Final Report **Project ER12**

PM$_{2.5}$ in the UK

December 2010

Dissemination status

Unrestricted

Project funders

Scottish Environment Protection Agency
Northern Ireland Environment Agency
Environment Agency

Whilst this document is considered to represent the best available scientific information and expert opinion available at the stage of completion of the report, it does not necessarily represent the final or policy positions of the project funders.

Research contractor

This document was produced by:

Prof. Duncan Laxen
Mr Stephen Moorcroft
Dr Ben Marner
Mr Kieran Laxen
Air Quality Consultants Ltd
23 Coldharbour Road
BRISTOL BS6 7HT

Dr Paul Boulter
Dr Tim Barlow
TRL Ltd
Crowthorne House
Nine Mile Ride
WOKINGHAM
RG40 3GA

Prof. Roy Harrison
Environmental Health and Atmospheric Sciences Research Group
University of Birmingham
BIRMINGHAM B15 2TT

Dr Mathew Heal
School of Chemistry
The University of Edinburgh
West Mains Road
EDINBURGH
EH9 3JJ

SNIFFER's project manager

SNIFFER's project manager for this contract is:

Vanessa Kind, SNIFFER

SNIFFER's technical advisory group is:

John Lamb, Scottish Environment Protection Agency – *Principal technical advisor*
Åsa Hedmark, Scottish Environment Protection Agency
Sarah Watkins, Environment Agency

SNIFFER
First Floor, Greenside House
25 Greenside Place
EDINBURGH EH1 3AA
Scotland
UK
www.sniffer.org.uk

Scotland & Northern Ireland Forum for Environmental Research (SNIFFER), Scottish Charity No SC022375, Company No SC149513. Registered in Edinburgh. Registered Office: Edinburgh Quay, 133 Fountainbridge, Edinburgh, EH3 9AG

EXECUTIVE SUMMARY

ER12: $PM_{2.5}$ in the UK

Project funders/partners: SNIFFER, Scottish Environment Protection Agency, Environment Agency, Northern Ireland Environment Agency

Background to research

It is recognised that exposure to airborne particulate matter (PM) can give rise to significant health effects and there is no evident safe level. Attention was initially directed towards PM of less than 10 μm diameter (PM_{10}), but epidemiological evidence regarding the health effects of smaller particles has now changed the focus to smaller particles less than 2.5 μm diameter ($PM_{2.5}$). As a consequence, new legislation has recently come into effect to control exposure to $PM_{2.5}$ (with a continuation of controls on PM_{10}), and the United Kingdom (UK) Government will need to develop approaches to reduce exposure to $PM_{2.5}$. To ensure this is carried out in the most cost-effective way it is first necessary to have a good understanding of the sources, pathways and health effects of $PM_{2.5}$, and of the legislation that can contribute to its control. The focus is on annual mean concentrations of $PM_{2.5}$, as these are the most significant in terms of health outcomes, and on concentrations within urban areas, where most people are exposed.

Objectives of research

This SNIFFER project is designed to provide regulators with an understanding of $PM_{2.5}$, including health effects, sources, speciation, pathways, regulatory requirements, monitoring and modelling tools. It will be used to advise regulators on how PM sources need to be regulated to achieve the air quality limit values and objectives.

Key findings and recommendations

Sources and behaviour of $PM_{2.5}$

- There are many different sources, both natural and anthropogenic, contributing to $PM_{2.5}$ concentrations. These can be primary particles, which are emitted directly into the atmosphere, or precursor gases, which form secondary particles through atmospheric reactions.
- Industrial sources and power stations contribute most to national primary anthropogenic emissions (35%), followed by road transport (24%), residential (13%) and shipping (10%).
- The $PM_{2.5}$ fraction of PM is removed only slowly from the atmosphere; the dispersion of $PM_{2.5}$ in the near-field can thus be treated like that of a gas.
- Primary anthropogenic particles (from all sources) make a relatively small contribution to urban background $PM_{2.5}$, probably less than 25% (although there is some uncertainty with regard to this estimate). Primary emissions from combustion sources are made up largely of elemental and organic carbon. These particles will have trace metals and trace organic compounds associated with them.

- In general terms, motor vehicle emissions (both exhaust, and brake and tyre wear) contribute slightly more than industrial sources (including point sources) to urban background $PM_{2.5}$.
- More specifically for $PM_{2.5}$ from traffic, the overall breakdown is broadly 64% vehicle exhaust, 32% brake and tyre wear, and 4% resuspended road particles. Thus around 36% of traffic emissions are likely to be from sources that are currently uncontrolled.
- Natural sources of PM include sea salt, which accounts for ~5-15% of urban background $PM_{2.5}$, with higher contributions found towards the western coastal areas of the UK, while windblown dust contributes ~5-8%.
- The formation of secondary particles happens relatively slowly (hours to days), thus secondary $PM_{2.5}$ is found well downwind of the sources of emission of the precursor gases. As a consequence there is a reasonably even distribution of secondary $PM_{2.5}$ on a regional scale.
- Secondary particles are mainly formed from emissions of sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOCs). Emissions of these precursor gases have declined significantly over the last three decades, especially for SO_2, followed by NO_x, VOCs and to a limited extent NH_3.
- Secondary particles dominate urban background $PM_{2.5}$ in the UK, accounting for some 30-50% of the $PM_{2.5}$ in urban areas. They are predominantly ammonium sulphate, ammonium nitrate, and organic particles. A significant proportion of secondary $PM_{2.5}$ is imported into the UK, having been formed from precursor emissions in continental Europe.

Exposure to $PM_{2.5}$

- Rural annual mean concentrations of $PM_{2.5}$ range from around 3.5 $\mu g/m^3$ in Scotland to around 10 $\mu g/m^3$ in southern England. Urban background concentrations are a few $\mu g/m^3$ higher, and are highest in central London at around 15 $\mu g/m^3$. Annual mean concentrations close to busy roads will be a few $\mu g/m^3$ higher still, although concentrations decline rapidly on moving away from the carriageway, such that they are indistinguishable from the background after about 20-50 m.
- Estimated population-weighted annual mean $PM_{2.5}$ concentrations for the UK in 2010 range from 5.5 $\mu g/m^3$ in Scotland, to 6.4 $\mu g/m^3$ in Northern Ireland, to 8.3 $\mu g/m^3$ in Wales, and to 10.6 $\mu g/m^3$ in England. These values will be determined largely by concentrations within urban areas, where the majority of the population is exposed. The population-weighted annual mean $PM_{2.5}$ concentration in inner London in 2010 is estimated to be 14.1 $\mu g/m^3$.

Health effects of $PM_{2.5}$

- Both short- and long-term exposure to $PM_{2.5}$ gives rise to a range of health effects. These include hospital admissions and mortality from respiratory and cardiovascular diseases.
- The effects of long-term exposure are more significant than those of short-term exposure, in terms of the overall impact on the nation's health.
- There is no recognised threshold below which there are no health effects.

- There is no clear evidence as to which of the $PM_{2.5}$ components give rise to the toxic effects. Thus, at this stage, all components need to be considered capable of giving rise to these effects.
- Interventions to reduce exposure to PM, such as the ban on coal burning in Dublin, have been shown to be beneficial in health terms, with fewer respiratory and cardiovascular deaths following the interventions.
- Exposure to $PM_{2.5}$ reduces life expectancy by around six months averaged over the whole of the UK. For those individuals who are particularly sensitive the reduction in life expectancy could be much greater. For instance, if 10% of the population is affected then the loss of life expectancy for these individuals would rise to an average of around 6 years. Whilst it is not straightforward to compare health risks, it is estimated that eliminating exposure to man-made $PM_{2.5}$ would yield greater benefits than eliminating road traffic accidents or exposure to passive smoking.
- Significant health benefits across the UK and European Union (EU) populations have been calculated for a given reduction in exposure to $PM_{2.5}$. These translate into financial benefits, which more than offset the costs of mitigation programmes currently in place.

Legislation to control exposure to $PM_{2.5}$

- The focus for $PM_{2.5}$ is on limiting long-term exposure through use of annual mean standards.
- There are two strands to the legislation to control exposure to $PM_{2.5}$:
  - exposure standards to define the level of control required; and
  - measures to limit emissions to meet the standards (including emission standards used to control emissions from both transport and industrial sources, and measures such as the National Emissions Ceilings Directive).
- Control strategies to reduce emissions that contribute to $PM_{2.5}$ include:
  - continued efforts to reduce emissions of precursor gases, to limit the formation of secondary PM, which will require national controls within a European-wide structure; and
  - continued efforts to reduce emissions from primary sources giving rise to the urban enhancement of $PM_{2.5}$ concentrations, which will need to focus on industrial sources, road transport and to a lesser extent on domestic sources.
- There is a large body of legislation already in place to control exposure to PM, much of it involving the control of emissions. This is often focused on PM in general and has not been specific to PM_{10} or $PM_{2.5}$. However, measures to control total PM and/or PM_{10} will in general result in lower $PM_{2.5}$ concentrations.
- The concentration standards to limit public exposure, which previously were focused on PM_{10}, have, in the last few years, been extended to include $PM_{2.5}$.
- A new exposure-reduction approach has been introduced for $PM_{2.5}$. This is in recognition of the absence of an identified safe level for exposure to PM, and requires a reduction of $PM_{2.5}$ background concentrations in urban areas across the UK over the period 2010 to 2020.

Measurements of $PM_{2.5}$ concentrations

- Little monitoring of $PM_{2.5}$ concentrations was carried out in the UK prior to the expansion of the national network (as part of the Automatic Urban and Rural Network (AURN)) in 2009 to meet the requirements of the EU Clean Air for Europe (CAFE) Directive. The AURN uses Filter Dynamic Measurement System (FDMS) analysers, which provide hourly mean concentrations that are equivalent to the EU reference method, *i.e.* the concentrations from these analysers are reference equivalent. The AURN currently comprises of 67 sites, of which 43 are classified as urban background[1], 17 as roadside/kerbside, 4 as industrial and 3 as rural.
- Additional monitoring is carried out by some local authorities, such as that undertaken within the London Air Quality Network (LAQN).
- Annual mean concentrations have been collated for 36 sites that had >90% data capture in 2009 (mostly AURN sites, but including some local authority sites). Urban background concentrations (27 sites) were mostly between 12-16 $\mu g/m^3$, with the highest concentration recorded at the London Eltham site in east London, at 17.6 $\mu g/m^3$.
- Analysis of the 2009 data indicates the following overall pattern for annual mean $PM_{2.5}$ concentrations:
  - rural background concentrations are ~3-10 $\mu g/m^3$, with the lowest values in Scotland and the highest in southern England;
  - urban background concentrations are ~ 3-6 $\mu g/m^3$ above the rural background in major urban areas;
  - roadside concentrations alongside busy roads are ~1-2 $\mu g/m^3$ above the urban background; and
  - kerbside concentrations alongside busy roads are ~7-8 $\mu g/m^3$ above the urban background.
- The limited evidence on trends in $PM_{2.5}$ concentrations over the last decade indicates:
  - no change in concentrations at rural background sites;
  - a very slight reduction in concentrations at urban background sites; and
  - a downward trend at roadside sites.
- The 2009 results show that concentrations of $PM_{2.5}$:
  - are substantially higher during the winter months than in the summer, with the highest monthly mean being around 10-15 $\mu g/m^3$ above the lowest monthly mean.
  - increase during the week, to give the highest levels on a Thursday/Friday, before dropping by around 4 $\mu g/m^3$ to a Sunday low. There is currently no full explanation available for this pattern; and
  - have a diurnal pattern that is consistent across central and southern parts of the UK (essentially south of the Lake District) with the lowest concentrations occurring during the afternoon and the highest during the early part of the night.

---

[1] There is one site, Harlington, officially classified as 'Airport', however this is around 1 km from Heathrow Airport, and as such, $PM_{2.5}$ concentrations will be affected by the airport to a very limited extent. It is thus better to treat this site as an urban background site for $PM_{2.5}$.

