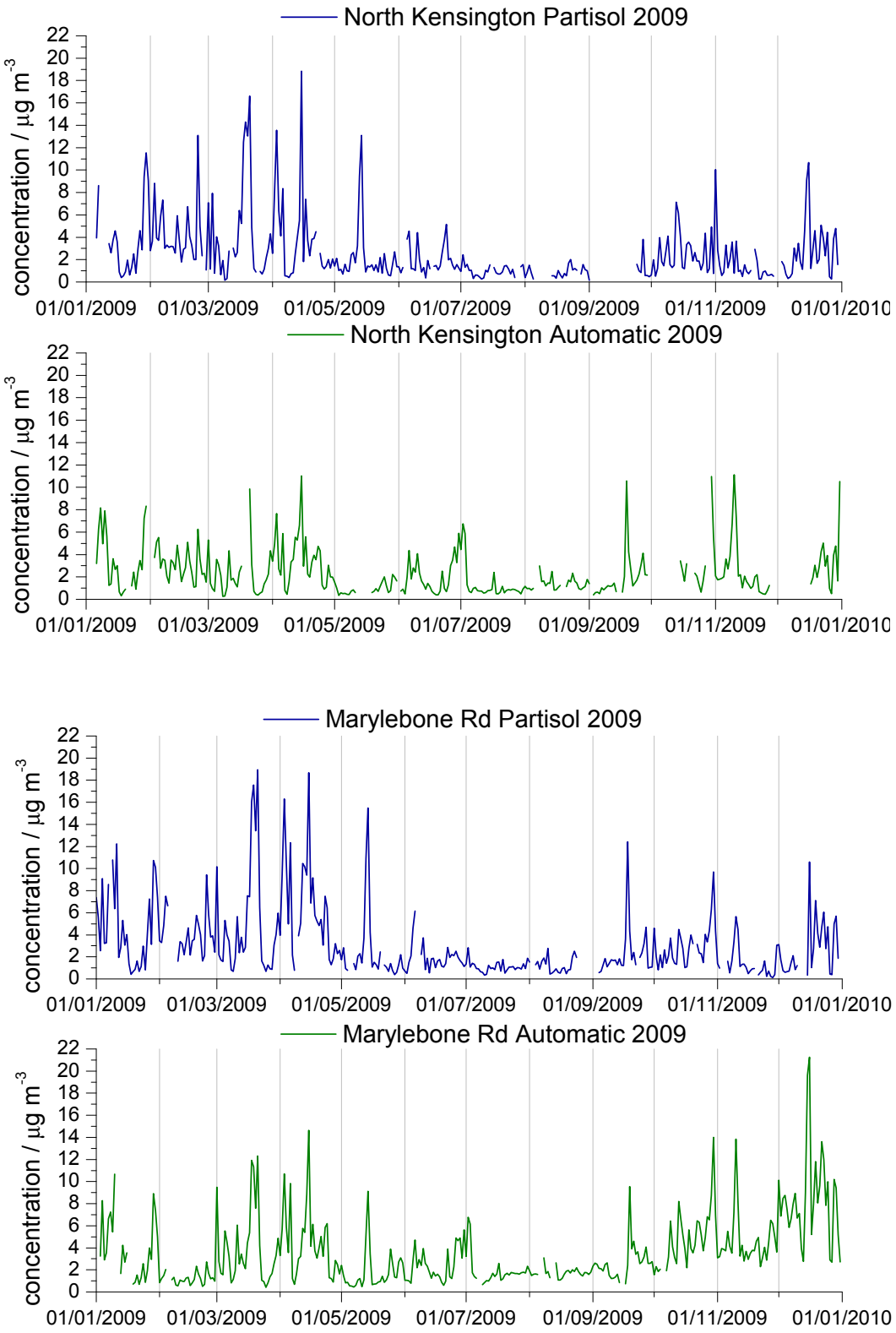


Figure 4-2: Comparison between automatic nitrate measurements ($PM_{2.5}$) and Partisol measurements (PM_{10}) at North Kensington and Marylebone Road in 2009

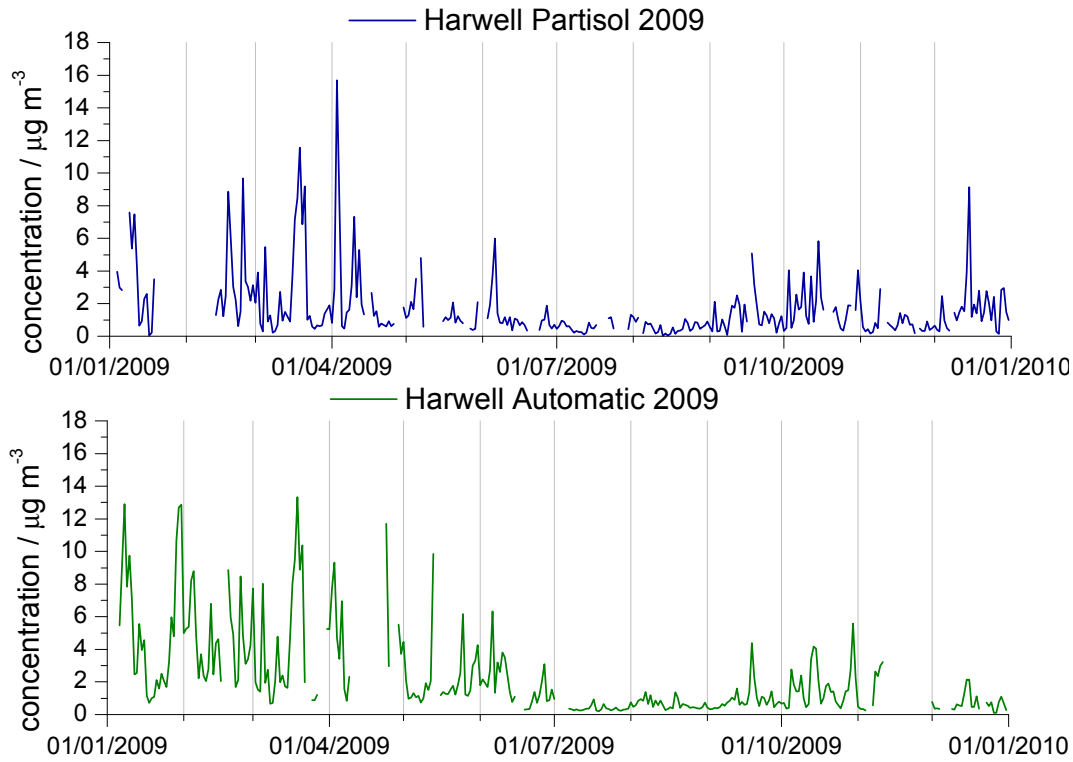


Figure 4-3: Comparison between automatic nitrate measurements ($PM_{2.5}$) and Partisol measurements (PM_{10}) at Harwell in 2009

4.2.3 Comparison of Automatic Nitrate ($PM_{2.5}$) and Partisol 2025 Nitrate (PM_{10})

Average daily concentrations of particulate nitrate have been derived from the hourly measurements made by the automatic analyser and these have been compared with the filter measurements of particulate nitrate.
Figure 4-4 presents scatter plots for each site. Only those days for which there are 20 or more hours of data have been included in the analysis.

Differences in the two measurement methods are expected to give rise to discrepancies in the results, and these are described in more detail in the NPL uncertainty report [NPL, 2007a]. As the measurements are from different PM size fractions (Partisol – PM_{10}, automatic nitrate – $PM_{2.5}$), a 1:1 correlation is not expected. Also, more sampling losses of volatile nitrate are expected from the daily Partisol method.

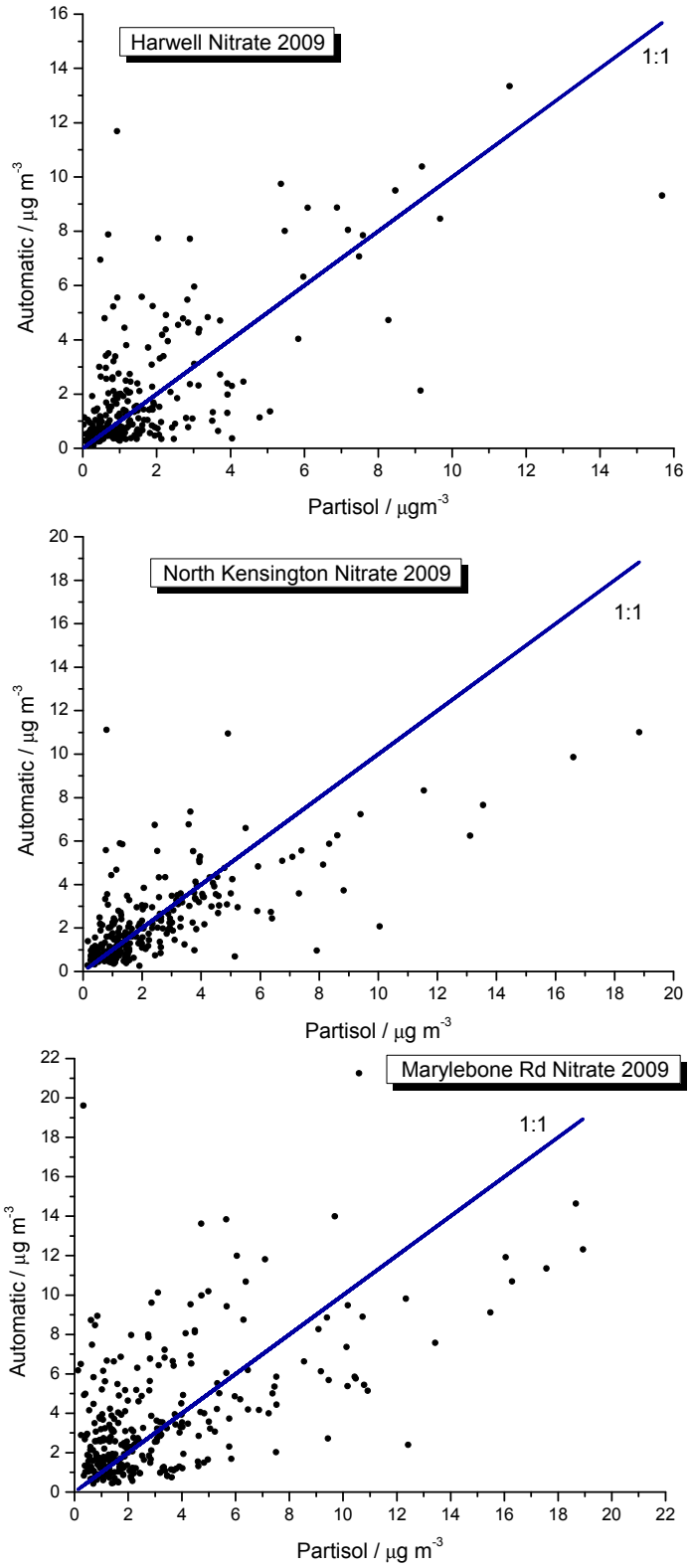


Figure 4-4: Scatter Plots of Nitrate Concentrations at the Network Sites during 2009

4.3 EC/OC Measurements (PM_{10})

4.3.2 Elemental and Organic Carbon

The filter measurements of EC, OC and TC (total carbon) made in 2009 have been combined and are displayed in Figures 4-5 for the 3 sites. All the concentrations are blank corrected.

4.3.3 Comparison between Elemental Carbon and Black Carbon

Using aethalometers, co-located measurements of black carbon ($PM_{2.5}$) have been made at the North Kensington and Marylebone Road sites, as part of the Defra Black Smoke Network [NPL report, 2009 Annual Report For The UK Black Carbon Network], and at Harwell as part of this Network (though only for the last few weeks of the year).
The time series of the elemental carbon (EC) and black carbon (BC) measurements have been compared and are shown in Figure 4-6 and Figure 4-7, as well as scatter plots.
The BC data collected at Harwell were not enough to make a significative comparison. The data capture for the period the instrument was operative is 67%.

The comparison for the North Kensington site shows very good agreement between the two techniques and presumably that the size fraction of PM in the elemental carbon contribution is mainly fine ($PM_{2.5}$). For the Marylebone Road site, from about the end of May to the end of December, however, BC concentrations are significantly higher than the EC concentrations when above about 10 $\mu g/m^3$. The reason for this step-change is not clear yet and it is under investigation. Historically there have been several other examples of anomalous measurement results at this highly trafficked kerbside site.
As mentioned above, the data available for Harwell are too limited and do not currently show any correlation between the two methods.

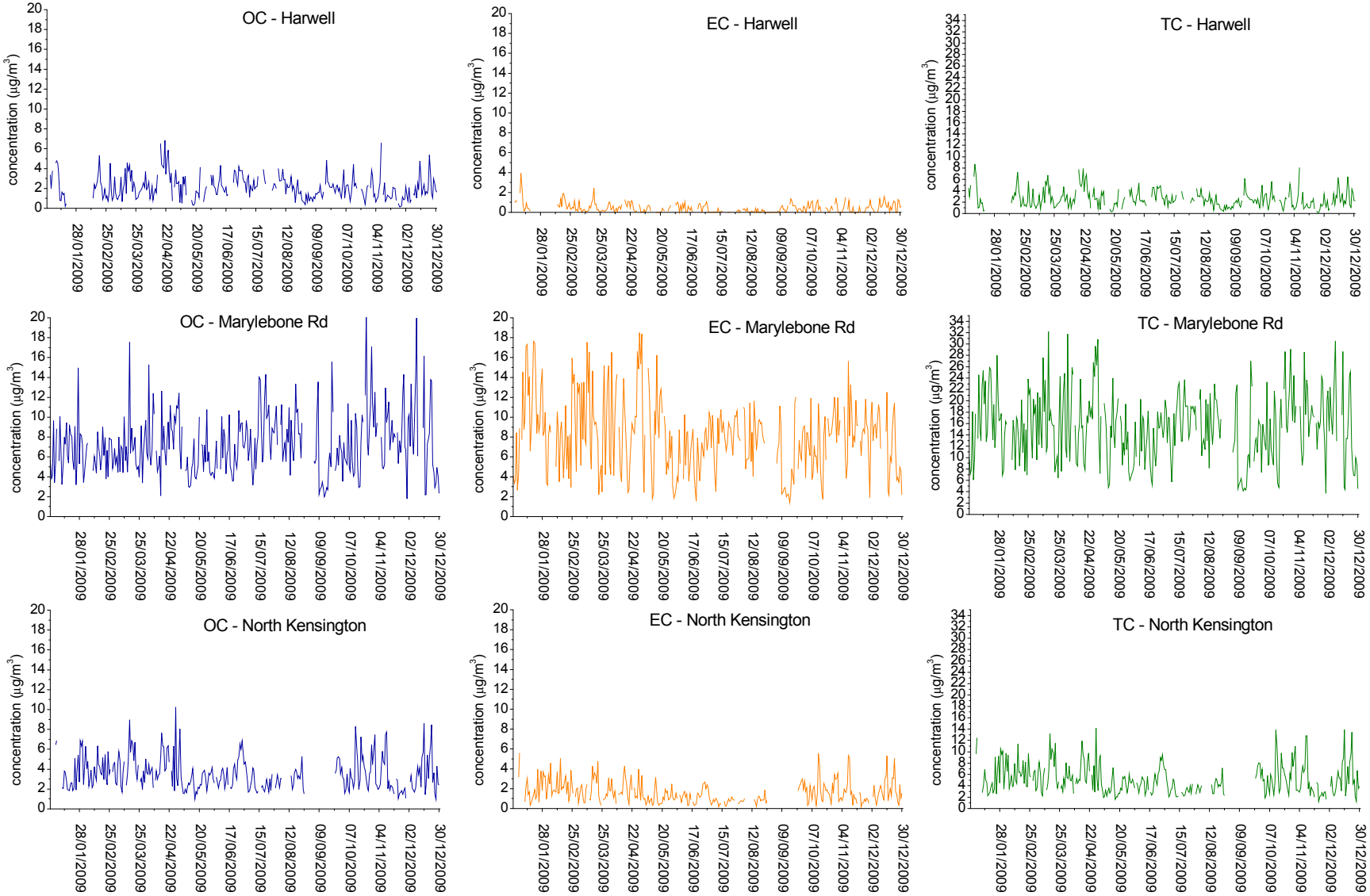


Figure 4-5: PM_{10} organic, elemental and total carbon trends at all sites in 2009