This may be due, at least in part, to loss of semi-volatile $PM_{2.5}$ from the atmosphere during the warmer part of the day, with this material then re-condensing during the evening and night time, although there is no clear-cut explanation for this pattern. The afternoon drop in concentrations is not apparent in the results for the northern parts of the UK.

- A detailed analysis taking into account wind direction and wind speed has shown a consistent pattern across the UK, with the highest concentrations being associated with winds from the northeast through to southeast. These higher concentrations are often associated with stronger winds >10 m/s, and occur throughout the day and night. This is evidence of a significant $PM_{2.5}$ contribution being imported from continental Europe, probably as secondary PM.

Modelling of $PM_{2.5}$

- Annual mean concentrations can normally be modelled with lesser uncertainty than is the case for short-term (hourly or daily mean) concentrations. Thus modelling of $PM_{2.5}$ should, in some respects, be more straightforward (than for PM_{10}, for which 24-hour concentrations are important) given that the focus is on the annual mean.
- A wide range of models can be applied to the modelling of $PM_{2.5}$, although these generally work at different spatial scales. Defra is currently undertaking studies to evaluate which models are best suited to meet policy needs and to identify those that should be investigated and developed.
- Two modelling suites are commonly used in the UK to estimate near-source concentrations of $PM_{2.5}$: ADMS (both ADMS4 for point sources and ADMS-Roads) and AERMOD. These are reasonably well-established and widely used for the assessment of development schemes, industrial permitting *etc*.
- There is only one national model applied routinely for $PM_{2.5}$. This is the semi-empirical Pollution Climate Mapping (PCM) model developed by AEA and used by Defra for policy development.

Policy implications

- Analysis of concentrations in relation to the EU limit values and UK objectives has shown that the key driver for action to control exposure to $PM_{2.5}$ will be the exposure-reduction targets that have been established. The UK target is for a 15% reduction in annual mean $PM_{2.5}$ at background locations across the major urban areas, while the EU target will be 10-15% (still to be determined, once monitoring results for the three years 2009-11 are available). These represent reductions in annual mean concentrations of around 1.5-2 $\mu g/m^3$ over the ten years between 2010 and 2020. If these reductions are to be achieved from the local sources that give rise to the urban background enhancement of around 3-6 $\mu g/m^3$, then these urban source contributions would need to be reduced by some 25-67%. On the other hand, if the whole of the reduction were to be achieved by reducing the secondary PM contribution, which accounts for ~30-50% of urban background $PM_{2.5}$ (around 4-6 $\mu g/m^3$), then the required reduction of this secondary PM would be some 25-50%. In either case the reductions required to meet what appears to be a small target reduction, are substantial. If both are tackled equally the percentage reductions would essentially be halved, but would still remain challenging.

- EU limit values established to ensure there are no hot-spots with excessive concentrations of $PM_{2.5}$ are unlikely to be exceeded in the UK. The PM_{10} objectives and limit values have been shown to be more stringent than the $PM_{2.5}$ objectives and limit values, thus the former will drive policies to reduce exposure in hot spots; this will, in general, help drive down $PM_{2.5}$ concentrations at these locations.
- The response of secondary PM concentrations to changes in precursor gas emissions is less than proportional. This implies that greater reductions in precursor gas emissions are required than might at first sight be the case. The chemistry of secondary inorganic PM formation is such that reductions in ammonia emissions may be more effective at reducing PM concentrations than equivalent reductions in sulphur dioxide and nitrogen oxides, although this requires further investigation. The chemistry of secondary organic PM formation is poorly understood and it is not clear which sources of non-methane VOCs (NMVOCs) should be targeted to reduce PM concentrations.

Recommendations

- The following recommendations are proposed for further work to provide a better understanding of $PM_{2.5}$ sources and concentrations, so as to allow appropriate control strategies to be developed. Their justification is described in Section 9:
    1. Modelling studies should be carried out to establish the contributions of UK and other EU emissions of precursor gases to annual mean $PM_{2.5}$ concentrations, and how these contributions will respond to changes in emissions. This will help guide the development of the most cost-effective control programme;
    2. A programme of field, chamber and modelling studies should be carried out to establish the contributions from anthropogenic and ‘natural’ sources of organic carbon to $PM_{2.5}$, and how these will respond to changes in emissions;
    3. Further work should be carried out on quantifying emissions, in particular from the road transport sector under ‘real-world’ driving conditions, and from industry. The contributions to urban background concentrations from other sources should be investigated using PM speciation studies. Such studies should then be used to inform the source contributions via modelling studies;
    4. Improved deterministic modelling approaches should be developed at the national and urban scale, with robust treatments of atmospheric dynamics, chemistry and aerosol processes, so as to give size and composition-resolved information on airborne concentrations. This will enable reliable projections to be made of future concentrations, to complement the findings of more empirical models.
    5. Seven additional $PM_{2.5}$ monitors should be set up at rural background sites, as a minimum. These could usefully be collocated with existing ozone monitors;
    6. Sufficient resources should be provided to ensure that the results of the PM monitoring programmes are subject to thorough analysis on an on-going basis;
    7. A programme of work should be established to support source apportionment of annual mean $PM_{2.5}$ at urban background locations in different areas of the UK; and
    8. Consideration should be given to ways of using the planning system to require reductions in $PM_{2.5}$ emissions to be incorporated into the planning of all new developments.

Delivery

- To deliver the additional work and subsequent control strategies:
  - The UK Government and devolved administrations will need to establish a work programme to define the sources contributing to urban background $PM_{2.5}$ in those urban agglomerations where compliance monitoring is being carried out and then predict the changes that will take place between 2010 and 2020, before developing measures to fill any short-fall in the predicted concentration reduction;
  - The Environment Agency, the Scottish Environment Protection Agency and the Environment Agency Northern Ireland should contribute to the understanding of primary and secondary emissions from industrial, agricultural and waste sources that they regulate. They should also assist in modelling the contribution of primary $PM_{2.5}$ emissions to urban background concentrations and identifying measures to limit primary and secondary emissions; and
  - Local Authorities could be expected to help implement national measures to reduce primary $PM_{2.5}$ emissions, as well as consider ways to control $PM_{2.5}$ from new developments. They could also help develop amendments to the Clean Air Act to control emissions from biomass boilers.

Key words: $PM_{2.5}$, health effects, ambient, emissions, sources, concentrations, legislation, monitoring, modelling.

LIST OF TABLES

LIST OF FIGURES

1. INTRODUCTION

Particulate matter (PM) in ambient air is a complex mixture of organic and inorganic substances. It is derived from a wide variety of sources, both natural and anthropogenic, and displays a range of physical and chemical properties. Particles are termed either ‘primary’, where they are emitted directly into the atmosphere, or ‘secondary’, where they are formed in the atmosphere by reactions between gases.

During the late 1980s and early 1990s the results of epidemiological studies in the USA identified PM less than 10 μm diameter (PM_{10}) as a key pollutant metric related to acute (short-term) and chronic (long-term) health effects. More recently, the growing body of research has pointed towards the smaller particles within PM_{10} as being the most significant in relation to health outcomes. In particular, attention has focused on PM less than 2.5 μm diameter ($PM_{2.5}$) as a metric more closely associated with adverse health effects than PM_{10}[2], although there is still considerable debate as to whether it is actually the ultrafine fraction ($PM_{0.1}$) (or indeed a non-mass metric, *e.g.* particle number) that is primarily responsible for the effects. A number of studies have also identified longer-term exposure to PM as being more significant than the short-term (daily) exposure to higher levels that had first been linked to health effects. These long-term effect studies have formed the basis for calculation of health outcomes from exposure to PM in the UK and Europe, which are not insubstantial.

The emphasis has thus shifted from the PM_{10} metric to $PM_{2.5}$, although it is still recognised that there are also health effects associated with exposure to the coarser particles within PM_{10}, termed either $PM_{2.5\text{-}10}$ or PM_{COARSE}. Until recently, however, the UK monitoring network has included relatively few measurements of $PM_{2.5}$, and the data are still relatively limited in terms of duration and geographical coverage.

Legislation to control exposure to PM developed during the 1990s. The focus was initially on controlling exposure to short-lived peak concentrations, as the epidemiological evidence at the time indicated that health effects were primarily associated with these peaks. A 24-hour standard of 50 $\mu g/m^3$ was therefore introduced in both Europe and the UK[3]. The use of such air quality standards has meant that control strategies have primarily been aimed at reducing pollutant concentrations at so-called ‘hot spots’, where monitoring has shown the standard can be exceeded. These hot spots have most commonly been identified alongside busy roads.

The re-orientation of attention towards $PM_{2.5,}$ coupled with the evidence that long-term concentrations were more significant in health terms than short-term peaks, has led to changes in legislation. The UK introduced the idea of a $PM_{2.5}$ standard in its Air Quality Strategy update (Defra, 2007). This was followed shortly after by the EU introducing its Clean Air for Europe (CAFE) Directive in 2008, which also included standards for $PM_{2.5}$. In both cases a new approach was introduced in recognition of the absence of a threshold for exposure to PM. This new approach involved a reduction in the overall exposure of the urban population to $PM_{2.5}$, based on the concept that greater benefits could be obtained from this general reduction in exposure than by a policy aimed at reducing exposure at hot spots. The CAFE Directive was implemented in UK legislation on 11 June 2010.

The current position is that exposure to $PM_{2.5}$ is constrained principally by the requirements of the CAFE Directive, which sets an exposure-reduction target that will involve a reduction in the 3-year average urban background concentrations of between

---

[2] Clearly $PM_{2.5}$ is a part of the PM_{10} metric, so the two metrics are not independent.

[3] An annual mean standard was also introduced, but in practice this was less stringent than the 24-hour standard.

0% and 20% between 2010 and 2020. The precise value for the reduction to be applied in the UK has yet to be announced, but is expected to be 10%. This will be supplemented by an annual mean target value of 25 $\mu g/m^3$ (to be met in 2010) a limit value of 25 $\mu g/m^3$ (to be met in 2015) and a 3-year average urban background concentration of 20 $\mu g/m^3$ (to be met by 2015). The limit value is mandatory, while the target is non-mandatory. The UK has set its own exposure-reduction target of 15% between 2010 and 2020 for background concentrations averaged across UK urban areas, with a 'backstop' annual mean objective of 25 $\mu g/m^3$ (to be met by 2020, *i.e.* this is less stringent than the EU limit value). The Scottish Government has set its own more stringent objective of an annual mean of 12 $\mu g/m^3$ (to be met at all relevant-exposure locations by 2020).

Both in the EU and the UK the current standards for PM_{10} are being retained. In practice it is expected that the annual mean target, limit value and objective for $PM_{2.5}$ of 25 $\mu g/m^3$ will be met if the UK 24-hour objective for PM_{10} (no more than 35 daily exceedences of 50 $\mu g/m^3$) is met.

The various standards for PM are set out in Table 1.

Meeting the requirements of the CAFE Directive is the responsibility of the UK Government. Some of the UK objectives have been set in Regulations, which require local authorities to take them into account (the obligation is for local authorities to develop Air Quality Action Plans in pursuit of the objectives where they are exceeded at points of relevant exposure, as defined in guidance set out in Defra (2009)). The PM_{10} objectives are set in Regulations, with the Scottish Government having set more stringent objectives than the rest of the UK. The $PM_{2.5}$ objectives are not set in Regulations, and thus only have an advisory role.

A number of legislative approaches are being taken to control exposure to PM in order to achieve the standards just described. These include controls on motor vehicle emissions, controls on industrial sources and controls introduced by local authorities to tackle hot spots. The National Emission Ceilings (NEC) Directive (2001/81/EC) underpins the controls at the national level. This sets national limits on emissions, with a date by which they are to be achieved. In relation to PM, the key controls have been on emissions of nitrogen oxides (NO_x), sulphur dioxide (SO_2) and ammonia (NH_3), as these are precursors of secondary inorganic PM (largely ammonium nitrate (NH_4NO_3) and ammonium sulphate ($(NH_4)_2SO_4$)), and on emissions of volatile organic compounds (VOCs), as these are precursors of secondary organic PM. The current version of the NEC Directive does not include protection of the public from exposure to (secondary) PM as an aim. A revision of the NEC Directive is currently underway, which includes a specific focus on PM, both as primary and secondary emissions. The NEC Directive leaves it to Member States to determine which control strategies to implement to meet the emission ceilings.

These matters are all explored in greater detail in this report. The first sections set out a review of the published literature to address the following issues:

- sources and pathways of $PM_{2.5}$;
- exposure to $PM_{2.5}$ in the UK;
- health effects of $PM_{2.5}$; and
- legislation to control exposure to $PM_{2.5}$,

This is followed by an analysis of $PM_{2.5}$ measurements in the UK and a description of approaches used to model $PM_{2.5}$. Finally, the policy implications of the findings are set out, together with recommendations for further work.

The approach applied throughout is to address a series of questions covering each of these topics.

Table 1 **Standards for $PM_{2.5}$ and PM_{10}.**

	Pollutant	**Time Period**	**Standard**	**To be achieved by**
UK	$PM_{2.5}$	Annual mean	Objective of 25 µg/m^3	2020
		3-year running annual mean	Objective of 15% reduction in concentrations measured across urban background sites	Between 2010 and 2020
	PM_{10}	24-hour mean	Objective of 50 µg/m^3 not to be exceeded more than 35 times a year	2005
		Annual mean	Objective of 40 µg/m^3	2005
Scottish	$PM_{2.5}$	Annual mean	Objective of 12 µg/m^3	2020
		3-year running annual mean	Objective of 15% reduction in concentrations measured across urban background sites	Between 2010 and 2020
	PM_{10}	24-hour mean	Objective of 50 µg/m^3 not to be exceeded more than 7 times a year	2005
		Annual mean	Objective of 18 µg/m^3	2005
European	$PM_{2.5}$	Annual mean	Target value of 25 µg/m^3	2010
		Annual mean	Limit value of 25 µg/m^3	2015
		Annual mean	Stage 2 indicative limit value of 20 µg/m^3	2020
		3 year Average Exposure Indicator (AEI)	Exposure-reduction target relative to the AEI depending on the 2010 value of the 3 year AEI (ranging from a 0% to a 20% reduction)[a]	2020
		3 year Average Exposure Indicator (AEI)	Exposure-concentration obligation of 20 µg/m^3	2015
	PM_{10}	24-hour mean	Limit value of 50 µg/m^3 not to be exceeded more than 35 times a year	2004
		Annual mean	Limit value of 40 µg/m^3	2004

[a] the exposure-reduction targets are outlined further in Table 14 in Section 6.3.4.

2. WHAT ARE THE CHARACTERISTICS AND BEHAVIOUR OF PM IN THE ATMOSPHERE?

2.1 What is the Distinction between Primary and Secondary Particles?

One of the most important distinctions when dealing with airborne PM is that between primary and secondary particles. Primary particles are emitted directly into the atmosphere from emission sources. Secondary particles are not emitted directly, but are formed by reactions between gas-phase components of the air. These reactions lead to the formation of substances of low volatility which consequently condense into the solid or liquid phase, thereby becoming particles (AQEG, 2005).

Primary and secondary particles are derived from a wide variety of sources, both natural and anthropogenic. The land and the sea are both major natural sources of primary particles, through entrainment of soils by the wind and the generation of marine aerosols by the bursting of air bubbles entrained in breaking waves. There are many anthropogenic sources of primary particles, including combustion (*e.g.* road-vehicle exhaust, domestic heating, and industry) and mechanical processes (*e.g.* quarrying, agriculture, tyre wear). Secondary particles generally result from atmospheric oxidation processes, and the substances oxidised may be either natural or anthropogenic in origin. These processes are not immediate and may occur over many hours or days. Secondary particles are thus formed many kilometres downwind of the source of the precursor emissions. Secondary particles are therefore less easy to ascribe to their original sources (AQEG, 2005).

2.2 What are the Physical Characteristics of Airborne Particles?

2.2.1 What are the sizes of airborne particles?

The behaviour of particles in the atmosphere and within the human respiratory system is determined to a large extent by their physical properties, notably their size. Moreover, particles in individual size modes are characterised by rather distinct sources, composition, chemical properties, lifetimes in the atmosphere and distances over which they can travel.

Particles in the atmosphere typically exhibit three distinct size modes, as shown in Figure 1. These are the nucleation mode, the accumulation mode and the coarse particle mode. The relative magnitudes of the modes in the size distribution depend on the nature of the contributing sources of PM, but crucially also upon the metric (mass, surface area or particle number). The nucleation mode has traditionally been defined as particles with a diameter of less than 0.05 micrometres (µm) (50 nanometres (nm))[4]. These particles are large in number but each one is so small that they form only a small proportion of the total mass. Accumulation mode particles range in size from around 50 nm to around 1 micrometre (µm). The coarse mode consists of particles larger than around 1 µm. At high relative humidity the larger particles in the accumulation mode can also reach 3 µm in diameter (USEPA, 2009). Particle number is most highly concentrated in the size range below 100 nm, but volume (or mass) is most concentrated in the larger size ranges.

---

[4] These are sometimes referred to as 'nanoparticles'. Some texts also describe the smallest particles, of 10 nm and below, as the nucleation mode and the particles falling between 10 and 50 nm as the Aitken mode (AQEG, 2005).

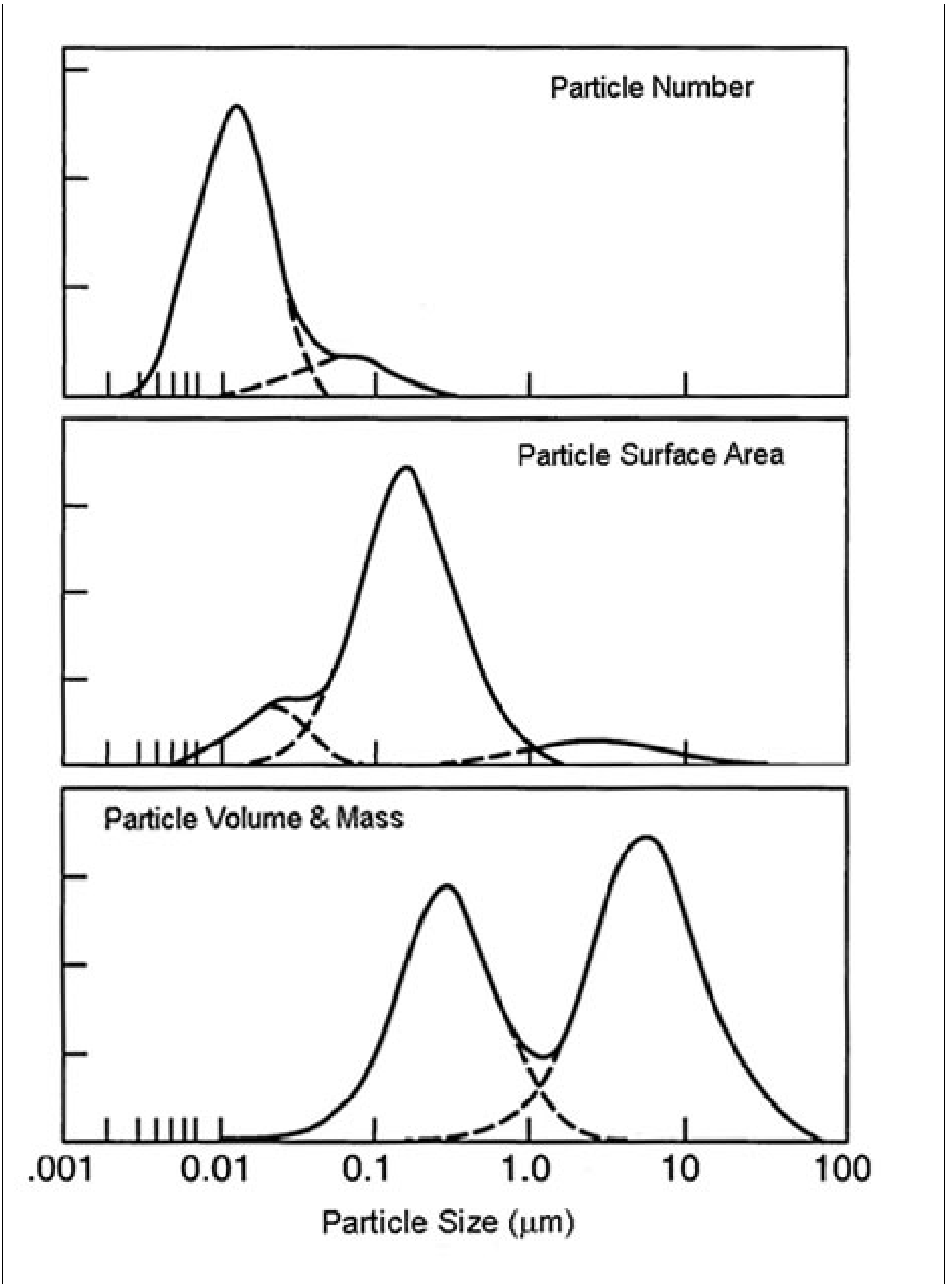

Figure 1 Representative size distribution of airborne particles in terms of particle number, surface area, and volume/mass. Based on Finlayson-Pitts and Pitts (2000). The volume to mass equivalence assumes uniform particle density.

Particles in the nucleation mode are newly formed, either by homogeneous nucleation[5] in the atmosphere or by nucleation processes that occur within the emissions from high-temperature combustion sources (leading to the emission of primary nucleation mode particles). Growth of nucleation mode particles, mainly by vapour condensation but also as a result of coagulation processes, leads to formation of accumulation mode particles. Secondary particles in the accumulation mode can also be formed via gas-to-particle transformations in the atmosphere. Coarse particles are usually generated by mechanical processes, such as the action of wind raising dust from bare soils and stockpiles, the breaking of waves giving rise to salt particles, the resuspension of road dust by passing traffic, and the handling of dusty materials during quarrying and construction activities (AQEG, 2005).

2.2.2 **What shapes are airborne particles?**

Particles appear in a wide variety of shapes, including spheres, ellipsoids, cubes, and irregular or fractal geometries, frequently occurring as agglomerations of particles. The shapes of particles and particle agglomerations are important for determining their optical properties, and also affect the area in contact with the surface they are deposited on, including cell membranes (USEPA, 2009).

2.2.3 **What terms are used to describe different particle size fractions?**

A variety of terminology is used to describe particles, being different for particle formation, ambient monitoring and occupational exposure. The terminology is summarised in Figure 2 in relation to particle size. $PM_{2.5}$ particles, that are the focus of this review, include all nucleation and accumulation mode particles and a fraction of particles in the coarse mode. $PM_{2.5}$ particles therefore do not match a natural break point in the particle size distribution. In terms of ambient particle measurements it has become convention to call $PM_{2.5}$ ‘fine particles’, while those between $PM_{2.5}$ and PM_{10} are ‘coarse particles’.

Ambient particles are usually described in terms of their ‘aerodynamic diameter’, as it is the aerodynamic behaviour of the particles that determines their separation during ambient sampling. Particles come in a wide variety of shapes, often as randomly shaped agglomerations. The aerodynamic diameter is the equivalent diameter of a spherical particle of unit density having equivalent aerodynamic behaviour (USEPA, 2009). In practice the size separation is not exact, with a proportion of particles larger than the cut point being collected, and a proportion of those smaller than the cut point not being collected. The median size of the particles passing through the size separator defines the size, thus $PM_{2.5}$ particles are those particles that pass through a size-selective inlet with 50% efficiency at 2.5 µm.