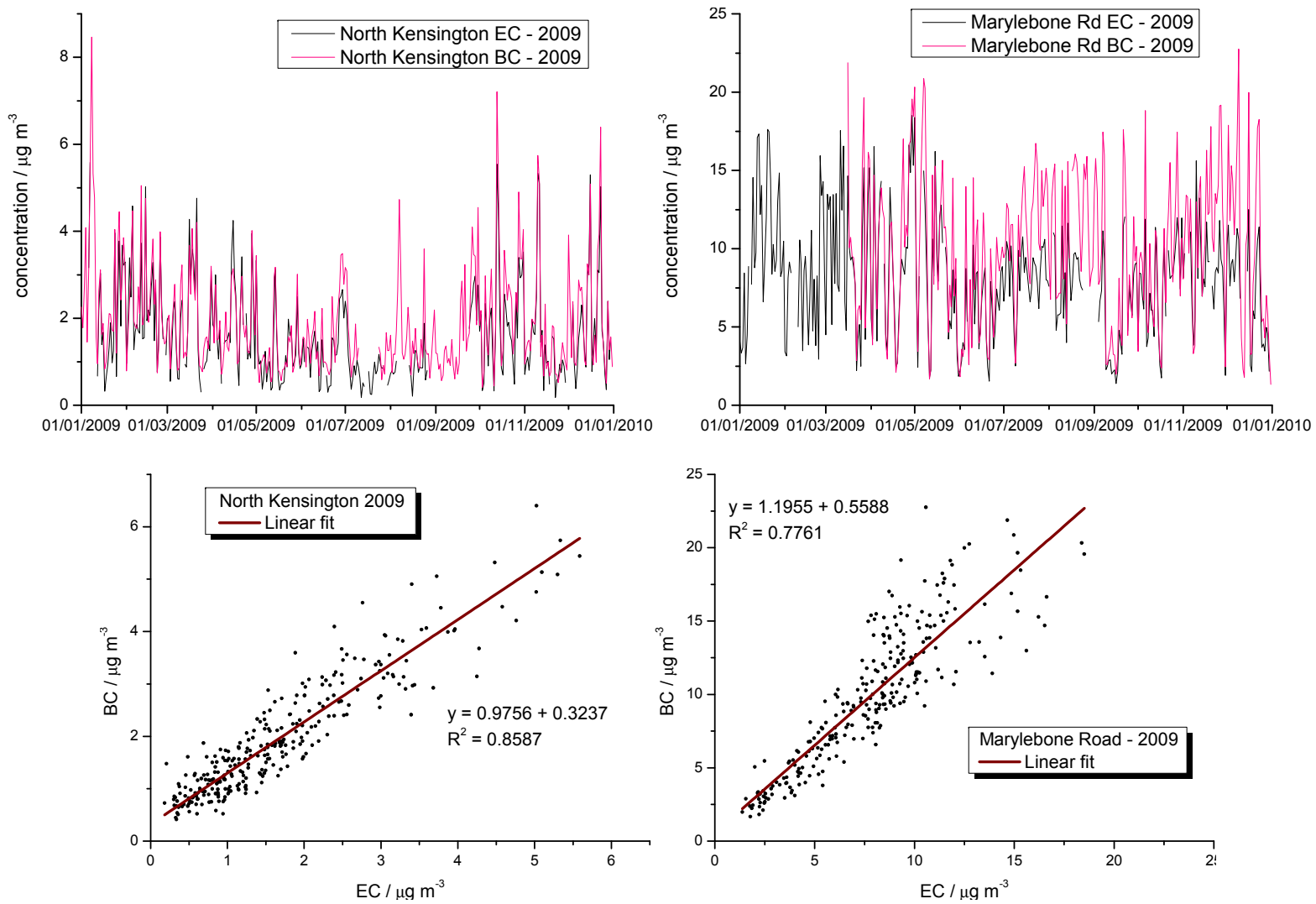


Figure 4-6: Comparison between BC ($PM_{2.5}$) and EC (PM_{10}) in 2009. [Left]: North Kensington site. [Right]: Marylebone Road site

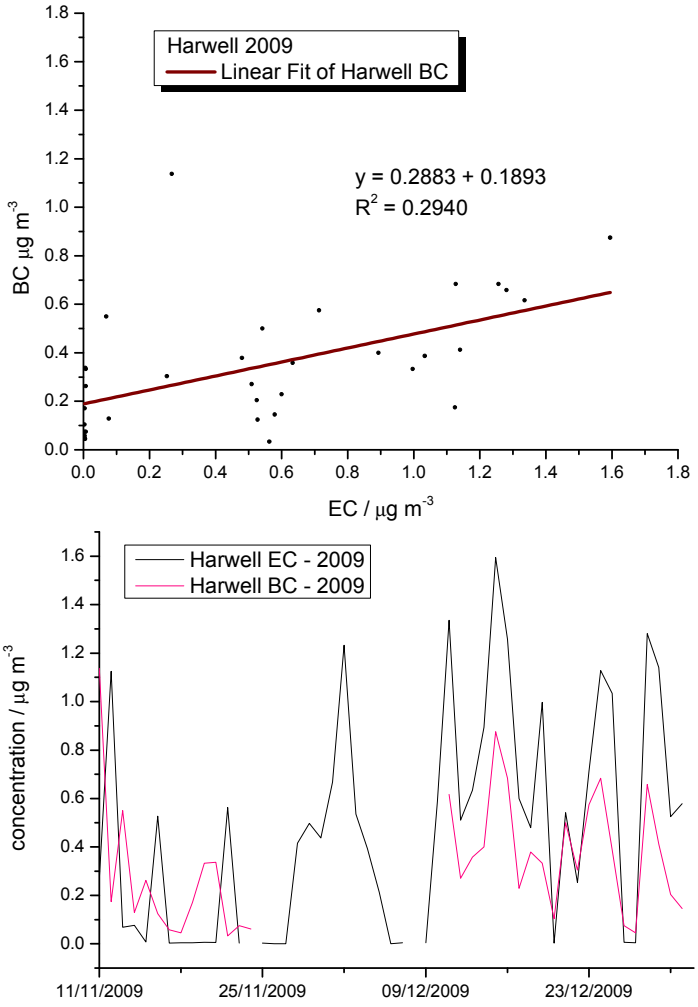


Figure 4-7: Comparison between BC ($PM_{2.5}$) and EC (PM_{10}) at Harwell in 2009

4.4 Particle Number and Size Distributions

4.4.1 Particle Number Concentrations (CPC)

Time series of hourly particle number concentrations (between about 7nm and several microns in diameter) measured at network sites during 2009 are shown in Figure 4-8 and 4-9.
The following should be noted:

- **Birmingham** –The instrument was removed from the Birmingham Centre site on 12/01/2009, as this site was closing. It was then sent to NPL for service and calibration, and installed at the Birmingham Tyburn Roadside site on 24/03/2009. On 5/11/2009 it was removed from site because the pump had failed, giving unstable flow. It was put back on site on 17/12/2009 after repair at TSI. It was moved to the Birmingham Tyburn (urban background) site in February 2010. Further details are given below.
- **Harwell –** The instrument was off site from the 2nd of March to the 13th of March for service and calibration at NPL. On the 9th of June it was removed from site and sent to Leipzig, Germany, for the EUSAAR 3rd NA3 S/DMPS Intercomparison Workshop together with the SMPS instrument. It was put back on site on the 7th of July.
- **North Kensington –** The instrument was off site from the 3rd of March to the 11th of April for service and calibration at NPL. Data from the 9th to the 18th was removed as the CPC had low flow.
- **Marylebone Road –** Data is missing up to the 5th of February as the instrument was at TSI for unstable flow, and from the 9th of April to the 12th of May for problems with the temperature condenser. On the 23rd of July it was removed from site and sent to TSI because the pump had failed. On the 13th of October the pump failed again and it was sent to TSI for repair. It was put back on site on 14th of November.

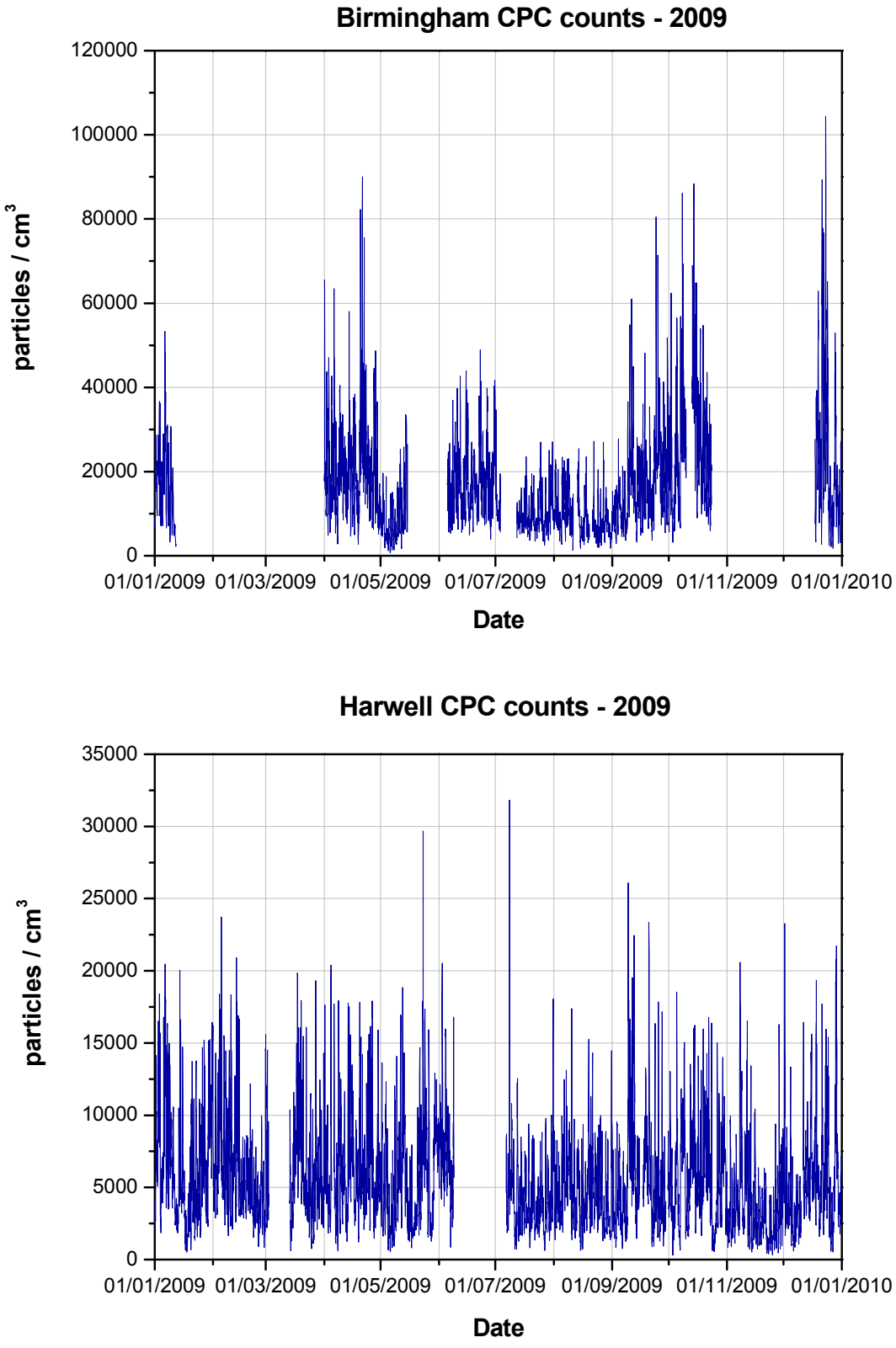
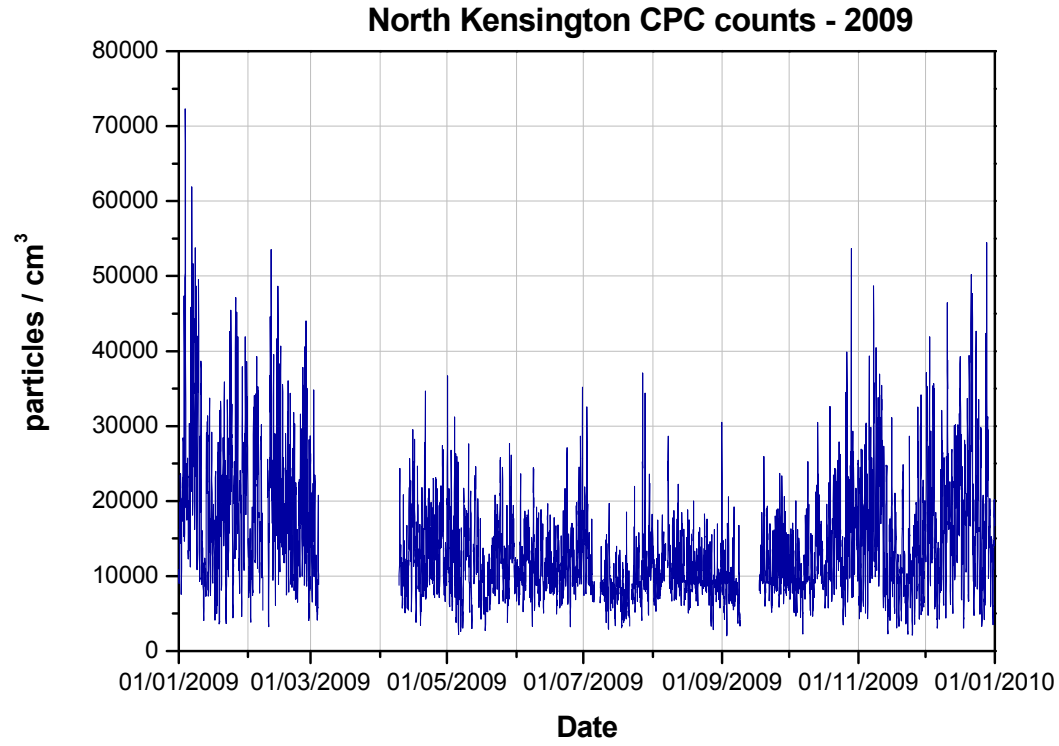


Figure 4-8: CPC Particles Counts at Harwell and Birmingham sites during 2009



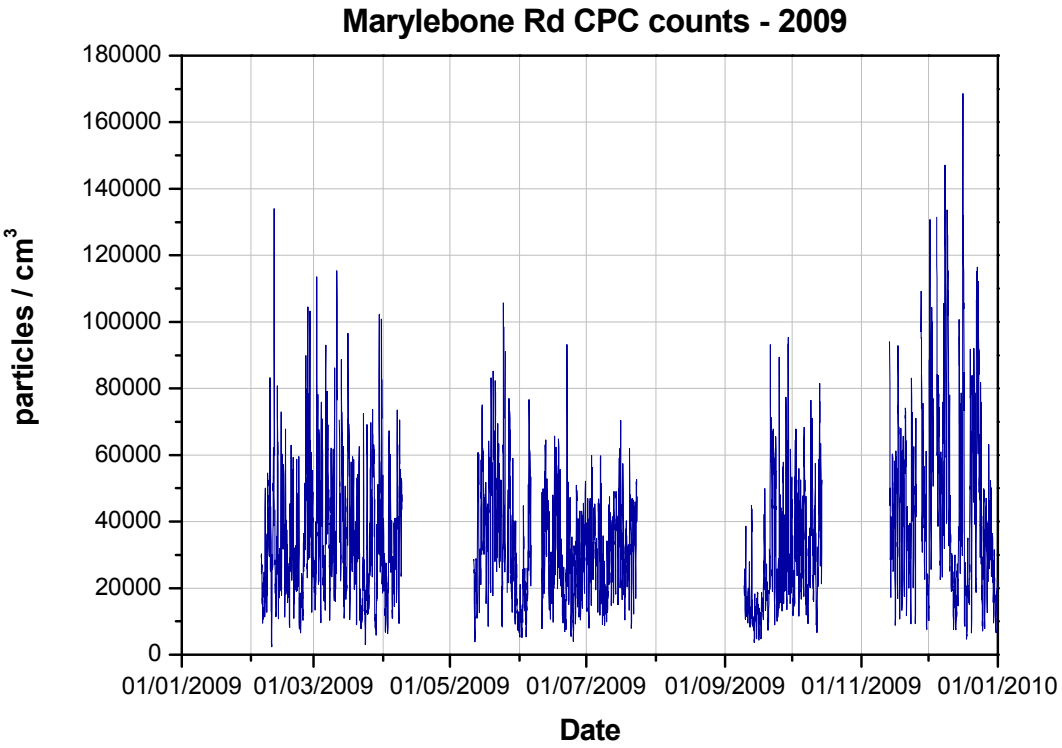


Figure 4-9: CPC Particles Counts at North Kensington and Marylebone Rd sites during 2009

Monthly data capture rates for the CPC instruments during 2009 are displayed in Table 4-3. Each instrument is removed from the site for a full service and calibration annually. This scheduled maintenance is expected to take three weeks, to include draining and drying, transit time, full service and re-installation. In the month(s) where the CPC was serviced, the data capture quoted in the table takes into account the scheduled downtime, and is denoted with an asterisk.