In terms of occupational exposure, the American Conference of Governmental Industrial Hygienists, the International Standards Organisation, and the European Standardisation Committee have adopted a 50% cut point of 4 µm as an indicator of ‘respirable’ particles, *i.e.* those particles that penetrate deep into the lung. However, in practice $PM_{2.5}$ is usually used as an indicator of respirable particles, even though it has a different cut point. The ‘thoracic’ fraction includes, in addition to the respirable fraction, all particles that penetrate below the larynx and into the lung and is thus similar to PM_{10}. Particles in the thoracic fraction larger than 4 µm, *i.e.* particles that tend to get trapped in the nasal passage way and upper airways, are sometimes referred to as ‘tracheobronchial’ or ‘thoracic coarse’ (USEPA, 2009). $PM_{2.5\text{-}10}$ is used as an indicator of this component of coarse particles.

---

[5] Homogeneous nucleation is a particle formation process which takes place within a uniform substance.

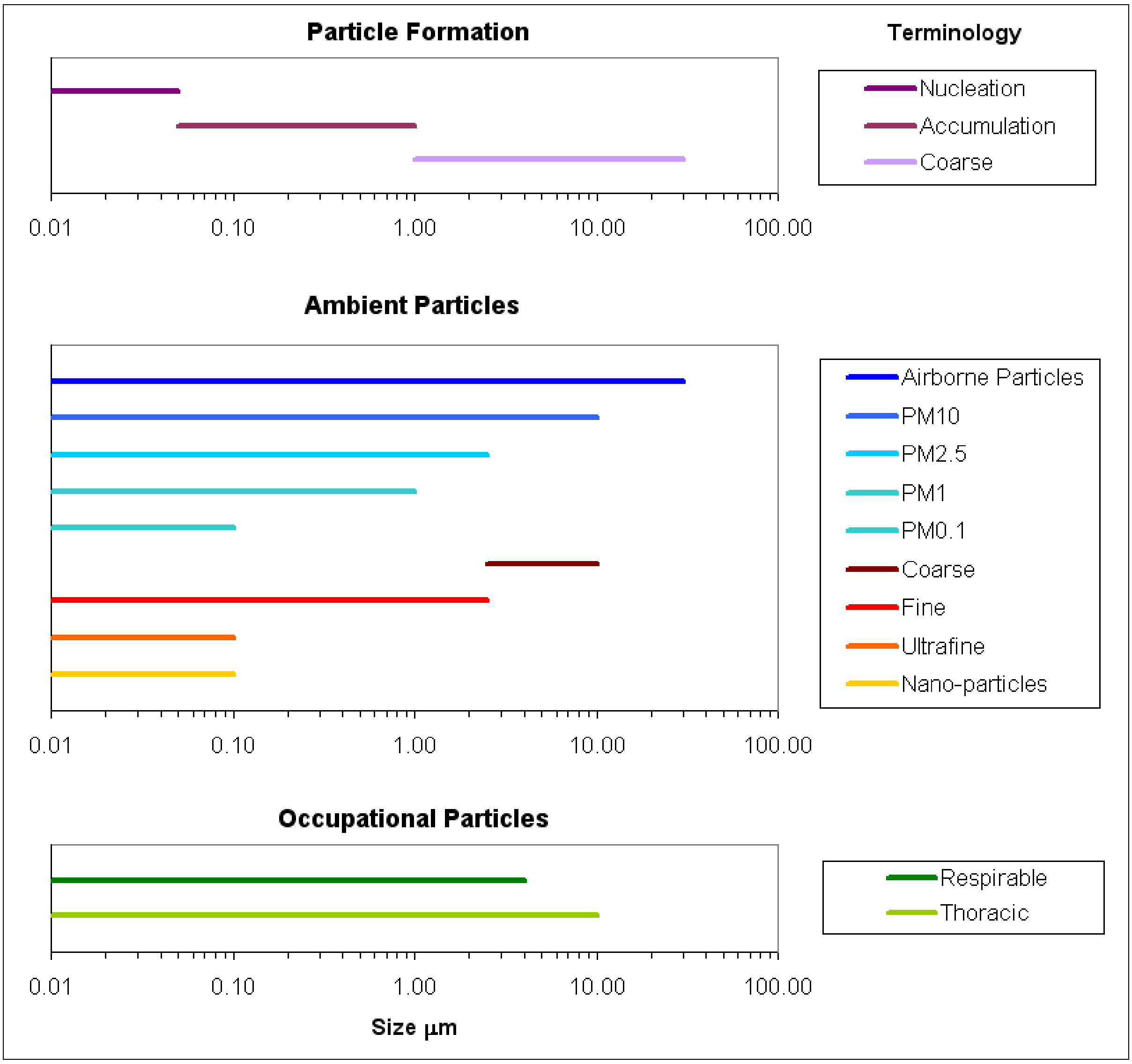

Figure 2 Terminology applied to different particle sizes (note logarithmic scale).

2.2.4 How is $PM_{2.5}$ measured in ambient air?

The reference method for the sampling and measurement of $PM_{2.5}$ is that described in *EN 14907:2005 Standard gravimetric measurement method for the determination of the $PM_{2.5}$ mass fraction of suspended particulate matter*. It is based on sampling ambient air through a filter, at a controlled flow rate for 24 hours. The filter is weighed under carefully controlled conditions prior to sampling and again following sampling. The mass difference between the two weighings is deemed to be attributable to the PM accumulated on the filter during sampling. A mean mass concentration, expressed in microgrammes per cubic metre ($\mu g/m^3$), is then calculated from the volume of air sampled and the mass of particulate material collected.

According to Directive 2008/50/EC, a Member State may use any alternative method which it can demonstrate displays a consistent relationship with the reference method. This may involve the results being corrected to produce concentrations equivalent to those that would have been achieved using the reference method.

Further details on monitoring $PM_{2.5}$ are provided in Section 7.

2.3 How Highly Correlated are $PM_{2.5}$ and PM_{10}?

$PM_{2.5}$ is a sub-set of PM_{10}, and therefore the correlation between measurements of the two ought to be high. According to De Leeuw and Horálek (2009), $PM_{2.5}$ and PM_{10} daily averages should have a correlation coefficient of at least 0.7 over a year. Horálek *et al.* (2008) showed that the correlation between $PM_{2.5}$ and PM_{10} exhibited no inter-annual variation and the correlation coefficients were between 0.86 and 0.88. They also noted that the correlation is lower at locations near roads, due to the direct emissions of coarse particles from road traffic (*e.g.* tyre and brake wear and resuspension). Heal *et al.* (2005) found a similar high correlation between daily $PM_{2.5}$ and PM_{10} at an urban background site in Edinburgh, with a correlation coefficient of 0.87. This is covered in greater detail using recent national monitoring data in the UK, in Section 7.

2.4 How do $PM_{2.5}$:PM_{10} Ratios Vary in Ambient Air?

De Leeuw and Horálek (2009) examined co-located $PM_{2.5}$ and PM_{10} measurements from 35 European countries (around 230 sites) for the period 2004-2006, taken from *AirBase*[6]. The annual mean concentrations of PM_{10} and $PM_{2.5}$ are shown by site type in Figure 3. The upper limit of the $PM_{2.5}$:PM_{10} ratio was typically around 0.8. However, the authors cautioned about the use of the ratios they identified, as information on the monitoring method and any adjustments carried out was not available. For instance, it was common practice in the UK during this time, and in other European countries, to report PM_{10} concentrations collected using a TEOM analyser that had been adjusted using a default 1.3 factor. This was known to be highly uncertain and the TEOM has since been shown to be unsuitable for reporting PM_{10}. If $PM_{2.5}$ was also collected using a TEOM analyser it is not clear what a suitable adjustment factor would have been (it would not be expected to be the same as the factor for PM_{10}). Thus, ratios between $PM_{2.5}$ and PM_{10} measured with a TEOM analyser would be highly uncertain. With this caveat, the results presented by De Leeuw and Horálek (2009) are sufficient to provide an indication of the overall ratios in rural, urban and traffic locations in different parts of Europe.

De Leeuw and Horálek (2009) calculated $PM_{2.5}$:PM_{10} ratios for four different European regions:

- *Northern Europe*: Norway, Sweden, Finland, Estonia, Lithuania, Latvia, Denmark and Iceland.
- *North-Western Europe*: United Kingdom, Ireland, the Netherlands, Belgium, Luxembourg, France north of 45 degrees latitude.
- *Central and Eastern Europe*: Germany, Poland, Czech Republic, Slovakia, Hungary, Austria, Switzerland, Liechtenstein.
- *Southern Europe*: France south of 45 degrees latitude, Portugal, Spain, Andorra, Monaco, Italy, San Marino, Slovenia, Croatia, Greece, Cyprus, Malta, Albania, Bosnia Herzegovina, Bulgaria, Romania.

The resulting values (Table 2) ranged from 0.42 to 0.78. In the North and Central-East regions of Europe the values were highest at rural sites and lowest at traffic sites, indicative of an increasing contribution of locally-emitted coarse particles at the traffic sites (which may be due to winter sanding of the roads). However, in North-Western and Southern Europe there was no such tendency; a possible explanation for the low rural ratio in Southern Europe might be a contribution of mineral (Saharan) dust. The rural sites in North-Western Europe had a ratio which was low compared with those at urban and traffic sites. This was believed to be a feature of the distribution of monitoring sites. When rural / urban comparisons were confined to nearby sites (within 75 km) the rural sites had a ratio 10% above the urban ratio. This is the source of the adjusted values in Table 2.

---

[6] http://air-climate.eionet.europa.eu/databases/airbase/

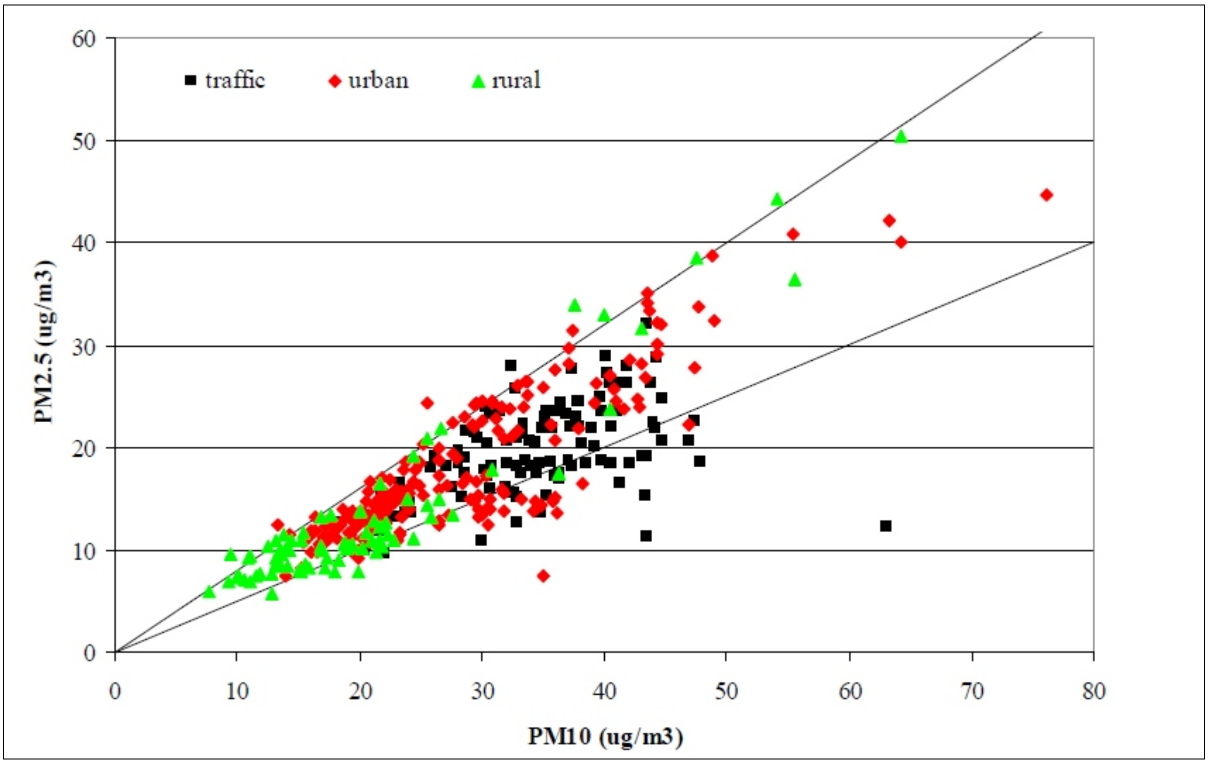

Figure 3 Annual mean concentrations of PM_{10} and $PM_{2.5}$ (period 2004-2006). The lines correspond to $PM_{2.5}$:PM_{10} ratios of 0.8 and 0.5. Source: De Leeuw and Horálek, 2009.

Table 2 $PM_{2.5}$:PM_{10} ratios as a function of region and site type. Source: De Leeuw and Horálek (2009).

Region of Europe	Rural	Urban	Traffic
Northern	0.78	0.55	0.42
North-Western	0.53/0.69*	0.63	0.59
Central-Eastern	0.75	0.71	0.65
Southern	0.57/0.64*	0.58	0.53
Europe	0.62	0.65	0.58

* The second value has been adjusted – see text.

Heal *et al.* (2005) used Partisol samplers to measure daily $PM_{2.5}$ and PM_{10} levels at an urban background site in Edinburgh for 12 months in 1999/2000. They found an average $PM_{2.5}$:PM_{10} ratio of 0.54, although on a daily basis the ratio varied highly, from 0.32 to 0.95. The Edinburgh average is somewhat lower than that given in Table 2 for urban sites, but well within the range shown in Figure 3.

2.5 How does $PM_{2.5}$ Relate to Other Particle Metrics?

In the European aerosol phenomenology study Putaud *et al.* (2010) observed a poor correlation between particle number and $PM_{2.5}$ mass concentration averaged over 10 µg/m^3 $PM_{2.5}$ concentration bins. However, the particle number concentration increased with $PM_{2.5}$ at most sites, but less than proportionally because the number of ultrafine particles does not generally increase with $PM_{2.5}$. In each geographical sector in Europe, the particle number:$PM_{2.5}$ mass ratio increased from rural to near-city, to urban, and to kerbside sites. The ratio between ultrafine and total particle number concentration decreased with increasing $PM_{2.5}$ concentration at all sites but one.

2.6 How does $PM_{2.5}$ Relate to Gaseous Pollutants?

Data from the USA show that PM_{10} and $PM_{2.5}$ are correlated better with each other than with the gaseous pollutants (*e.g.* SO_2, NO_2, CO and O_3). Correlations between PM size fractions, and between PM and some gaseous pollutants (*e.g.* CO), vary both seasonally and spatially between and within metropolitan areas. There is relatively little seasonal variability in the mean correlation between $PM_{2.5}$ (or PM_{10}) and SO_2 and NO_2. In the case of ozone the correlation is stronger than for other constituents in the summer (correlation coefficient 0.56), while in winter there is a negative correlation (correlation coefficient -0.30) (USEPA, 2009). This is likely to reflect the photochemical formation of both ozone and $PM_{2.5}$ in the summer months, while in winter an increasing $PM_{2.5}$ concentration will reflect increasing nitrogen oxides concentrations, which will remove ozone.

2.7 How Spatially Variable is $PM_{2.5}$?

The spatial variability of $PM_{2.5}$ across Europe is illustrated in Figure 4. The highest concentrations are found in Northern Italy and in Eastern Europe. Concentrations over much of England are in the 10-15 $\mu g/m^3$ concentration range, being lower in Scotland, Northern Ireland and Wales (see Section 4.1 for further details of the pattern of $PM_{2.5}$ concentrations across the UK). Background concentrations in Belgium, the Netherlands, and northern Germany are somewhat higher than those in England, falling in the 15-25 $\mu g/m^3$ range.

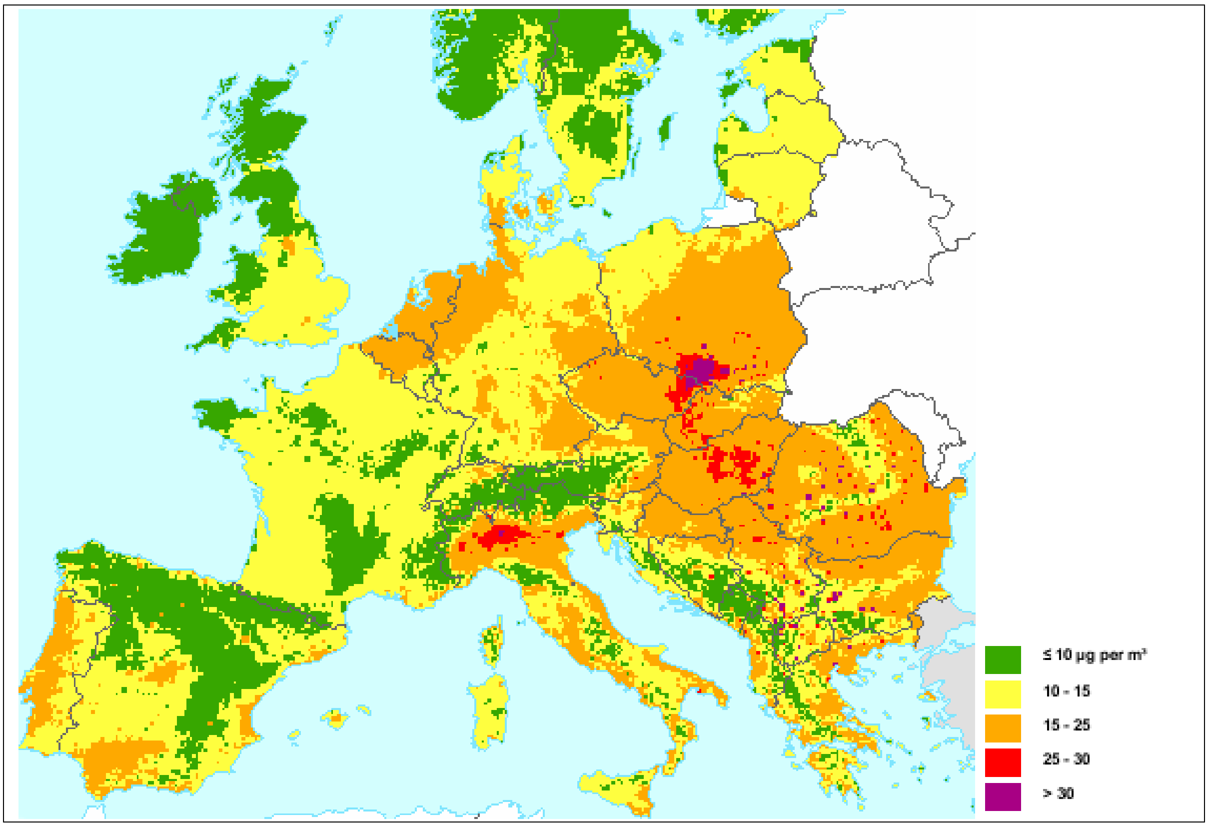

Figure 4 Background $PM_{2.5}$ concentrations mapped across Europe in 2005. Source: De Leeuw and Horálek, 2009.

Data from the United States show that $PM_{2.5}$ is less spatially variable than PM_{10} across urban areas (USEPA, 2009). For instance, in Boston the correlation between 24-hour $PM_{2.5}$ concentrations was high (correlation coefficient over 0.8) for samplers up to 70 km apart. This contrasted with PM_{10}, for which the correlation was poorer (correlation coefficients more typically 0.6 to 0.8). This reflects the greater role of local sources for PM_{10}, and the more regional nature of $PM_{2.5}$. Further information on correlation between 1-hour $PM_{2.5}$ data from samplers up to 140 km apart in southern England is provided in Section 7.5.4.

$PM_{2.5}$ concentrations have been mapped across London using a semi-empirical model (see Section 4.1) and show limited spatial variability in background concentrations (*i.e.* concentrations away from the direct influence of nearby sources) Figure 5. The M25 motorway, which encircles Greater London, stands out as a source giving rise to slightly higher background concentrations in the nearby 1 x 1 km grid squares. Concentrations in Central London rise to just over 15 µg/m^3, which is only some 3-5 µg/m^3 above values in outer London of 10-12 µg/m^3.

Figure 5 Background $PM_{2.5}$ concentrations mapped across Greater London in 2010. Source: Defra (2010a).

Concentrations vary to a greater extent closer to sources. Annual mean roadside concentrations decline rapidly on moving away from the edge of the road. This is illustrated in Figures 6 and 7 (Hickman *et al.*, 2002). Figure 6 shows PM_{10} concentrations measured at different distances from the edge of the M25 using MiniVol samplers. The measurements were taken over 28 days during the winter of 1998/99. Figure 7 shows PM_{10} and $PM_{2.5}$ concentrations measured at the same location during a 14 day period in 2000. The values have been normalised to 100% at 20 m. A best-fit log-linear relationship has been fitted to the PM_{10} data in Figure 6. The same relationship is shown in Figure 7. The results are broadly similar for both PM_{10} and $PM_{2.5}$, with a sharp decline over about the first 30 m from the carriageway. Beyond about 20-50 m from the edge of the road the concentrations become largely indistinguishable from the local background. It is also clear that the roadside increment is only around 30-40% above background levels.

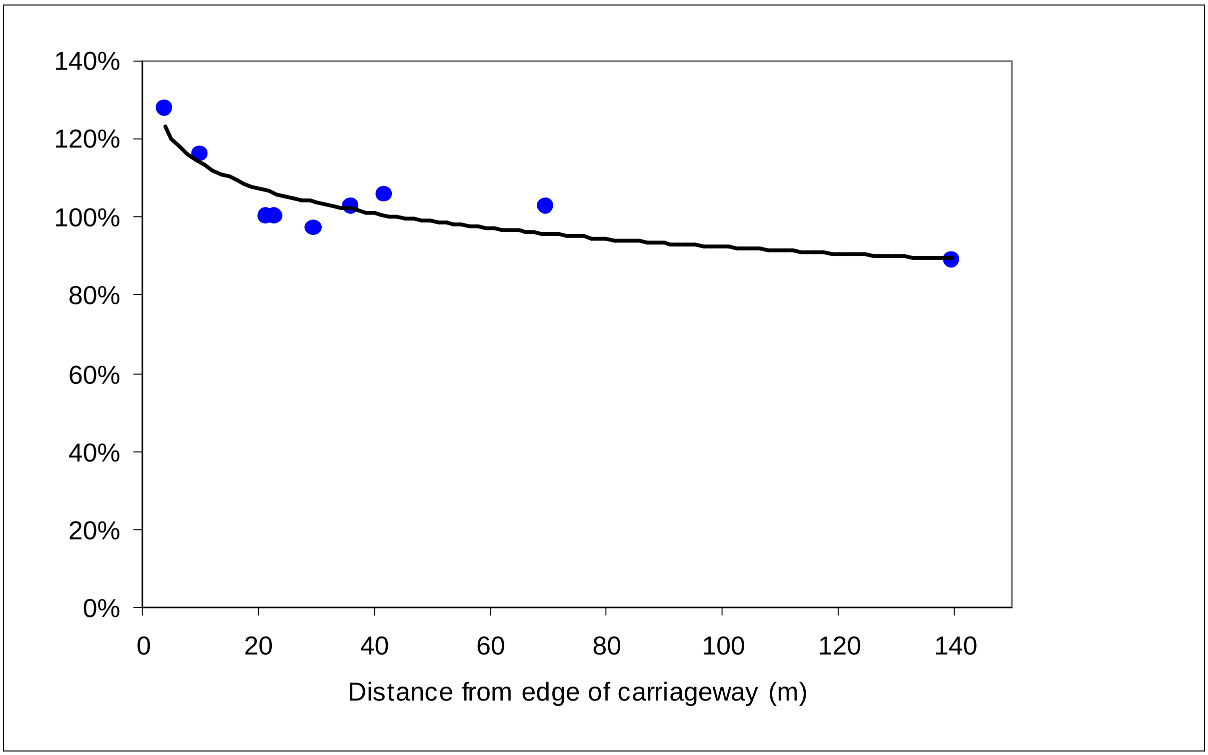

Figure 6 **PM_{10} concentrations measured on a transect away from the M25 motorway, over 28 days in 1998/99. Values normalised to 100% at ~20 m from the edge of the carriageway. Best fit logarithmic relationship. Source: Hickman *et al*. (2002).**