Two data capture sets are presented for the Birmingham CPC: the data capture in January is calculated according to the days the CPC was up and running on the old site in Birmingham centre before removal; the data capture for the rest of the year is for the new site in Birmingham Tyburn Roadsite. The average indicated in the table is for the new site. February was not taken into account, as neither site was operative.

The Birmingham centre site closure was an AURN decision and all the instruments present in that cabin, including the CPC, were relocated on the Birmingham Tyburn Roadside site. In February 2010 the CPC was moved to Birmingham Tyburn (urban background) for a better continuity with the previous site.

The CPC at Marylebone Rd seems to break down very frequently. The TSI 3022A CPC model, which is the model used for all the stand-alone CPCs in the Network, is an old model that has been discontinued by TSI. Some of the 3022A Network CPCs have been working since 2000 and need to be replaced by new ones.

The 3375 CPC model, which is the one used in the SMPS system, is a suitable replacement of the 3022A model. The 3775 CPC has a much improved flow control system using critical flow orifices. It is more reliable and laboratory measurements at NPL show that the flow is 10 times more stable than the old 3022A model.

Site	North Kensington	Birmingham	Marylebone Road	Harwell
January	100.0%	100%(**)	67.7%(*)	100.0%
February	83.6%	---	83.5%	90.3%
March	79.2%(*)	67.7%(*)	97.2%	100.0%(*)
April	69.4%	96.8%	28.3%	93.5%
May	98.4%	46.0%	63.2%	100.0%
June	96.8%	82.5%	81.5%	100%(**)
July	89.2%	72.2%	72.8%	100%(**)
August	97.2%	87.5%	0.0%	100.0%
September	68.3%	96.6%	69.2%	86.2%
October	99.5%	63.3%	43.1%	99.9%
November	96.8%	0.0%	49.6%	96.6%
December	100.0%	46.4%	95.0%	96.9%
Average	**89.9%**	**65.8%**	**62.5%**	**97.0%**

Notes: '*' denotes the month in which the instrument calibration and service was performed
Notes: '**' data capture calculated over the days the CPC was operative

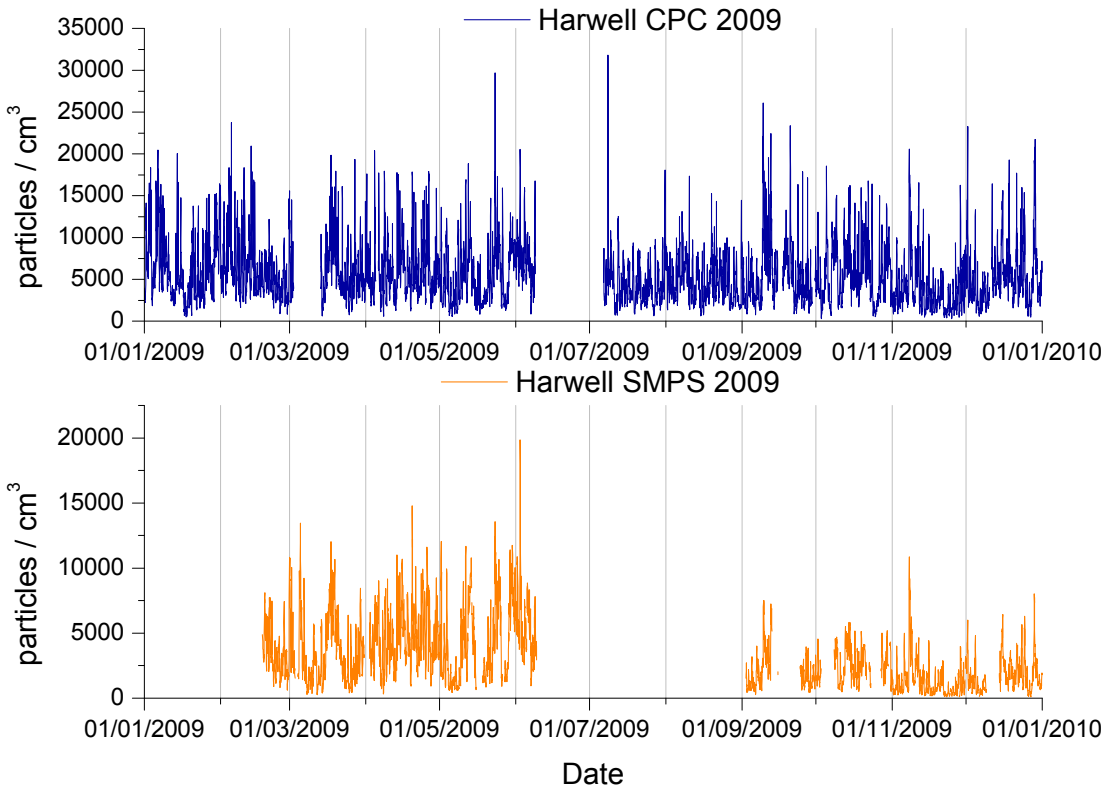
Table 4-3: Monthly Data Capture for CPC Instruments during 2009

4.4.2 Particles Number and Size Distributions (SMPS)

Total particulate counts (between about 16 nm and 605 nm aerodynamic diameter) have been plotted in Figure 4-10 and 4-11.

The following should be noticed:

- **North Kensington –** Instrument was at TSI in December 2008 and January 2009 for service and calibration. In February 2009 it was sent to TSI again for flow problems.
- **Harwell –** The instrument was at TSI until the 18th of February for service and calibration. On the 9th of June it was sent to Leipzig, Germany, for the 3rd EUSAAR NA3 S/DMPS Intercomparison Workshop. The CPC showed a fault and it was sent to TSI for repair. It was reinstalled on site on the 2nd of September after repair.
- **Marylebone Road –** From the 8th of April to the 12th of May, it was off site for flow problem with the CPC. On the 25th of October it was sent to TSI for a leak in the classifier. It was put back on site on the 15th of November.



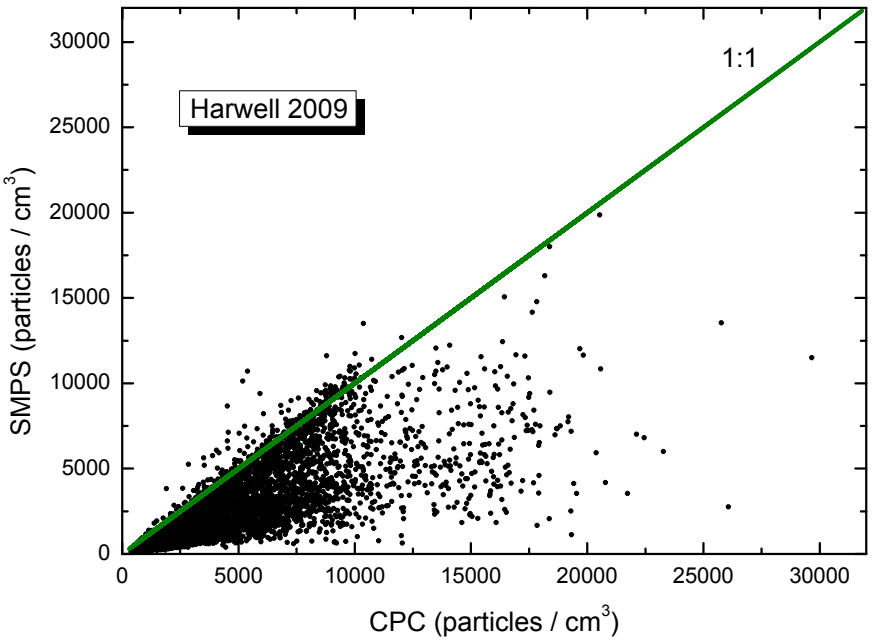
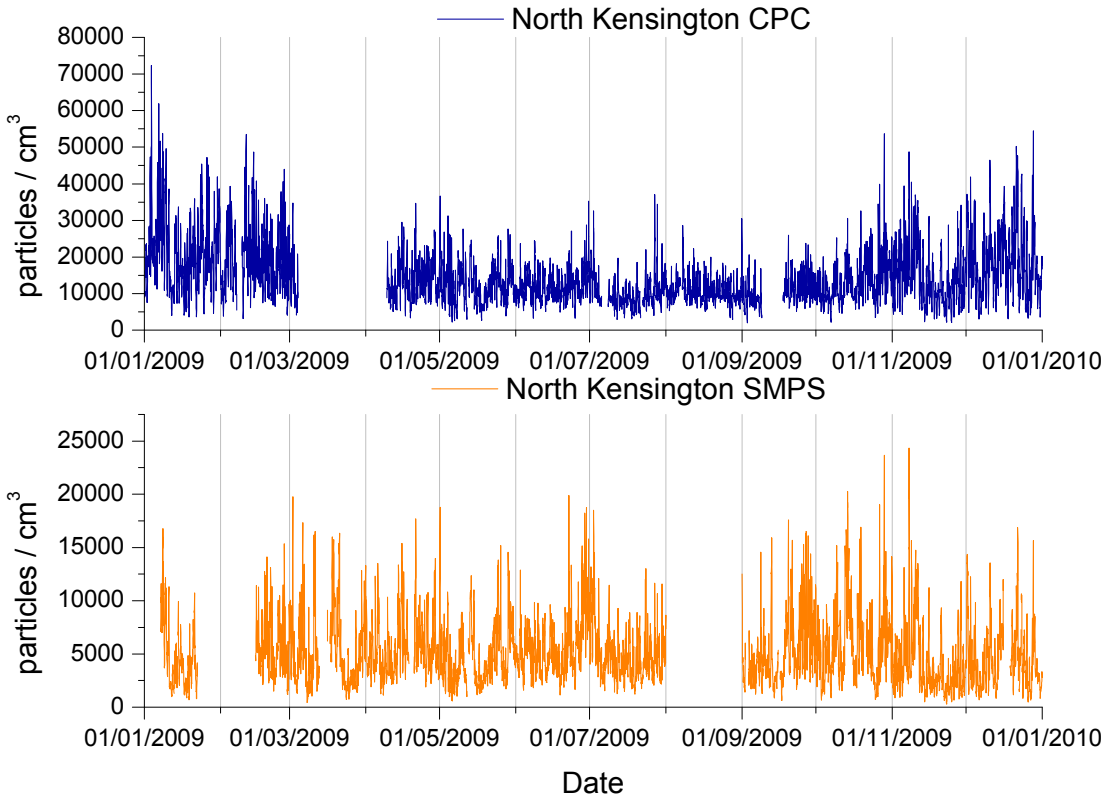


Figure 4-10: [Top] CPC and SMPS 2009 time series at Harwell. [Bottom] Harwell CPC and SMPS 2009 scatter plot



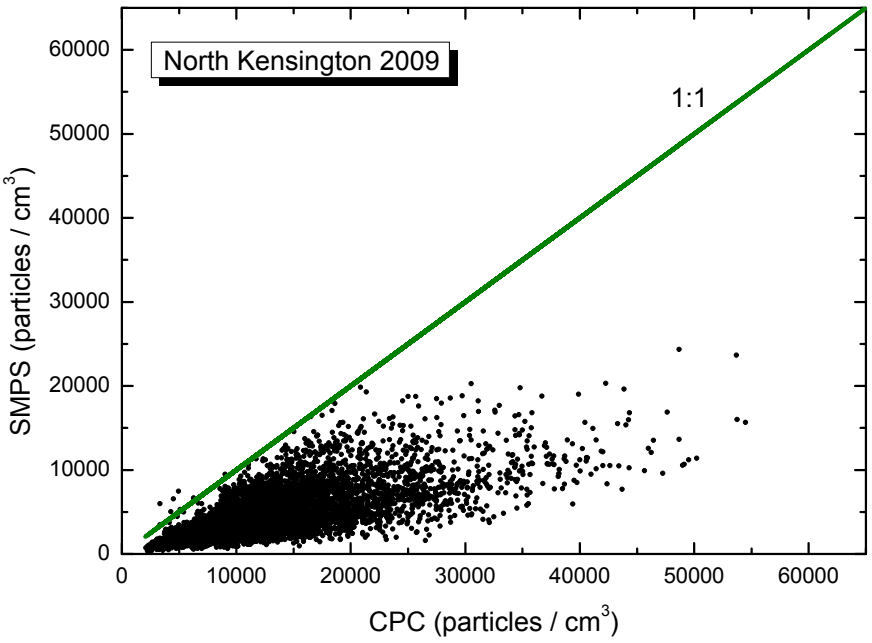


Figure 4-11: [Top] CPC and SMPS 2009 trends at North Kensington. [Bottom] North Kensington CPC and SMPS 2009 scatter plot

Monthly data capture rates for the SMPS instruments during 2009 are displayed in Table 4-4.

Month	Harwell	North Kensington	Marylebone Road
January	45.2%(*)	93.1%(*)	99.9%
February	64.3%(*)	49.3%	96.0%
March	93.8%	86.2%	100.0%
April	95.0%	92.9%	25.0%
May	88.0%	98.4%	58.2%
June	100%(**)	99.7%	86.8%
July	100%(**)	100.0%	92.7%
August	100%(**)	97.2%	61.7%
September	56.7%	93.3%	71.8%
October	63.6%	96.5%	71.1%
November	98.9%	100.0%	42.5%
December	82.4%	90.7%	91.1%
Average	**82.4%**	**91.7%**	**74.8%**

Notes: '*' denotes the month in which the instrument service was performed.
'**' Data capture calculated over days SMPS was on site

Table 4-4: Monthly Data Capture for SMPS Instruments during 2009

In June the Harwell SMPS was sent to Leipzig, Germany, for the EUSAAR 3rd NA3 S/DMPS Intercomparison Workshop, and when taken back to site the CPC was showing a flow fault that required a repair at TSI. It was not back on site before September. Therefore, the data capture for those 3 months indicates the time the instrument was on site and running.

The production of data from SMPS instruments is a complicated process, summarised schematically in Figure 4-12. Many stages of data processing are carried out by proprietary manufacturer's software to convert the raw data (number count versus Differential Mobility Analyser voltage) into the final data (number concentration versus particle size). While the size axis can be reliably calibrated using certified spheres, the number concentration axis, and hence both the scale and shape of the size distribution, is much less amenable to direct evaluation.

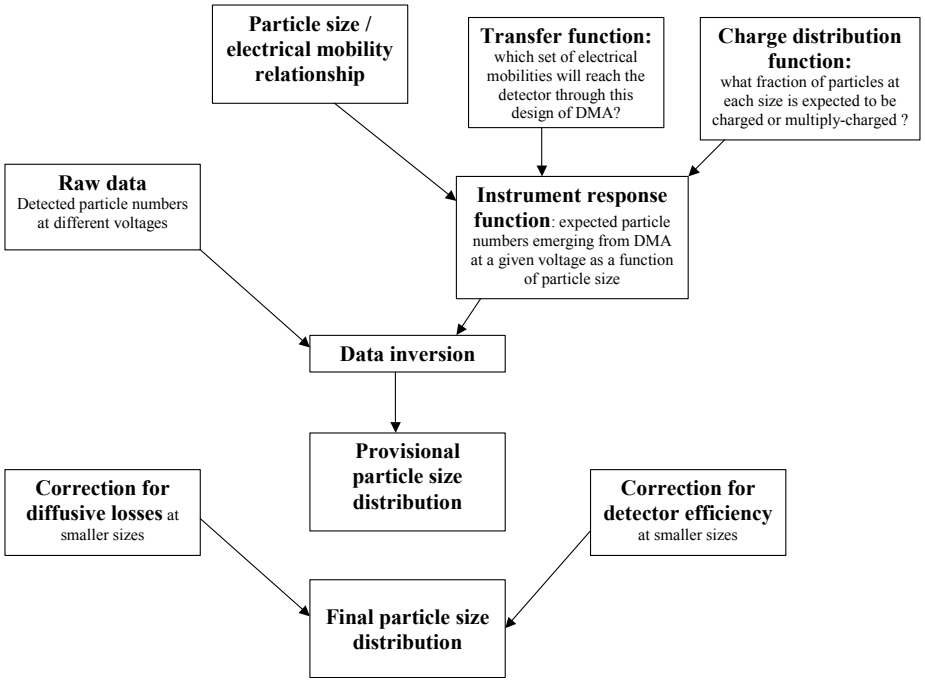


Figure 4-12: Schematic of the Internal Data Processing of the Current SMPS Instrument in the Network.