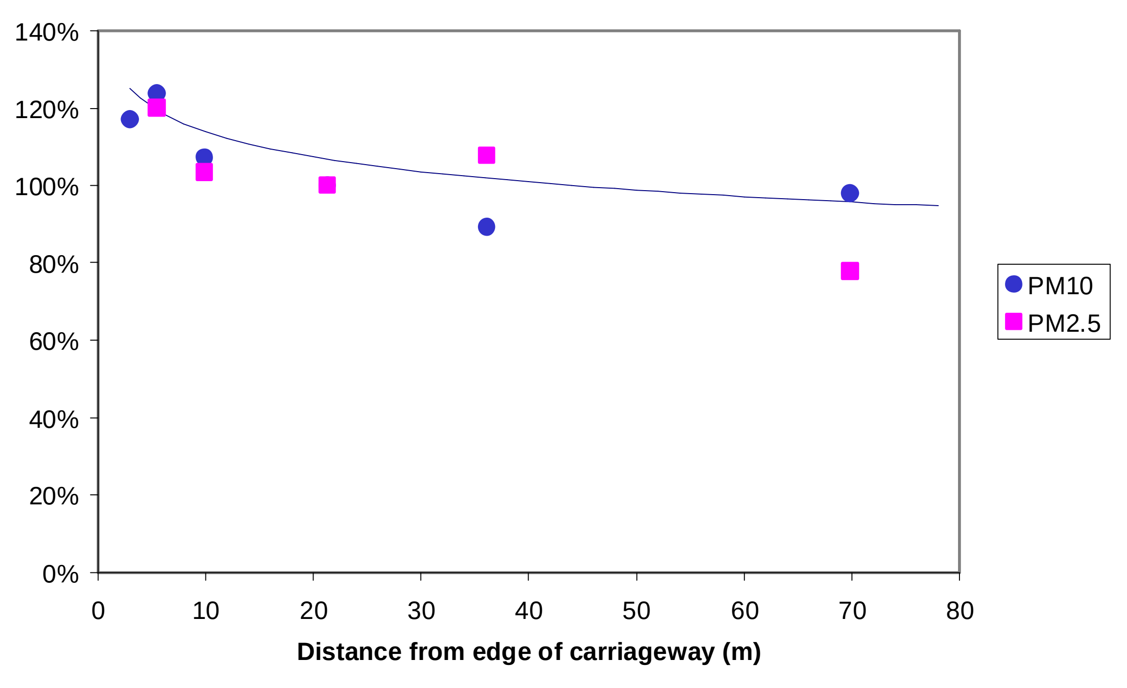

Figure 7 **PM_{10} and $PM_{2.5}$ concentrations measured on a transect away from the M25 motorway, over 14 days in 2000. Values normalised to 100% at ~20 m from the edge of the carriageway. Best fit logarithmic relationship taken from Figure 6. Source: Hickman *et al*. (2002).**

Concentrations around industrial sources will depend on the height of the emission, and whether building downwash affects dispersion. Emissions from power stations burning coal are emitted from tall stacks, at a height of over 100 m, and maximum contributions to ground-level annual mean PM_{10} concentrations have been estimated at ~0.25 µg/m^3, with contributions to 24-hour concentrations of much less than 5 µg/m^3 (AQEG, 2005). $PM_{2.5}$ contributions will be somewhat smaller. Emissions from Port Talbot steelworks have contributed to exceedences of the 24-hour mean PM_{10} objective; however, analysis of PM results collected with Partisol samplers in 2003, showed that the steelworks were contributing PM_{COARSE} particles with no clear contribution to $PM_{2.5}$. The $PM_{2.5}$ concentrations appeared to be dominated by long-range transport from the east (AQEG, 2005). Measurement of PM_{10} and $PM_{2.5}$ around the steelworks in Scunthorpe have recently been analysed by the Environment Agency (2009a). Pollution roses allowed different sources to be identified, some of which contributed $PM_{2.5}$, including the coke ovens and blast furnaces, while others contributed to PM_{10}, without contributing significantly to $PM_{2.5}$, including the sinter plant and coal handling plant.

2.8 What are Airborne Particles Typically Composed of?

Airborne particles are made up of a large number of organic and inorganic substances. The chemical composition varies in time and space, and also with particle size. Due to the processes of condensation and coagulation, particles can become internally mixed, *e.g.* an organic coating on a carbon particle, with the constituents having arisen from different sources. In contrast, in externally mixed aerosols, the individual particles have a composition that reflects the particular source (AQEG, 2005).

The main constituents of the three particle modes, and typical sources of these constituents, are given in Table 3 and Table 4. Although much of this information is from work in the USA, it will be equally applicable in the UK.

Some general statements are possible for the different size modes. For example, particles in the nucleation mode are composed mainly of lower volatility organic and sulphur compounds, but also some solid carbon and metals. Secondary particles are composed mainly of ammonium sulphate and nitrate. These originate from the oxidation of sulphur and nitrogen oxides in the atmosphere to acids, which are then neutralised by atmospheric ammonia derived mainly from agricultural sources.

The major constituents usually represent at least a few per cent of the particle mass. Either in addition to or within some of the major constituents there are many minor chemical constituents, the detection of which is often a function of the sensitivity of the analytical procedure. These include trace metals and trace organic compounds. Trace metals, such as lead, cadmium, mercury, nickel, chromium, zinc and manganese are used in metallurgical processes. Some occur as impurities or additives in fuels and others are used in industrial products. Concentrations are generally very low. The total mass of organic compounds can represent a significant part of the overall mass, but is made up of a very large number of individual organic compounds, each of which is present at a very low concentration. Such organic compounds include aliphatic and aromatic hydrocarbons, heterocyclic compounds, and oxygenates such as aldehydes, ketones and carboxylic acids (AQEG, 2005).

It should be recognised that considerable uncertainty may apply to results of particle composition studies due to changes in composition (and possibly morphology) that can occur during the sampling, collection and analysis of aerosol samples. For example, particles may be coated with semi-volatile material that can evaporate when subjected to certain analytical procedures. (USEPA, 2009).

Table 3 Constituents of airborne particles (adapted from USEPA (2009)).

	Nucleation	Accumulation	Coarse
Formation process	Combustion, high-temperature processes, and atmospheric reactions.	Combustion, high-temperature processes, and atmospheric reactions.	Break-up of large solids/droplets.
Formed by	Nucleation of atmospheric gases including H_2SO_4, NH_3 and some organic compounds. Condensation of gases.	Condensation of gases. Coagulation of smaller particles. Reactions of gases in or on particles. Evaporation of fog and cloud droplets in which gases have dissolved and reacted.	Mechanical disruption (crushing, grinding, abrasion of surfaces). Evaporation of sprays. Suspension of dusts. Reactions of gases in or on particles.
Composed of	Sulphate. Elemental Carbon. Metal compounds. Organic compounds with very low saturation vapour pressure at ambient temperature.	Sulphate, nitrate, ammonium, and hydrogen ions. Elemental carbon. Large variety of organic compounds. Metals: compounds of Pb, Cd, V, Ni, Cu, Zn, Mn, Fe, *etc*. Particle-bound water. Bacteria, viruses.	Nitrates / chlorides / sulphates from HNO_3/ HCl / SO_2 reactions with coarse particles. Oxides of crustal elements (Si, Al, Ti, Fe). $CaCO_3$, $CaSO_4$, NaCl, sea salt. Bacteria, pollen, mould, fungal spores, plant and animal debris.
Solubility	Not well characterised	Largely soluble, hygroscopic[7], and deliquescent[8].	Largely insoluble and non-hygroscopic.
Sources	High temperature combustion. Atmospheric reactions of primary gaseous compounds	Combustion of fossil and biomass fuels.	Resuspension of particles deposited.

---

[7] A hygroscopic material is one which attracts moisture from the atmosphere.

[8] Deliquescent substances have a strong affinity for moisture, and will absorb water from the atmosphere to dissolve.

Table 4 Sources of major constituents of airborne particles. Source: AQEG (2005).

Component	Notes
Sulphate	Mainly a secondary component from atmospheric oxidation of SO_2, although there may be a small primary component from sea salt or mineral matter such as gypsum.
Nitrate	Normally present as ammonium nitrate (NH_4NO_3), which results from the neutralisation of HNO_3 vapour by NH_3, or as sodium nitrate ($NaNO_3$), due to displacement of hydrogen chloride from NaCl by HNO_3 vapour.
Ammonium	Generally present in the form of ammonium sulphate ($(NH_4)_2SO_4$) or NH_4NO_3.
Sodium and chloride	From sea salt.
Elemental carbon	Black, graphitic carbon formed during the high-temperature combustion of fossil and contemporary biomass fuels.
Organic carbon	Carbon in the form of organic compounds, either primary (from automotive or industrial sources) or secondary (from the oxidation of VOCs).
Minerals	Crustal materials are rich in elements such as Al, Si, Fe and Ca. These are present in coarse dusts that arise from processes such as wind-driven entrainment, quarrying, construction and demolition.
Water	Water-soluble components, especially $(NH_4)_2SO_4$, NH_4NO_3 and NaCl, take up water from the atmosphere at high relative humidity, turning from crystalline solids into liquid droplets. The transition occurs at a specific relative humidity known as the deliquescence point, at which dry crystals are exposed to increasing relative humidity. There is, however, a hysteresis effect: upon a reduction in the relative humidity, water is lost only slowly and is retained at relative humidities well below the deliquescence point. This means that particles sampled from the atmosphere, even after drying at 40–50% relative humidity (as is customary before weighing), will still retain bound water, often representing a significant component of the mass.

As well as requiring the monitoring of $PM_{2.5}$ mass concentrations, Directive 2008/50/EC calls for measurements of the chemical composition of $PM_{2.5}$ at rural background sites. Measurements are required of the following minimum list of chemical species (which are part of $PM_{2.5}$): sulphate (SO_4^{2-}), sodium (Na^+), ammonium (NH_4^+), calcium (Ca^{2+}), elemental carbon (EC), nitrate (NO_3^-), potassium (K^+), chloride (Cl^-), magnesium (Mg^{2+}) and organic carbon (OC). Compositional analysis allows the assessment of sources and is also a check on the mass measurement (*i.e.* the sum of the mass of the chemical compounds should be equal to the directly measured mass).

2.9 What are the Chemical Characteristics of $PM_{2.5}$?

2.9.1 What are the chemical characteristics of $PM_{2.5}$ in Europe?

When considering $PM_{2.5}$ in the Netherlands, Matthijsen and ten Brink (2007) observed that more than 75% of the regional $PM_{2.5}$ is composed of secondary inorganic constituents (sulphate, nitrate and ammonium), elemental carbon (EC) and organic carbon (OC). EC contributed 20% to the 'total carbon' (TC), and derived mostly from diesel traffic emissions. The contribution of natural sources to $PM_{2.5}$ was much lower than the contribution to PM_{10}.

Putaud *et al.* (2010) carried out a detailed analysis of the chemical composition of PM in samples collected by various researchers across Europe. The annual average chemical composition of PM_{10}, $PM_{2.5}$ and PM_{COARSE}, is shown in Figure 8 and Table 5. It was found that the main constituents of $PM_{2.5}$ (and PM_{10}) all over Europe were generally organic matter (OM), SO_4^{2-} and NO_3^-. Mineral dust ('min dust' in the Figure) and sea salt were generally the main constituents of PM_{COARSE}. Mineral dust can also be a major constituent of PM_{10} at kerbside sites. The other constituents are elemental carbon (EC), ammonium (NH_4^+) and carbonaceous matter (CM), with a proportion unaccounted for ('unacc').