Some elements of the software in the current TSI instruments (Model 3936L75) are more transparent than for the previous TSI 3071 model used in the Network (in 2005). The multiple charge correction and diffusion loss correction software can be switched on and off by the user. Both of these corrections are used in the data reported here.

The effect of the diffusion loss and multiple charge corrections can be seen in Figure 4-13. The uncorrected spectrum is shown in black. The effects of the multiple charge correction and diffusion loss corrections are shown in green and red, respectively. The blue curve is the combined effect of the two corrections. The overall effect of the two corrections is to increase the particle number counts at smaller sizes and to increase the total particle count [EURAMET Project 1027, Comparison of nanoparticle number concentration and size distribution].

It is clear that great care needs to be applied when comparing SMPS data from similar instruments on different settings, and even more when comparing SMPS data from different instruments.

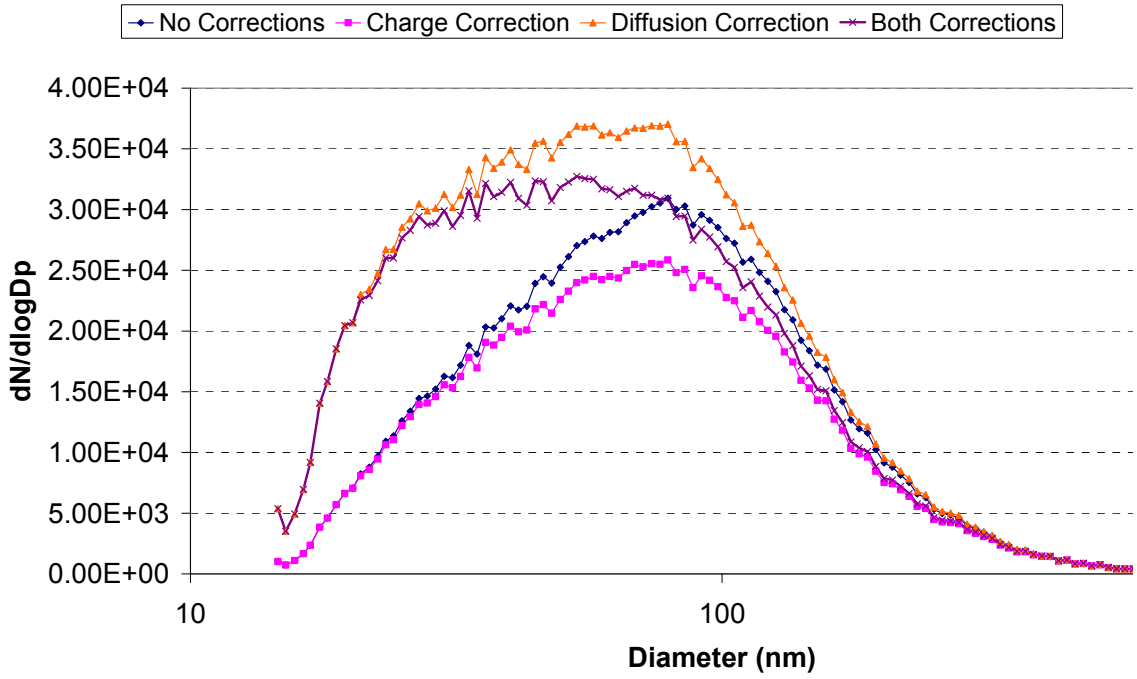


Figure 4-13: Effect of (i) the multiple charge, (ii) the diffusion loss and (iii) the combined correction on the SMPS size spectra.

4.4.3 SMPS versus CPC data

A scatter plot between the CPC particles counts and total SMPS particle counts (all size fractions) at all sites in 2008 is presented in Figure 4-17.

An example of the historical relationship of the ratios of the CPC to SMPS total counts is shown in Table 4-5.

July 2005	2.7
August 2005	2.4
September 2005	2.3
October 2005	2.5
November 2005	2.6
December 2005	2.6
January 2006	3.0

Table 4-5: Monthly Ratio of CPC to SMPS Particle Counts in 2005 and 2006 at Marylebone Road Site.

CPC to SMPS particle count ratios for the three sites with co-located instruments in 2007, 2008 and 2009 are shown in Table 4-6.

The complicated data processing within SMPS instruments, described in Section 4.4.2 above, means that the total particle number concentration obtained by integrating the SMPS size distribution is subject to much greater uncertainties than CPC instruments,

which measure number concentrations much more directly. In addition, the results cannot be compared directly because the instruments measure particles over different size ranges – the SMPS covering approximately 16-600 nm aerodynamic diameter, and the CPC covering from around 7 nm to several microns. Clearly this means that the CPC should inherently record higher concentrations than the SMPS.

	2007			2008			2009		
	Marylebone Road	Harwell	North Kensington	Marylebone Road	Harwell	North Kensington	Marylebone Road	Harwell	North Kensington
January				2.4	5.1	2.6			4.6
February				2.0	1.0	2.2	2.1	1.7	3.3
March	2.9			-	1.8	2.6	2.1	1.7	2.6
April				-	2.0	2.5	2.1	1.3	2.5
May	4.1			-	1.4	1.9	2.1	1.3	2.5
June	2.6			1.8	1.4	1.6	1.6	1.3	2.1
July	2.6			1.1	1.3	1.1	1.3		2.0
August	2.4			1.1	1.2	1.2			
September	2.2			1.0	1.1	1.1	1.4	2.7	2.0
October	1.9	1.8		1.1	1.7	1.3	1.6	2.3	2.2
November	3.6		8.8	1.3	1.6	-	2.0	2.8	3.6
December	3.4		3.5	1.4	1.4	-	2.7	2.9	4.1

Table 4-6: Monthly Ratio of CPC to SMPS Particle Counts in 2007, 2008 and 2009

4.4.4 EUSAAR upgrade inlet systems

The EUSAAR project (European Super-sites for Atmospheric Aerosol Research) includes 20 sites across Europe including Harwell. It has recommended that SMPS instruments should standardise the aerosol size distribution by measuring under dry conditions (<40% RH). The SMPS and CPC instruments at Harwell underwent an audit by the EUSAAR project team during November 2007. Several recommendations were made:

- Replace inherited copper pipe work and 'funnel' sampling inlet with a PM_1 sampling head and stainless steel tubing.
- Provide a calibrated flow meter (for example, a BIOS dry-cal meter or Gilian Gilibrator bubble meter) for the LSO to measure the flow rate of the CPC and SMPS fortnightly.
- Provide an ultrasonic bath for the cleaning of the SMPS impactor.
- Install humidity control and monitoring on the sample line, in the form of a drier and humidity sensor.
- Ensure that the Harwell site operator has access to documentation on instrument performance and measurement quality, either in written form at the site or as a detailed online log accessible form the site at any time.

To pursue these recommendations, NPL has provided the following:

- The copper pipe work and 'funnel' sampling inlet has been replaced with a PM_1 sampling head and stainless steel tubing, as part of a contract to supply improved inlet systems for the CPC and SMPS instruments.
- A calibrated flow meter has been provided to the local site operators to measure the flow rate of the CPC and SMPS on a fortnightly basis.
- Ultrasonic baths have been provided to each site.
- The humidity of the sample air is controlled and monitored through the use of Nafion driers and installation of humidity sensors.
- The site operators are provided with copies of the documentation on instrument performance and measurement quality. A web portal, operated by the network management team at King's College London, provides information on site visits, instruments' performance and calibration and call-outs to the instruments' manufacturers.

The upgrade systems were installed on September 2nd at Harwell, on October 19th at North Kensington, on December 2nd at Marylebone Rd and on February 17th 2010 at Birmigham for the stand-alone CPC. On this occasion the instrument was moved to Birmingham Tyburn site (urban background) for continuity with the previous site in Birmingham centre.

4.4.5 SMPS size distributions

The counts in each particle size bin measured during 2009 are presented as monthly averages in the left-hand panels of Figure 4- and as an annual average in the right-panel. It has not been possible to ratify discrete 15 minute averaged data for the Marylebone Road SMPS due to instrument performance problems, which led to unacceptable levels of uncertainty in the data set and to a longer averaging period being used. Therefore, data are presented as monthly averages to give an indication of concentrations and size distributions.

In some of the size distributions the lowest size bins have been excluded from these data due to an artifact associated with the operation of this SMPS model under network conditions. Each 15 minute average consists of several size scans which are done without a pause between them. Any residual particles that have not cleared the CPC before the next scan begins are erroneously counted in these smallest size bins. A strategy to remove this has been identified and implementation is under discussion.

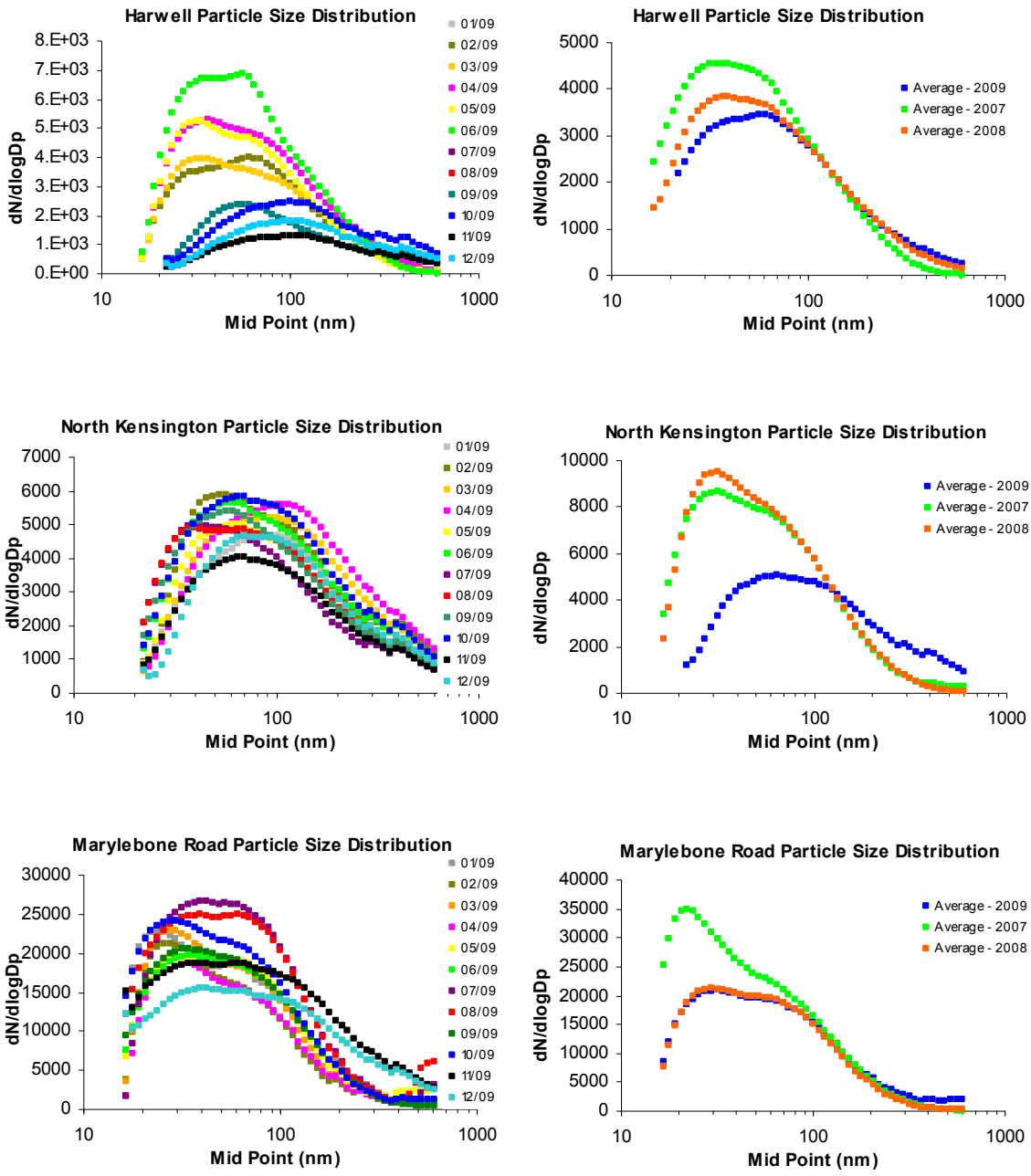


Figure 4-14: Monthly-averaged Particle Size Distributions at Network Sites during 2009 [Left-hand Panels] and Comparison of the 2007, 2008 and 2009 Annual-averaged Size Distributions [Right-hand Panels].

4.5 Meteorological Data

Although not a formal part of this measurement programme, meteorological data have been collated from the measurements made at Harwell and Rochester in other Defra monitoring networks. Monthly data capture rates for the meteorological masts during 2009 are displayed in Table 4-7.

Site	Harwell	Rochester
January	100%	100%
February	100%	100%
March	100%	100%
April	100%	100%
May	100%	100%
June	99%	100%
July	100%	100%
August	100%(*)	100%
September	---	100%
October	100%(*)	80%
November	98%	100%(*)
December	98%	---
Average	**99%**	**98%**

(*) Data capture calculated over the days the site was operational

Table 4-7: Monthly Data Capture for Meteorological Instruments during 2009

The Harwell meteorological station has been upgraded as part of the EMEP network at the end of August. The collection of the data was passed over to AEA at the beginning of October. No data is available for September.
The site at Rochester was switched off at the beginning of November due to a leak in the roof. The site is still not operational.
Wind roses have been derived for Harwell and Rochester for each month in 2009. These can be found in Annex 1. As the Rochester site was operational for only 3 days in November, the wind rose for this month is not shown.

4.6 Trends and Profiles

4.6.1 Annual Mean Concentrations

Annual mean concentrations have been derived for the inorganic components, as shown in Table 4-8. The table also includes the annual mean concentrations for the years 2003 to 2008 for comparison.

All anions concentrations are lower than previous years. Figure 4-17 shows a downward trend of sulphate concentration at all sites. The reason for this drop in concentration will be investigated further.

Component	2003 Mean	2004 Mean	2005 Mean	2006 Mean	2007 Mean	2008 Mean	2009 Mean	Units
Harwell Nitrate ($PM_{2.5}$)	2.66	4.27	1.94	1.61	2.26	3.03	2.5	$\mu g.m^{-3}$
Marylebone Road Nitrate ($PM_{2.5}$)	-	-	3.33	3.18	3.25	2.33	3.4	$\mu g.m^{-3}$
North Kensington Nitrate ($PM_{2.5}$)	-	-	-	-	-	2.84	2.3	$\mu g.m^{-3}$
Harwell Daily Nitrate (PM_{10})	3.90	2.76	3.17	3.21	3.06	2.96	1.67	$\mu g.m^{-3}$
Marylebone Road Daily Nitrate	5.06	3.93	3.97	4.39	4.05	3.86	3.06	$\mu g.m^{-3}$
North Kensington Daily Nitrate	4.32	3.38	3.91	3.74	3.76	3.24	2.63	$\mu g.m^{-3}$
Harwell Daily Sulphate (PM_{10})	2.44	2.29	2.40	3.01	2.40	2.14	1.30	$\mu g\ m^{-3}$
Marylebone Road Daily Sulphate	3.18	3.16	3.21	4.00	3.15	2.74	1.79	$\mu g\ m^{-3}$
North Kensington Daily Sulphate	2.61	2.95	3.02	3.51	2.79	2.57	1.72	$\mu g\ m^{-3}$
Harwell Daily Chloride (PM_{10})	0.91	1.04	1.19	1.32	1.38	1.52	1.11	$\mu g\ m^{-3}$
Marylebone Road Daily Chloride	1.45	1.54	1.64	2.01	1.91	2.16	1.47	$\mu g\ m^{-3}$
North Kensington Daily Chloride	1.11	1.24	1.33	1.57	1.66	1.91	1.36	$\mu g\ m^{-3}$

Table 4-8: Annual Mean Concentrations of Inorganic Anions, 2003–2009

4.6.2 Monthly Trends

Using data from this contract, this section seeks to show the trends in the average monthly values for all species at all sites during the past several years (see Figure 4-15 to 4-20). All available data has been used, regardless of the data capture in any particular month.

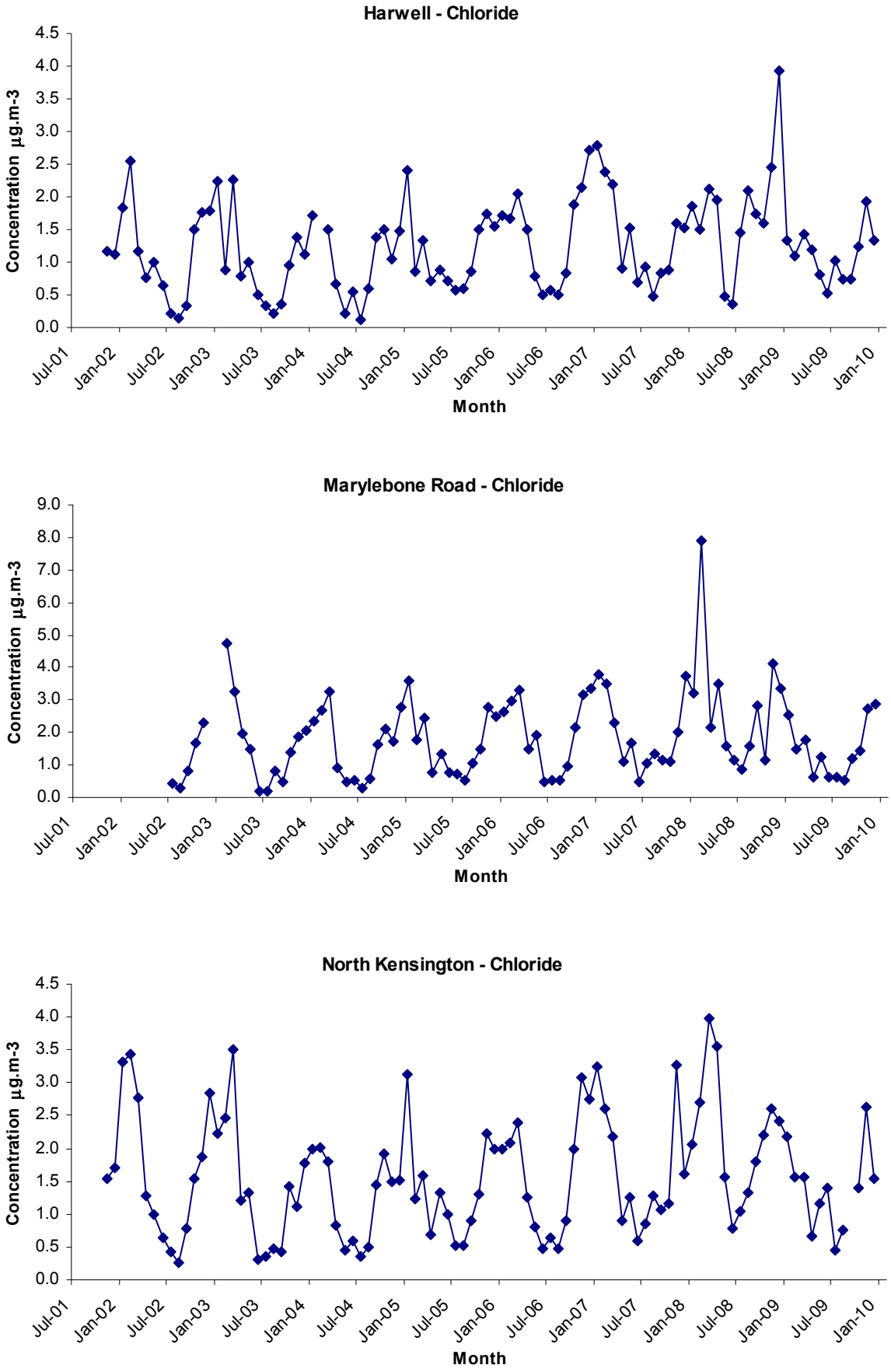


Figure 4-15: Monthly trends for PM_{10} chloride concentrations at Network sites

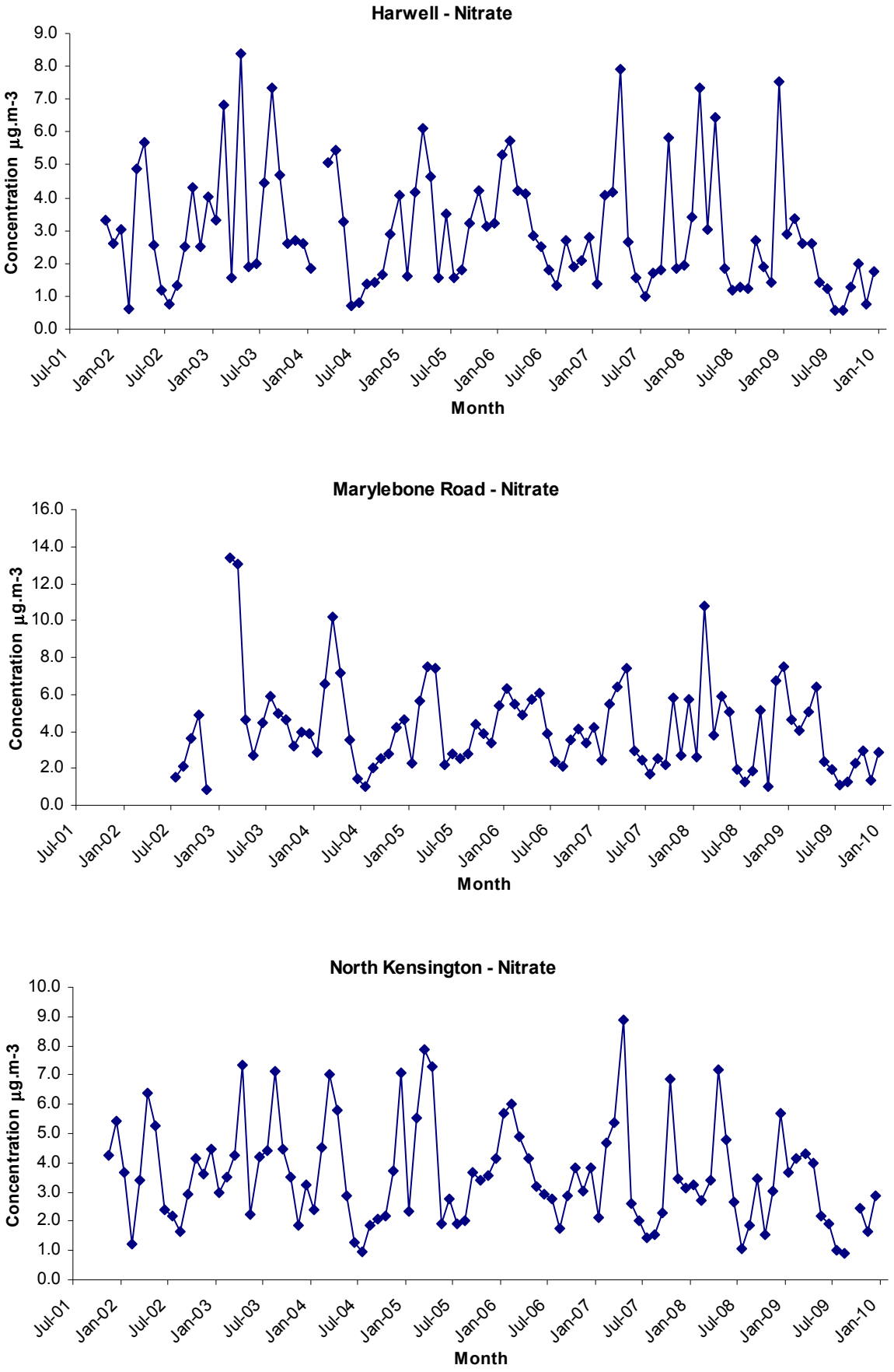
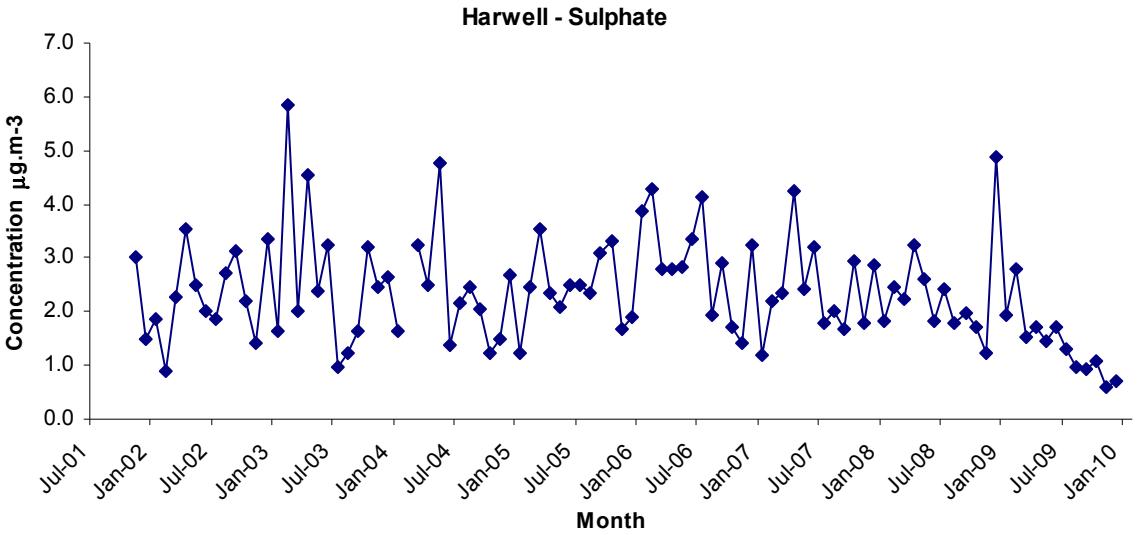
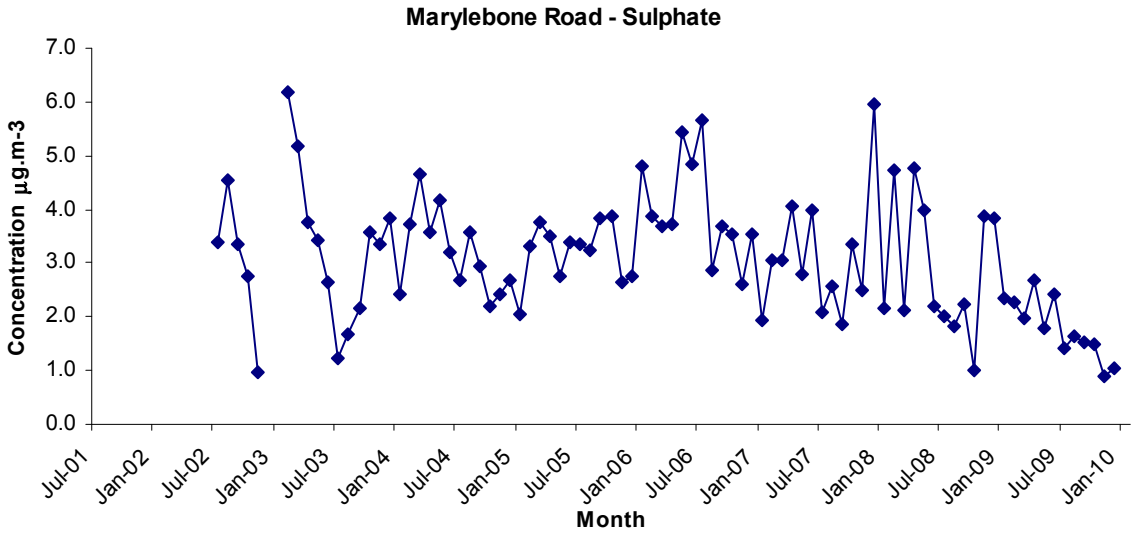


Figure 4-16: Monthly trends for PM_{10} nitrate concentrations at Network sites





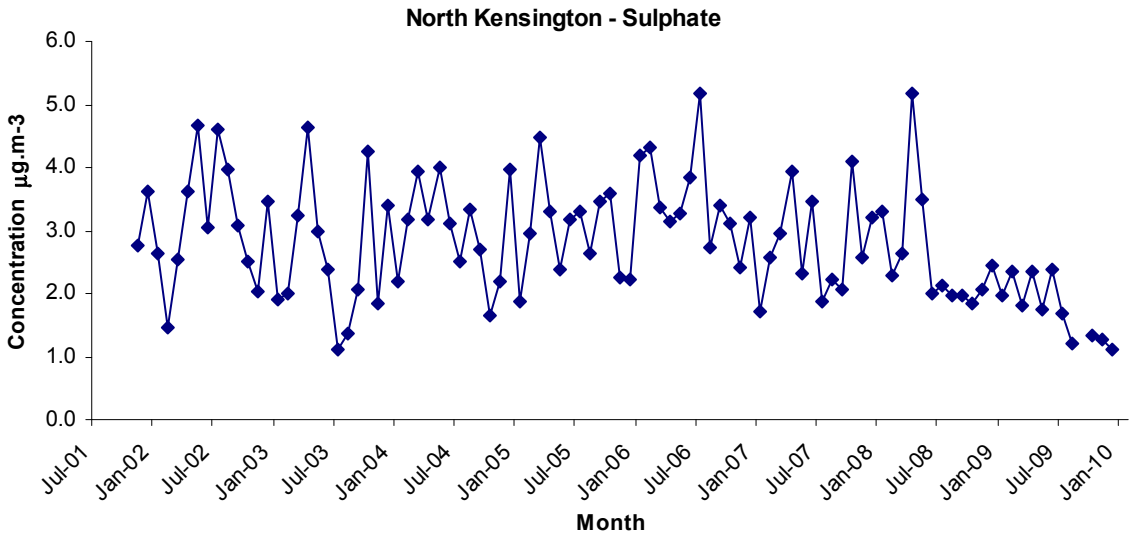


Figure 4-17: Monthly trends for PM_{10} sulphate concentrations at Network sites

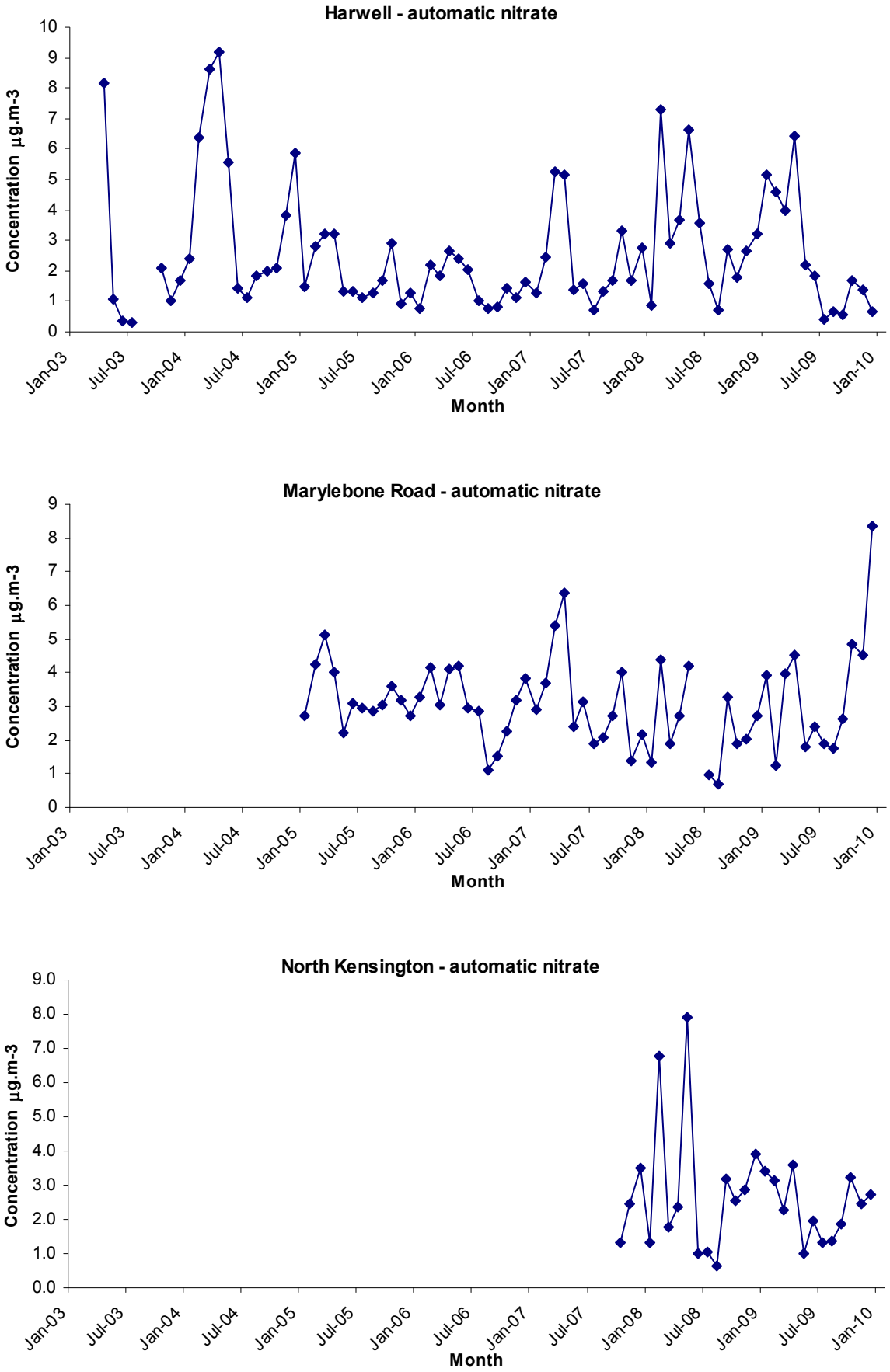


Figure 4-18: Monthly trends for $PM_{2.5}$ automatic nitrate concentrations at Network sites