OM was the major single component of PM_{10} at almost every site where its contribution was determined. However, PM_{10}, $PM_{2.5}$ and PM_{COARSE} exhibited differences across Europe: on average there was more carbonaceous matter in PM_{10} in Central Europe, more nitrate in North-Western Europe, and more mineral dust in all fractions in Southern Europe.

There was no general relationship between PM mass concentration and PM chemical composition for each type of site in all three sectors of Europe (North-Western, Central and Southern Europe). The NO_3^- contribution to $PM_{2.5}$ (and PM_{10}) increased with PM mass concentrations at almost all urban background sites, and at several other locations across Europe. It was therefore argued that NO_x emissions should be particularly targeted to reduce the number of exceedences of the 24-hour PM_{10} limit value (Putaud *et al.*, 2010).

For $PM_{2.5}$, between-sector comparisons were only possible for urban sites. Some of the conclusions for PM_{10} also applied to $PM_{2.5}$ (mineral contribution larger in Southern Europe, sea salt percentage smaller in Central Europe, and total carbon (TC) percentage smaller in North-Western Europe), but others did not. For instance, the $SO_4^{2-}/PM_{2.5}$ and $NO_3^-/PM_{2.5}$ ratios in Southern Europe were significantly lower than in the other sectors.

Differences in PM chemistry between the three types of site were also observed within each sector of Europe. Both SO_4^{2-} and NO_3^- contributions to PM_{10} generally decreased when moving from rural to kerbside sites. The same was observed for NO_3^- in PM_{COARSE} and for SO_4^{2-} in both $PM_{2.5}$ and PM_{COARSE} in Central Europe. In contrast, the contribution of EC to PM_{10} increases from rural to urban to kerbside sides in all sectors. The contribution of OM to $PM_{2.5}$ was similar at all types of site.

It was also noted by Putaud *et al.* (2010) that there are still very few sites where all the major constituents of PM_{10} and $PM_{2.5}$ have been measured over time periods long enough to obtain representative averages, and even less is known about sub-micrometre and ultrafine particle number concentrations. Another issue is the lack of control on data comparability. There are currently no reference methods for measuring aerosol characteristics, except for PM mass concentrations. Even for this metric, the reference methods (EN12341 and EN14907 for PM_{10} and $PM_{2.5}$ respectively) do not guarantee the comparability of measurements performed at various locations across Europe, since significant sampling and analytical artefacts remain, which are PM chemistry and meteorology dependent.

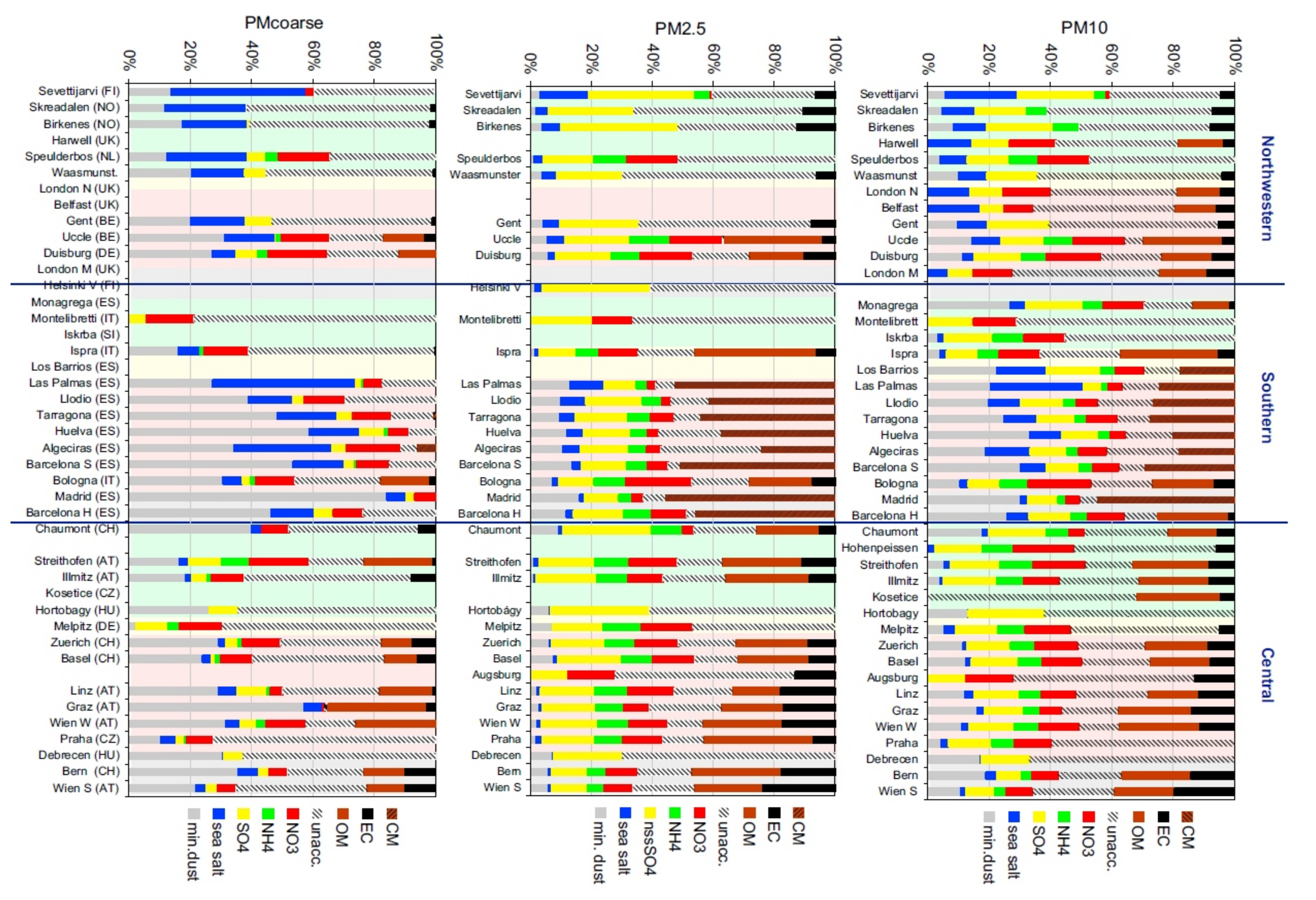

Figure 8 PM_{10}, $PM_{2.5}$ and PM_{COARSE} annual mean chemical composition by site and region. Pastel background colours indicate the site types (light green: rural background, light yellow: near-city, rose: urban background, grey: kerbside). Source: Putaud et al. (2010).

Table 5 Major constituents of PM_{10}, $PM_{2.5}$ and PM_{COARSE}. Source: Putaud *et al.* (2010).

		PM_{10}			$PM_{2.5}$			PM_{COARSE}		
		Rural	**Urban**	**Kerbside**	**Rural**	**Urban**	**Kerbside**	**Rural**	**Urban**	**Kerbside**
N-Western Europe	Min. dust	4%	12%			5%	1%		26%	
	Sea salt	12%	10%	7%		4%	1%		15%	
	SO_4^{2-}	13%	14%	8%		21%	18%		6%	
	NO_3^-	16%	14%	12%		16%			20%	
	OM	15%	18%	16%		25%			14%	
	EC	4%	5%	9%		7%			1%	
	TC	*15%*	*18%*	*20%*		*25%*			*12%*	
Southern Europe	Min. dust	15%	21%	28%		11%	14%		42%	69%
	Sea salt	3%	12%	5%		6%	2%		22%	11%
	SO_4^{2-}	16%	12%	12%		15%	15%		4%	5%
	NO_3^-	14%	9%	8%		7%	7%		11%	9%
	OM		26%			23%			13%	
	EC		6%			8%			2%	
	TC	*13%*	*21%*	*28%*		*30%*	*38%*		*11%*	
Central Europe	Min. dust	9%	12%	15%	3%	5%	6%	22%	25%	29%
	Sea salt	2%	2%	2%	1%	1%	1%	3%	3%	5%
	SO_4^{2-}	19%	15%	9%	17%	19%	12%	4%	4%	4%
	NO_3^-	13%	12%	8%	6%	13%	10%	7%	7%	6%
	OM	23%	21%	21%	15%	22%	26%	15%	15%	13%
	EC	6%	10%	17%	5%	14%	21%	3%	3%	10%
	TC	*32%*	*32%*	*38%*	*19%*	*31%*	*35%*	*14%*	*14%*	*19%*

Water is a normal component of PM, but the amount present is very variable and depends on the measurement method. Water binds to hydrophilic components in PM such as sulphate, ammonium, nitrate and sea salt. Reducing emissions of SO_2, NO_x and NH_3 lowers the concentration of their secondary PM components and therefore reduces the overall $PM_{2.5}$ concentration. Lower secondary PM levels may also reduce the uptake of water by fine particles. This leads, in turn, to a further reduction in the $PM_{2.5}$ concentration. In this way water can magnify trends in secondary PM. The amount of water associated with secondary inorganic aerosol is, however, highly uncertain (Matthijsen and ten Brink, 2007).

2.9.2 What are the chemical characteristics of $PM_{2.5}$ in the UK?

Yin and Harrison (2008) collected $PM_{2.5}$ from three sites in and around Birmingham. A central urban background site was sampled for 12 months, a roadside site for 6 months (mainly summer) and a rural site for 6 months (mainly winter). The samples were taken sequentially, not consecutively. A pragmatic mass closure model was used to assign compounds to the measured constituents. This model is a way of accounting for the measured mass concentration of airborne particles through summing scaled values of the concentrations of major chemical components in such a way as to allow for those components that are not directly measured (Harrison *et al.*, 2003). The overall results for the central urban background site are shown in the top pie-chart in Figure 9. Three components account for a large proportion of the total mass: nitrates, sulphate and organics. Iron-rich dusts are attributed to non-exhaust traffic emissions, calcium salts are attributed to construction/demolition dust and wind-blown soil and sodium chloride to sea salt (although this should also include re-suspended de-icing salt). Elemental carbon will relate to primary combustion. Organic carbon (organics) will be both from combustion sources and natural sources. The $PM_{2.5}$ composition seen in Birmingham is similar to that reported by Putaud *et al.* (2010) for north-western Europe (see Table 5)

Yin and Harrison (2008) also report the $PM_{2.5}$ composition on high pollution episode days (*i.e.* days with PM_{10} >50 $\mu g/m^3$). They report a substantial increase in nitrates, which account for just under half the $PM_{2.5}$ on episode days (lower pie-chart in Figure 9). The authors note this emphasises the importance of developing a better understanding of the sources of nitrate in airborne PM. Putaud *et al.* (2010) also report higher nitrate contributions at higher $PM_{2.5}$ concentrations.