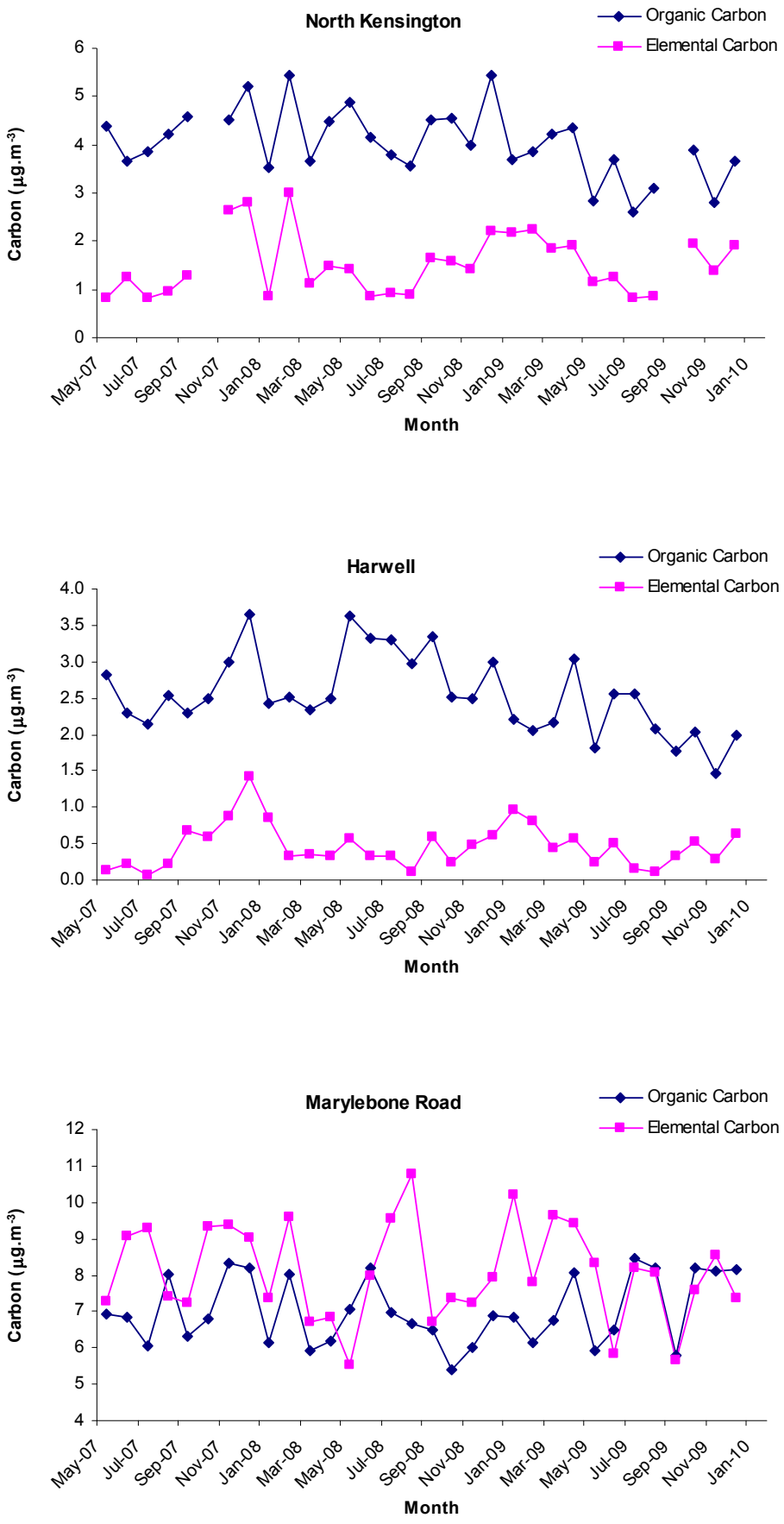


Figure 4-19: Monthly trends for PM_{10} OC/EC concentrations at Network sites

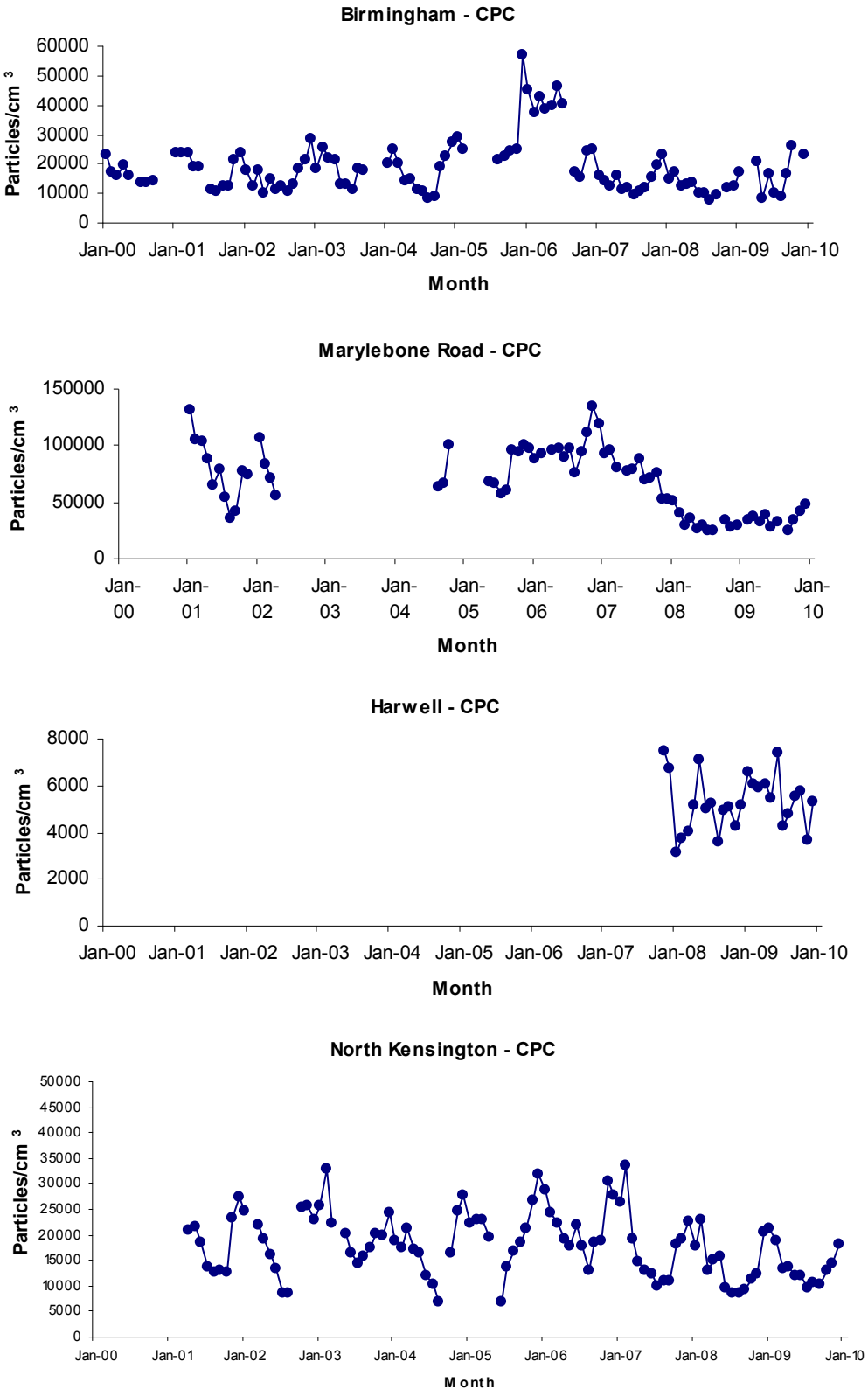


Figure 4-20: Monthly trends CPC counts at Network sites

4.6.3 Diurnal Profiles

Diurnal profiles have been derived for the automatic 'continuous' instruments. The profiles by quarter (January-March, April-June, July-September and October-December) are shown in Figure 4-21 and 4-22 for the CPC and in Figure 4-23 and 4-24 for the automatic nitrate measurements.

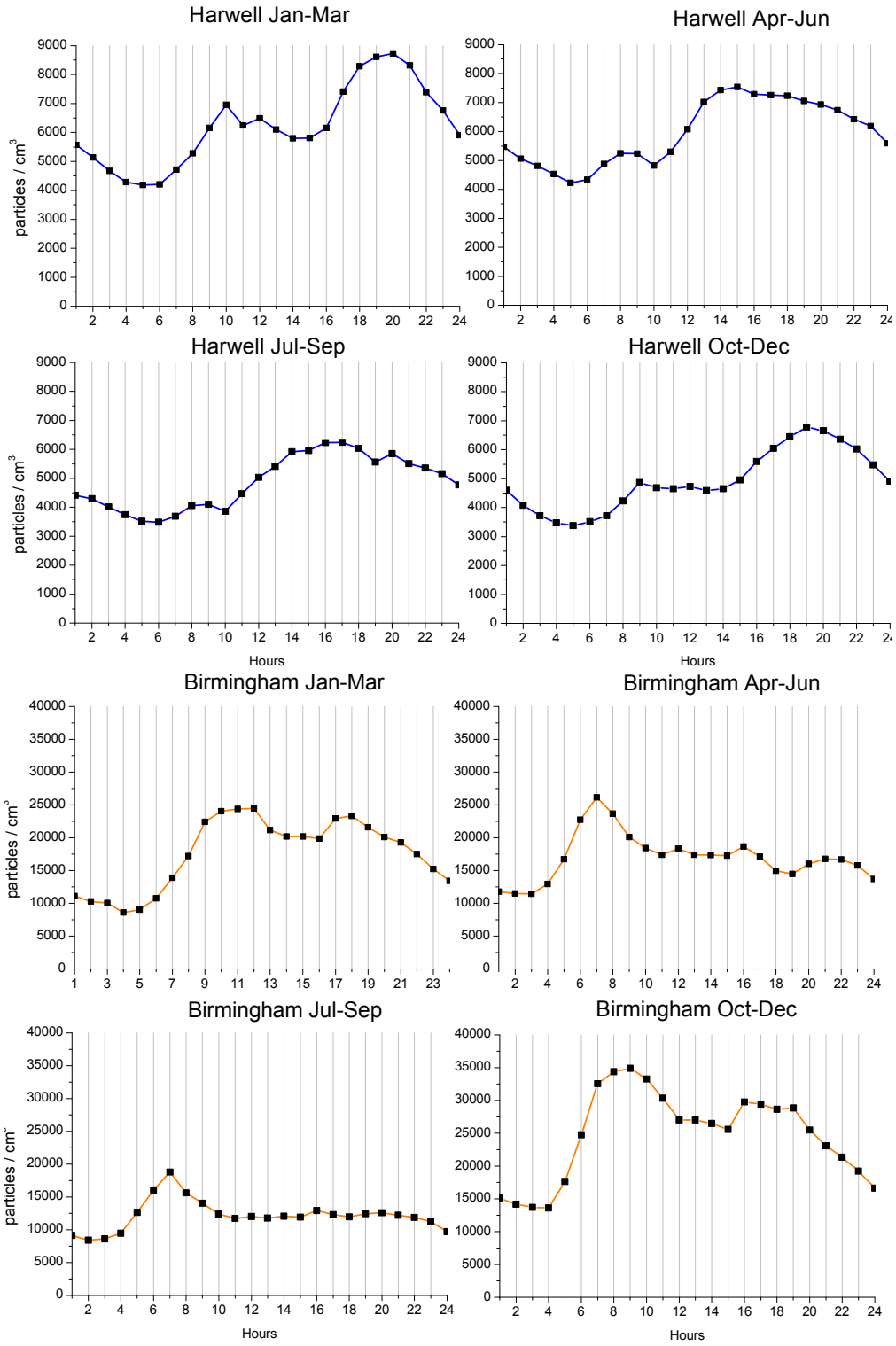


Figure 4-21: Diurnal trend for CPC counts in 2009 for Harwell and Birmigham sites

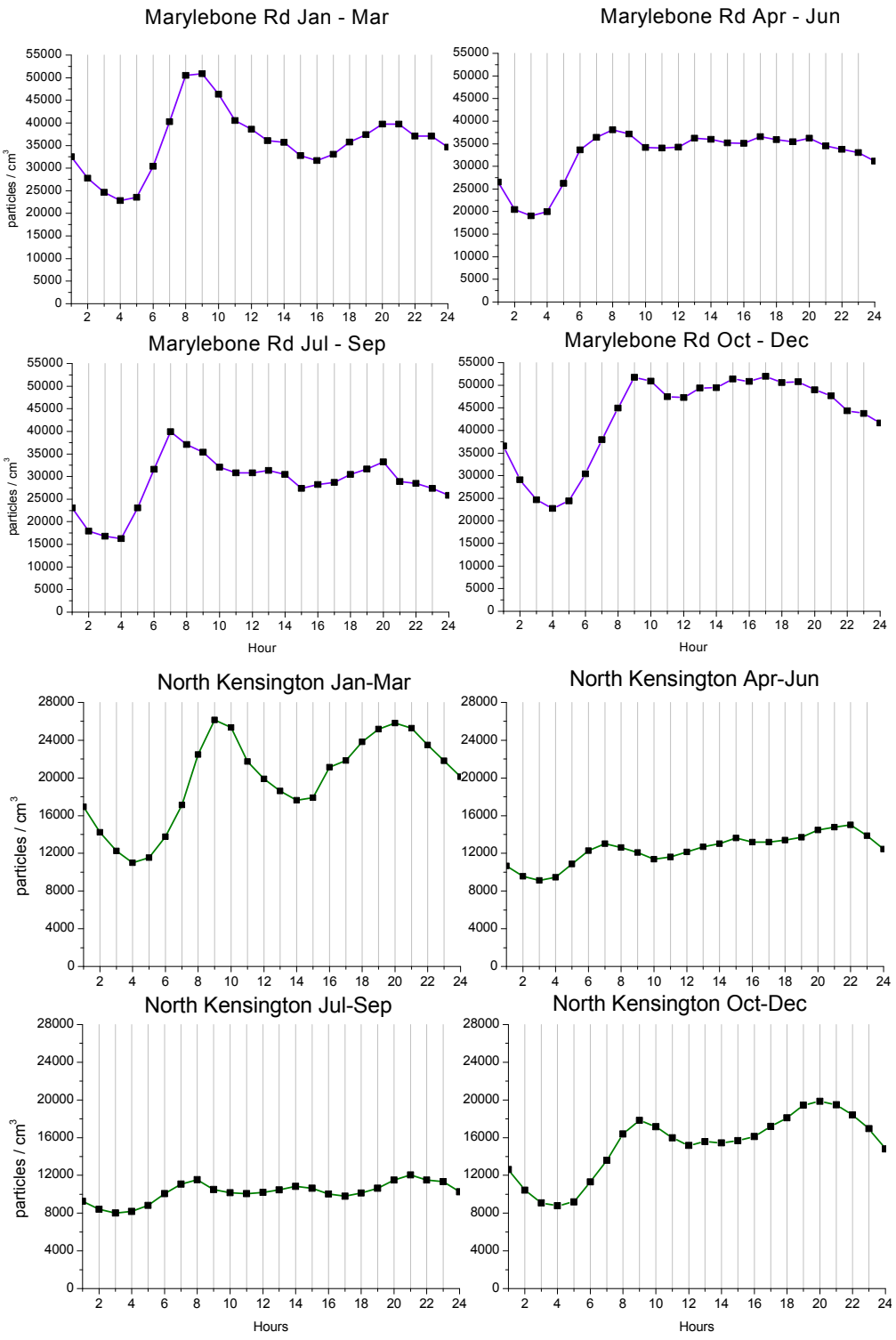


Figure 4-22: Diurnal trend for CPC counts in 2009 for Marylebone Rd and North Kensington sites

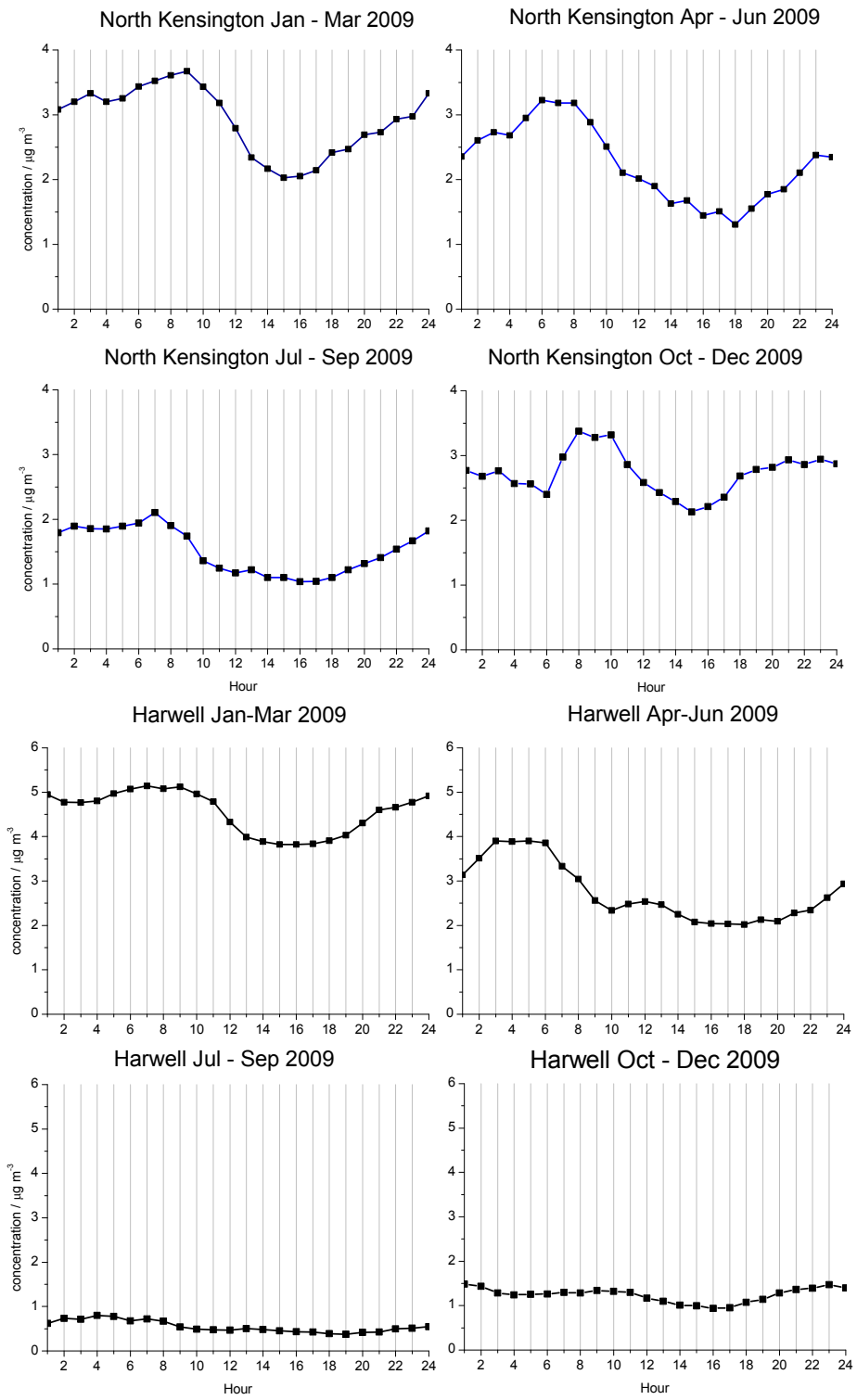


Figure 4-23: Diurnal trend for automatic nitrate concentrations in 2009 for North Kensington and Harwell sites

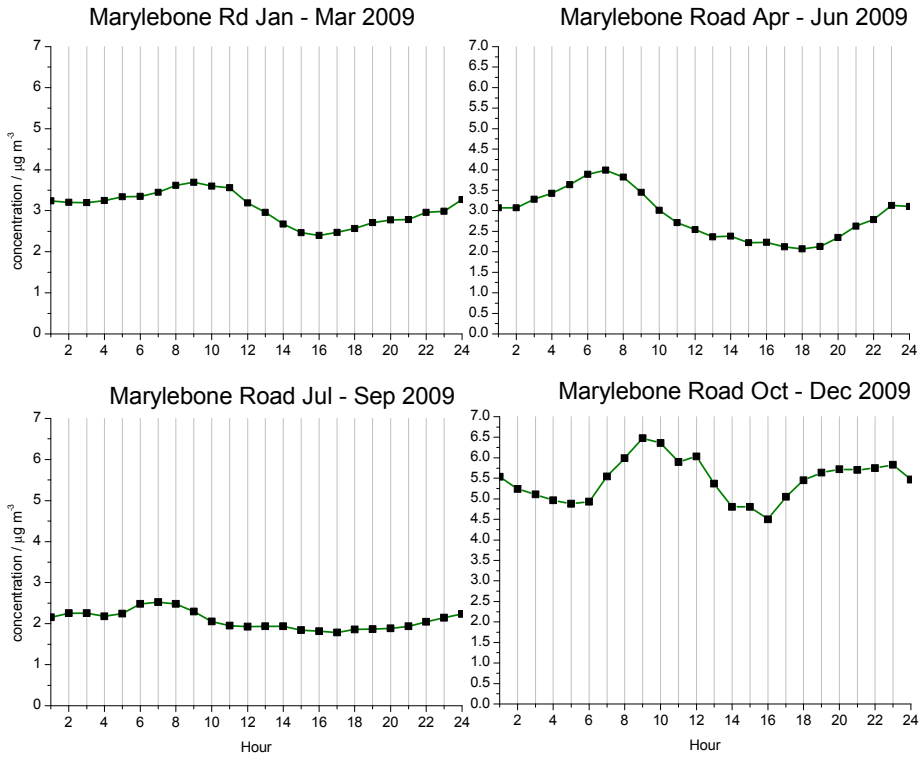


Figure 4-24: Diurnal trend for automatic nitrate concentrations in 2009 for Marylebone Road site

5 Update on the Wider Policy and Research Context

The measurements made within this Network are one research resource in the area of particulate matter. Other sources of data should be borne in mind. In this Section, we identify complementary measurement activities, which will provide additional data (a) to confirm the measurements made in this network or (b) to assist the interpretation of the measurements.

5.1 Update on Related UK Activities

5.1.1 London Specific Measures

The characterisation of the chemical composition of PM is of particular importance in London. Roadside locations in London were the only areas identified as likely to exceed the PM_{10} objective in the recent UK application to the European Commission for an extension to meet PM_{10} air quality limits. An understanding of the chemical composition is vital to understand the impact of local, regional, national and international emissions abatement.

London is also the subject of the largest Low Emission Zone in the UK, which began in February 2008 with emissions restrictions for heavy lorries. Further emissions restrictions for lighter lorries were bought in during July 2008; the emissions standards for both these vehicle classes will be further tightened in January 2012. The extension of the LEZ to large vans and minibuses was planned for 2010 but was postponed in the Mayor's Draft Air Quality Strategy.
The draft strategy focuses effort on three key corridors into London, which are the locations of most of the pollution hot spots, one of these corridors includes the Marylebone Road. Many of the novel approaches to PM abatement, such as power washing roads and the application of dust suppressants, will require detailed validation using the chemical composition measurements made using this network at this site. The promotion of cleaner vehicles on these routes and across London will also require assessment using particle number concentrations.

5.1.2 Defra and Other National Monitoring Activities

AURN measurements of PM_{10} and $PM_{2.5}$

Any investigation of PM must be linked to the officially reported PM_{10} and $PM_{2.5}$ measurements. The discrepancies between the reference gravimetric method and the standard TEOM instrument are well known, and it has also become apparent that the reference method needs to be very carefully implemented to produce reliable results. The effects of humidity on gravimetric PM measurements made on different filter types have been clarified by a study co-funded by the EU. [*A study on the effects of humidity on the mass of UK PM samples*, P G Quincey and D M Butterfield, NPL Report AS 40, 2009.]

Black Smoke / Black Carbon Measurements

There are currently 21 sites in the Black Smoke Network that measure Black Carbon optically using aethalometers, a metric designed to approximate to elemental carbon. Although the conventional Black Smoke index (given in units of μg m^{-3}) is quite different from a measure of elemental carbon, NPL has provided evidence that the black smoke index and the aethalometer measure of black carbon are closely related [Quincey, 2007], allowing better interpretation of historical data. Three aethalometers are located at sites in this network (Harwell, North Kensington and Marylebone Road).

Rural Monitoring

Daily measurements of sulphate, and monthly measurements of nitrate, chloride and ammonium are made at a number of rural sites through the Ammonia and Acid Deposition Monitoring Networks.

As part of the UK implementation of the EMEP monitoring strategy, two sites, Auchencorth Moss and Harwell, have been established to monitor, *inter alia*, particulate matter. The measurements of relevance to this network are those of:

- Sulphate, nitrate, ammonium, sodium, potassium, calcium and magnesium ions in both the PM_{10} and $PM_{2.5}$ size fractions, on an hourly basis, using a steam-jet aerosol collector.

- EC by aethalometry with supplementary analysis of filter samples for EC and OC.

5.2 Update on European Activities

5.2.1 New EU Air Quality Directive 2008/50/EC

The new Directive on Ambient Air Quality and Cleaner Air for Europe came into force in 2008, and is transposed into UK legislation by a Statutory Instrument that comes into force 11 June 2010.

This directive revises and combines the Framework Directive 96/62/EC, the first three "Daughter" Directives, covering sulphur dioxide, NO_x, PM, lead, benzene, carbon dioxide and ozone, and the "Exchange of Information Decision".

Three aspects are relevant to this Network:

- The previous PM legislation was based on control of PM_{10}, while the new Directive places an emphasis on $PM_{2.5}$. Research-led measurements such as those on this Network should therefore make due emphasis on the $PM_{2.5}$ size fraction.

- There is more explicit allowance for exceedences arising from "natural sources" to be excluded. Again, research-led measurements should make the determination of the "natural" fraction of PM a clear aim. Wind blown, long-range, transported mineral dust and sea salt are the most important natural sources, but there are many other sources with potentially significant effects that

can be attributed to natural sources. The implications for Member States are still being discussed. The issues have been summarised in *Contribution of natural sources to air pollution levels in the EU – a technical basis for the development of guidance for the Member States*, Report EUR 22779 EN (2007).

- Annex IV includes the statement "Measurement of $PM_{2.5}$ must include at least the mass concentration and appropriate compounds to characterise its chemical composition. At least the list of chemical species below shall be included."

SO_4^{2-}	Na^+	NH_4^+	Ca^{2+}	Elemental carbon (EC)
NO_3^-	K^+	Cl^-	Mg^{2+}	Organic carbon (OC)

Many of these components are those currently measured by this Network. There is, however, an evident need to standardise these measurements for EU reporting purposes, which is being addressed as described in the next section.

5.2.2 CEN standards

2009 has seen considerable CEN activity in the areas covered by this Network.

CEN TC 264 Working Group 15 is in the process of updating the gravimetric PM_{10} standard EN 12341:1998. One aim is to tighten the specifications for filter handling and transport in line with the EN 14907 standard for $PM_{2.5}$. The main outstanding issues are the specifications for "reference" filter material, and the humidity level during weighing. These issues have been the subject of experimental studies funded through JRC Ispra, and supplemented in the UK by Defra, that were mentioned above.

The working group is also in the process of drafting a CEN standard for automatic PM_{10} measurement.

CEN TC 264 WG 32 covers particle number concentration and size distribution measurements (ie CPC and SMPS-type). It is producing two separate Technical Specifications (as distinct from full Standards), covering:

1) A standard method for measuring "single parameter" particle number concentration, ie a "total" number concentration covering a broad size range, as typically covered by CPCs in ambient measurements. This will provide a "standard" low size cut-off, sampling, operating, QA/QC and calibration procedures, and be readily adoptable as a reference method. The WG is close to agreeing some key parameters such as standard CPC low size cut-off characteristics and calibration procedures that will be directly relevant to this Network.

2) Standard methods for measuring particle number concentration over more limited size ranges, as used to form size distributions, ie SMPSs, optical particle spectrometers, time-of-flight spectrometers, electrical low pressure impactors, etc, with appropriate sampling, operating, QA/QC and calibration procedures. This document will be addressed after the first one.

CEN TC 264 WG 34 covers anion and cation analysis, while CEN TC 264 WG 35 covers Elemental Carbon and Organic Carbon. These two groups held their first meetings in May 2009. In both cases documents are being drafted to provide guidance on methods for Member States to use from June 2010, but in view of the timescale these documents will not be CEN standards for the time being.

5.2.3 EUSAAR

The EUSAAR project (European Super-sites for Atmospheric Aerosol Research) includes 20 sites across Europe including Harwell. Amongst other topics it makes recommendations for standardizing measurements with CPCs and SMPSs, and of EC/OC.

For example, it has recommended that SMPS instruments should standardise the aerosol size distribution by sampling under dry conditions (<40% RH). Changes to sampling systems in the UK Network to conform with this have been implemented.

To a large extent the EUSAAR proposals are being dealt with within CEN WGs 32 (which includes Alfred Wiedensohler) and 35 (which includes Jean-Philippe Putaud).

5.3 ISO Standards

The most relevant standards being developed by ISO are within TC24 SC4.

ISO/DIS 15900, *Determination of Particle Size Distribution – Differential Electrical Mobility Analysis for Aerosol Particles* – i.e. measurements using SMPS. This was published in 2009. It is a very useful description of the principles of SMPS operation (for all purposes), but does not include requirements for calibrating the number concentration part of the measurement – i.e. CPCs, and other field operation requirements, which will be addressed by the CEN WG 32.

ISO Preliminary Work Item 27891, *Aerosol particle number concentration – calibration of condensation particle counters* is at an advanced draft stage. It will be aimed at users (especially those with internal calibration programmes), instrument manufacturers who certify their equipment, and specialist calibration laboratories (including NMIs). Traceability is ultimately expected to be to the NMI structure. This will be a very useful underpinning standard for the CEN WG 32 work.

The overlap in scope with CEN TC 264 WG32 has been noted. Paul Quincey (NPL) is a member of both committees.

6 Topic Reports and Publications

Reports and papers produced or published since the start of the contract include:

6.1 Project and Topic Reports

May 2005-April 2006

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). State of Network Report, NPL Report DQL-AS 019, September 2005

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Strategic Network Review, NPL Report DQL-AS 020, November 2005

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Annual Report 2005, NPL Report DQL-AS 028, Revised July 2006

May 2006-April 2007

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Annual Audit Report, NPL Report DQL-AS 031, July 2006

Comparison of Methods for Organic and Elemental Carbon PM_{10} Concentrations at Marylebone Road for the Period 07/09/06 to 31/12/06, NPL Report DQL-AS 035, February 2007

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Estimation of Measurement Uncertainty in Network Data, NPL Report DQL-AS 037, March 2007

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Annual Report 2006, NPL Report AS4, Revised April 2007.

May 2007-April 2008

Monitoring of Particulate Nitrate by Rupprecht & Patashnick 8400N Ambient Particulate Nitrate Monitors, A.M. Jones and R.M. Harrison, August 2007.

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2). Annual Audit Report, NPL Report DQL-AS 016, October 2007.

Comparison of Cluster Analysis Techniques Applied to Rural UK Atmospheric Particle Size Data, D.C.S. Beddows and R.M. Harrison, Draft, December 2007.

Change in particle number concentration from 2000 to 2006 at four UK sites, A.M. Jones and R.M. Harrison, March 2008.

The weekday-weekend difference and the estimation of the non-vehicle contributions to the urban increment of airborne particulate matter, A.M. Jones, J.Yin and R.M. Harrison,

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2), Annual Report 2007.

May 2008-April 2009

CPEA 28: Airborne Particulate Concentrations and Numbers in the UK (phase 2), Annual Report 2008.

The Merging of Atmospheric Particle Size Distribution Data Measured using Electrical Mobility and Time-of-Flight Analysers, David C. S. Beddows and Roy M. Harrison

Review and Interpretation of Black Carbon Data Measured by Magee Aethalometers, Alan M. Jones And Roy M. Harrison

The Temporal Trends in Particulate Sulphate and Nitrate Concentrations at UK Sites, Alan M. Jones And Roy M. Harrison

Quantifying the London Specific Component of PM10 Oxidative Activity,
Ian S Mudway, Gary Fuller, David Green, Chrissi Dunster and Frank J Kelly

6.2 Publications

Multisite Study of Particle Number Concentrations in Urban Air, R.M. Harrison and A.M. Jones, Environmental Science and Technology, **39**, 6063-6070 (2005).

The Use of Trajectory Cluster Analysis to Examine the Long-Range Transport of Secondary Inorganic Aerosol in the UK, S.S. Abdalmogith and R.M. Harrison, Atmospheric Environment, **39**, 6686-6695 (2005).

Interpretation of Particulate Elemental and Organic Carbon Concentrations at Rural, Urban and Kerbside Sites, A.M. Jones and R.M. Harrison, Atmospheric Environment, 39, 7114-7126 (2005).

Fine ($PM_{2.5}$) and Coarse ($PM_{2.5-10}$) Particulate Matter on a Heavily Trafficked London Highway: Sources and Processes, A. Charron and R.M. Harrison, Environmental Science and Technology, **39**, 7768-7776 (2005).

An Analysis of Spatial and Temporal Properties of Daily Sulphate, Nitrate and Chloride Concentrations at UK Urban and Rural Sites, S.S. Abdalmogith and R.M. Harrison, J. Environmental Monitoring, **8**, 691-699 (2006).

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What are the Sources and Conditions Responsible for Exceedences of the 24 h PM_{10} Limit Value (50 μg m^{-3}) at a heavily trafficked London site? A. Charron, R.M. Harrison and P.G. Quincey, Atmospheric Environment, **41**, 1960-1975 (2007).

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The Weekday-Weekend Difference and the Estimation of the Non-Vehicle Contributions to the Urban Increment of Airborne Particulate Matter, A.M. Jones, J. Yin and R.M. Harrison, Atmos. Environ., **42**, 4467-4479 (2008).

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Paul Quincey (2007) **A relationship between Black Smoke Index and Black Carbon Concentration**, Atmospheric Environment, Volume 41, Issue 36, Pages 7964-7968.

US EPA (2005) **Comparison of Integrated Filter and Semi-Continuous Measurements of $PM_{2.5}$ Nitrate, Sulfate, and Carbon Aerosols in the Speciation Trends Network (STN)**, US Environmental Protection Agency EPA 454/R-05-004, 2005.

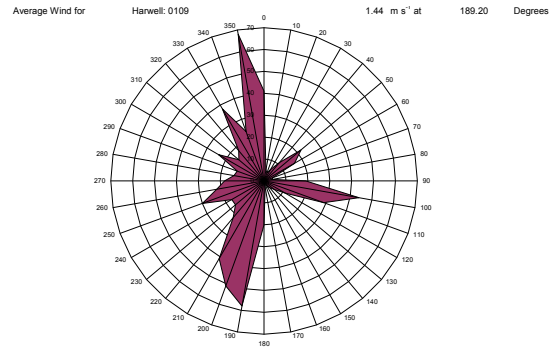
9 Acknowledgements

We wish to acknowledge the support provided by the Department for Environment, Food and Rural Affairs (Defra) and the Devolved Administrations (the Scottish Executive, Welsh Assembly Government and the Department of the Environment Northern Ireland) for this work under contract CPEA 28.

Annex 1. Wind Roses at Harwell and Rochester

Wind Roses – Harwell, January-June 2009

January 2009



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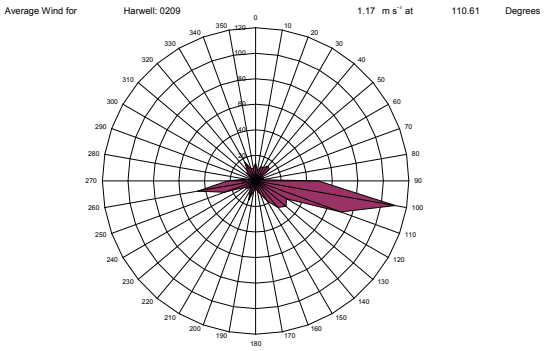

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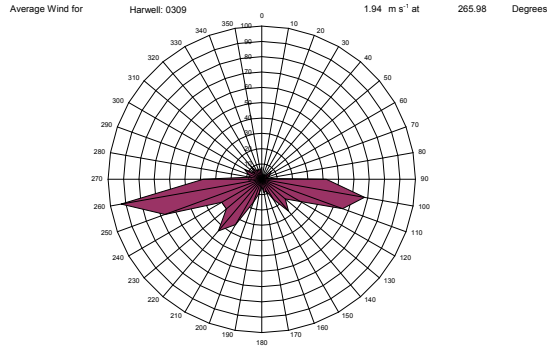

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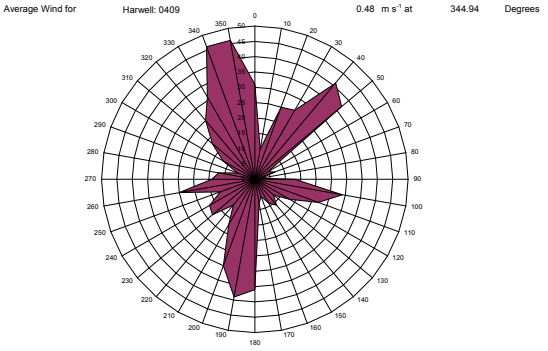

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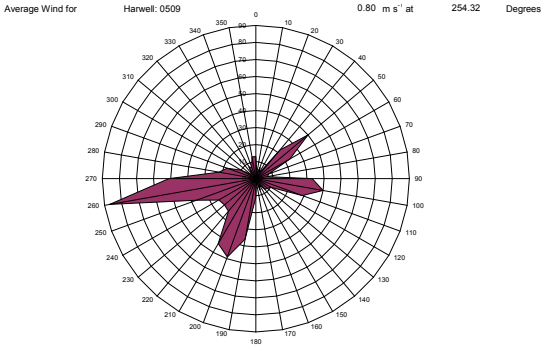

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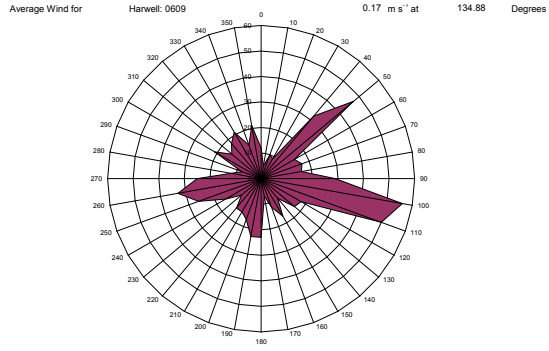
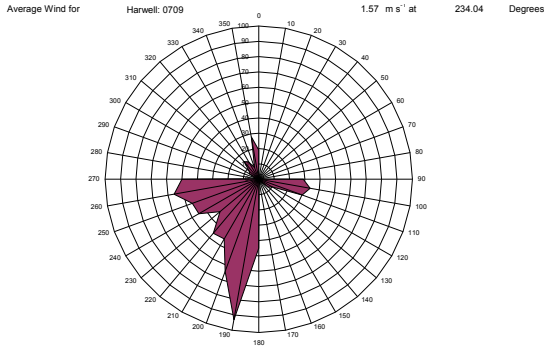


Wind Roses – Harwell, July-December 2009

July 2009



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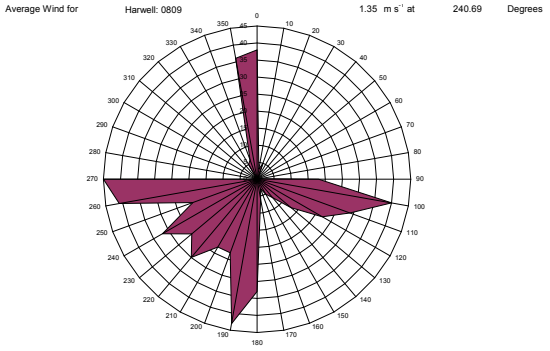

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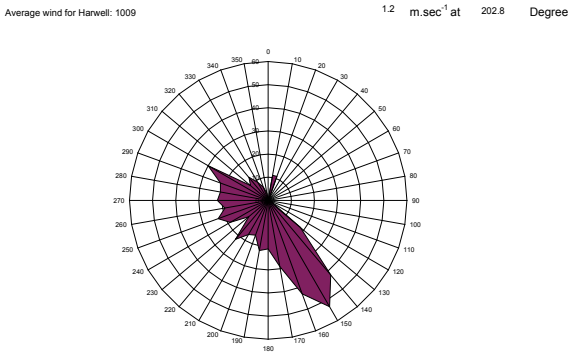
October 2009



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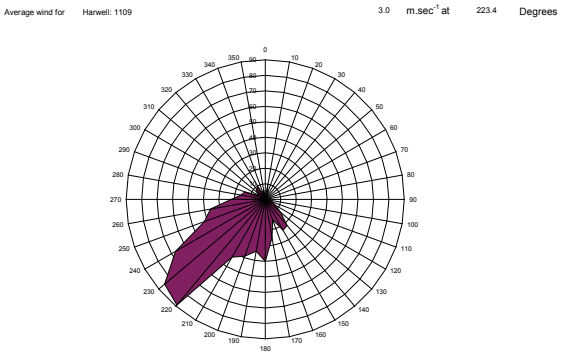

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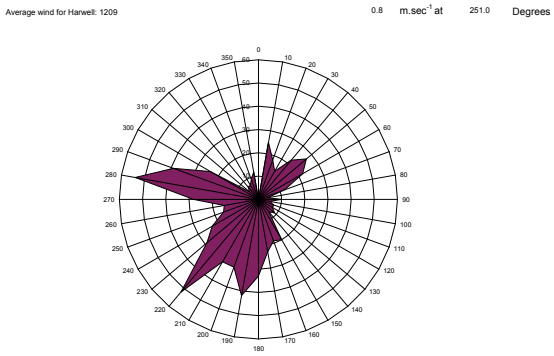
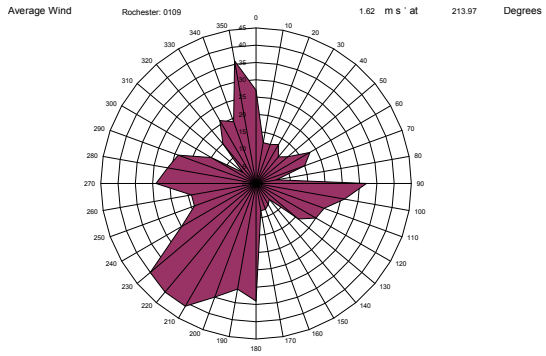


Wind Roses – Rochester, January-June 2009

January 2009



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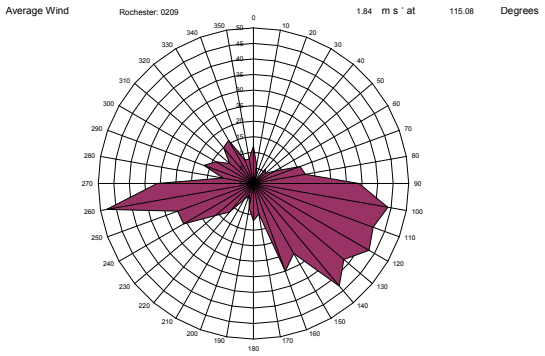

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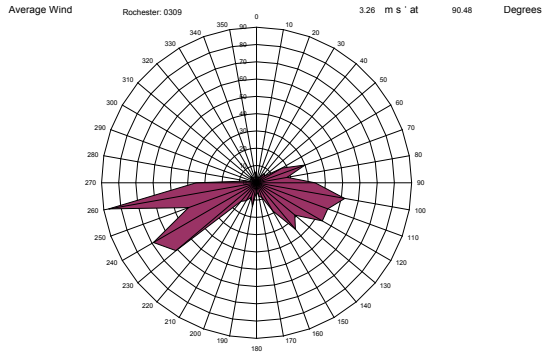

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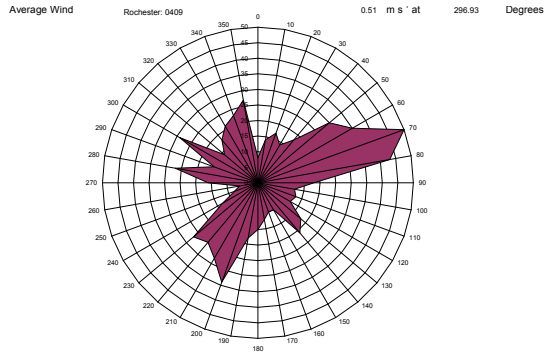

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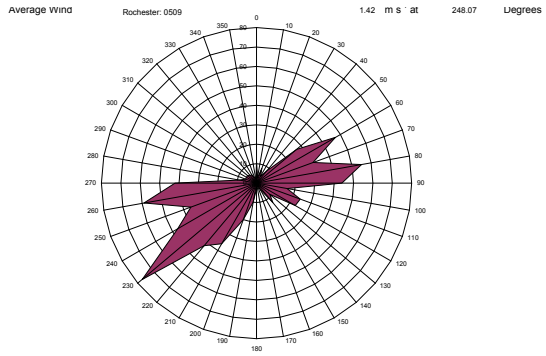

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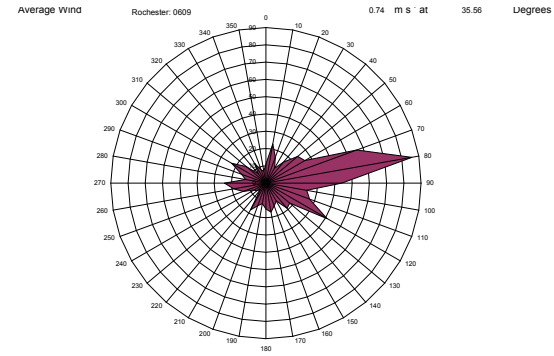
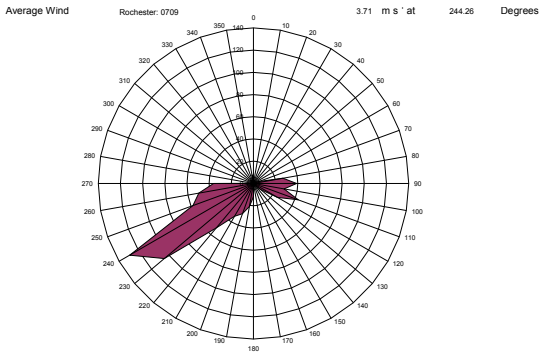


Wind Roses – Rochester, July-December 2009

July 2009



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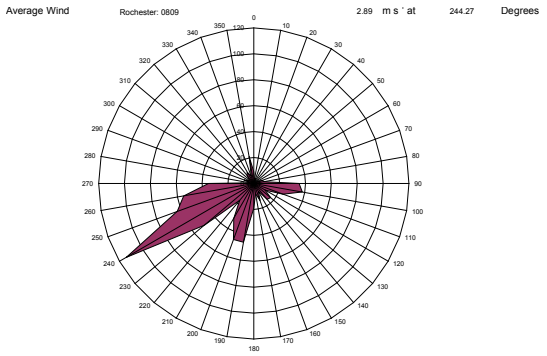

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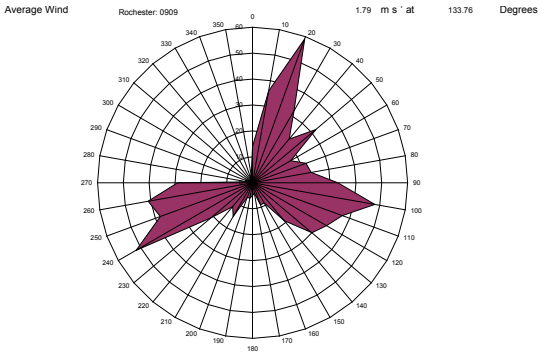

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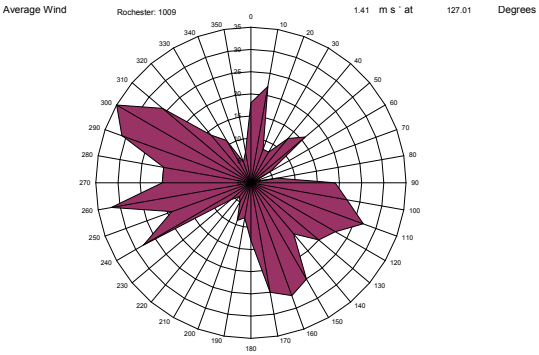

November 2009

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December 2009

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