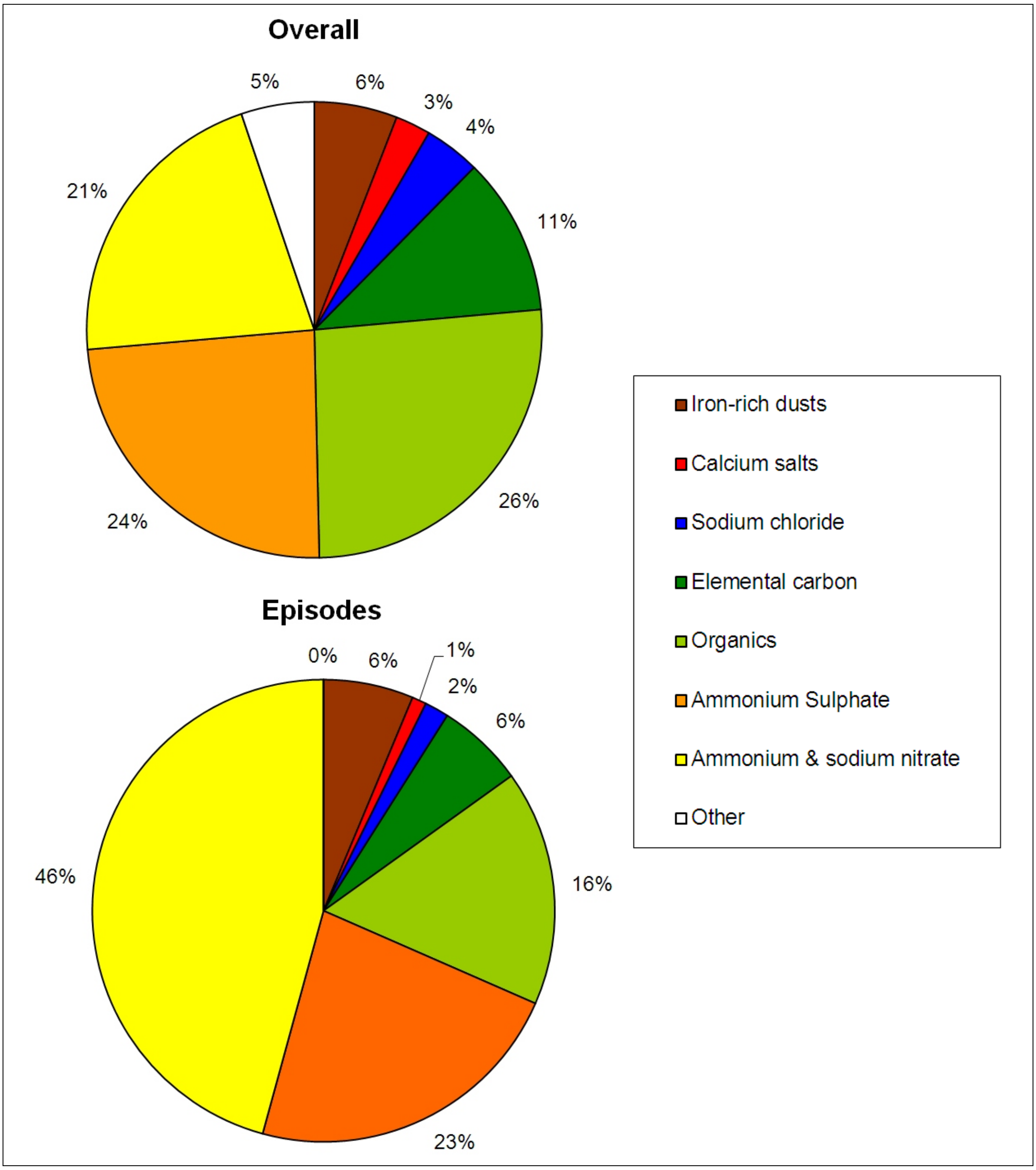

Figure 9 Composition of $PM_{2.5}$ on all days (overall) and episode days (PM_{10} > 50 µg/m^3) at a central urban background site in Birmingham. Source: Yin and Harrison (2008).

2.10 How is Secondary $PM_{2.5}$ Formed?

Secondary particles are formed as the result of atmospheric reactions involving both inorganic and organic gaseous precursors. For example, particles can be produced by the oxidation of precursor gases such as SO_2 and NO_x to acids followed by neutralisation with NH_3, and by the partial oxidation of organic compounds (USEPA, 2009).

The process by which secondary particles are formed is termed nucleation; this term refers to the processes by which molecules of low volatility condense to form solid or liquid matter. There are two distinct types of nucleation process. The first is heterogeneous nucleation. Most secondary particle formation in the atmosphere occurs by this process, whereby newly formed substances condense onto existing particles causing the growth of those particles. The condensation processes are most effective on small and medium sized particles, causing these to grow larger. The second process is called homogeneous nucleation. Some newly formed molecules have extremely low vapour pressure and, in the absence of an abundance of pre-existing particles, will condense with one another to form wholly new particles (AQEG, 2005).

2.10.1 How are nitrate, sulphate and ammonium particles formed?

The best known process of homogeneous nucleation occurs when sulphuric acid (H_2SO_4) is formed from the atmospheric oxidation of SO_2. This H_2SO_4 can nucleate with water vapour (binary nucleation) or, more readily, with water vapour and ammonia (ternary nucleation) to form droplets of sulphate solution. It is not wholly clear whether particle formation results from homogeneous nucleation of the oxidation product itself, or whether condensation occurs on a primary nucleus formed from H_2SO_4 nucleation (AQEG, 2005). A number of mechanistic details still remain to be determined, including the interactions with other species (USEPA, 2009). The overall oxidation rate of SO_2 is around 1% per hour, thus sulphate particles are only formed slowly (AQEG, 2005). SO_2 emission sources thus typically contribute to sulphate formed hundreds to thousands of kilometres downwind.

Nitrate aerosol is formed from the oxidation of NO_2, which itself is mainly derived from the oxidation of NO released during fossil fuel combustion. The predominant daytime oxidation route for NO_2 is initiated by its reaction with the hydroxyl radical (OH). This leads to the formation of gaseous nitric acid (HNO_3), which is highly soluble and also shows a strong affinity for surfaces. HNO_3 is scavenged by existing particles and droplets to form nitrate aerosol. Nitrate aerosol is also generated during night-time via the formation of the higher oxide N_2O_5, and the subsequent reaction of N_2O_5 with water or sea salt aerosol (AQEG, 2005). The overall oxidation rate of NO_2 is around 5% per hour, thus nitrate particles are formed more rapidly than sulphate particles (AQEG, 2005). NO_x emission sources typically contribute to nitrate formed tens to hundreds of kilometres downwind.

The precursor to ammonium aerosol formation is NH_3. NH_3 emissions are dominated by agricultural sources, which are mainly due to the decomposition of urea and uric acid in livestock waste. NH_3 is the most abundant alkaline gas in the atmosphere, and therefore plays a significant role in neutralising acids. It is efficiently taken up into acidic sulphate and nitrate aerosols, formed by the processes described above, leading to the formation of ammonium aerosol (AQEG, 2005).

2.10.2 How is secondary organic aerosol (SOA) formed?

SOA is formed when the oxidation of atmospheric non-methane volatile organic compounds (NMVOCs) generates oxygenated products that undergo gas-to-particle transfer. The precursor NMVOCs may be emitted from both anthropogenic and biogenic sources. Although hundreds of individual NMVOCs are emitted, the gas-phase oxidation of each compound broadly follows the same pattern. An initial oxidation step leads to the production of a set of products containing polar oxygenated functional groups - such as aldehyde, ketone, alcohol, nitrate and carboxylic acid groups - which tend to make the products less volatile and more water soluble. The gas-phase oxidation ultimately converts all the carbon in the emitted NMVOCs into CO_2. This is in contrast to the situation for SO_2 and NO_x, for which the ultimate oxidation products (sulphate and nitrate) are the forms that are most associated with the particle phase. SOA is therefore formed from the gas-to-particle transfer of partially oxidised organic material, which occurs in competition with further oxidation in the gas phase (AQEG, 2005).

As a result of the complexity of the emitted NMVOC mixture and the oxidation chemistry, the atmosphere contains many thousands of different organic oxygenates possessing a wide range of properties and, therefore, different propensities to undergo gas-to-particle transfer. However, certain classes of NMVOC are more likely to lead to aerosol formation by virtue of their high reactivity and the types of oxidation product formed. Of particular significance are large, cyclic, unsaturated compounds: the oxidation products tend to be of comparatively high molecular weight and contain two or more polar functional groups, these products are, therefore, of lower volatility. Two such classes of emitted NMVOCs, which generate SOAs, are monoterpenes and aromatic hydrocarbons (AQEG, 2005). A review of SOA studies by USEPA (2009) noted that oligomers[9] are likely to be a major component of OC in aerosol samples, and that small but significant quantities of SOA are formed from the oxidation of isoprene.

USEPA (2009) concluded that ambient samples can contain mixtures of SOA from different sources at different stages of processing, some with common reaction products making source identification of SOA problematic. The USEPA therefore noted the importance of accurately describing the phase distribution of semi-volatile organic compounds emitted by combustion sources under atmospheric conditions, and of atmospheric photochemical reactions in modifying the composition of emissions.

2.11 What Happens to Particles Once in the Atmosphere?

Particles smaller than 10 nm within the nucleation mode rapidly increase in size by condensational growth to move into the 0.01-0.05 µm range. These very small particles therefore have a relatively transient existence in the atmosphere (seconds or minutes) since they readily transform into larger particles and also deposit quite efficiently to surfaces because of their highly diffusive nature, which results from their low individual mass and high tendency to Brownian motion (AQEG, 2005).

Accumulation mode particles are too large to be subject to rapid Brownian motion and too small to settle from the air rapidly under gravity. Their further growth is inhibited because they do not coagulate as rapidly as fine and coarse particles, and there are diffusion barriers to their growth by condensation. Particles in the accumulation mode can therefore have a long atmospheric lifetime (typically 7–30 days), although they are subject to removal by incorporation into rain and this can significantly shorten their atmospheric lifetime. Gravitational settling velocities become appreciable within the coarse particle size range and therefore atmospheric lifetimes are much shorter than for the accumulation mode particles. Particles larger than ~100 µm in diameter rapidly settle out of the air and are of minor health significance because, although they can be inhaled, they do not generally penetrate beyond the nose and mouth (AQEG, 2005).

The chemical processes involved in the formation of secondary particles are relatively slow and because of their small size their persistence in the atmosphere is prolonged. Thus, while road traffic may be the main source of the original NO_x, and coal and oil burning the main source of SO_x, secondary particles are distributed more evenly throughout the air with fewer differences between urban and rural areas. They can also travel large distances, resulting in the transport of particles across national boundaries (AQEG, 2005).

---

[9] An oligomer consists of a few monomer units, in contrast to a polymer which can consists of an unlimited number of monomers.

2.12 How is $PM_{2.5}$ Removed from the Atmosphere?

Wet deposition and dry deposition are important processes for removing $PM_{2.5}$ from the atmosphere on urban, regional, and global scales. Wet deposition includes particles incorporated into cloud droplets that then fall as rain (rainout) and particles removed by collision with falling rain (washout). Snow and ice can also serve the same purpose. Dry deposition involves transfer of particles through gravitational settling and/or by impaction on surfaces by turbulent motions (USEPA, 2009). Table 6 summarises the atmospheric lifetime, travel distance and removal processes for particles in the three main size modes. $PM_{2.5}$ particles include the nucleation and accumulation fractions, together with some of the coarse fraction. They will have a long lifetime in the atmosphere, typically days to weeks, and a typical travel distance of several hundred kilometres. Their main removal mechanism will be rainout and dry deposition.

Table 6 Fate of airborne particles of different sizes (adapted from USEPA (2009)).

	Nucleation (<0.05 μm)	**Accumulation** (0.05-1 μm)	**Coarse** (1-100 μm)
Atmospheric half-life	Minutes to hours	Days to weeks	Minutes to hours
Removal processes	Grows into accumulation mode	Forms cloud droplets and rains out	Dry deposition by fallout
	Diffuses to raindrops and other surfaces	Dry deposition	Scavenging by raindrops
Travel distance	<1 to 10s of km	100s to 1000s of km	<1 to 10s to km (100s to 1000s of km in dust storms for small size tail)

3. WHAT ARE THE SOURCES OF $PM_{2.5}$?

3.1 What are the Sources of Primary $PM_{2.5}$?

The main anthropogenic and natural sources for the major constituents of primary $PM_{2.5}$ are summarised in Table 7. Only major sources for each constituent within each broad category are shown, and not all sources are equal in magnitude. Anthropogenic sources can be further divided into stationary and mobile sources (Table 8).

Table 7 Constituents of primary $PM_{2.5}$ and main sources (adapted from USEPA (2009)).

Constituent	Natural sources	Anthropogenic sources
Sulphate (SO_4^{2-})	Sea spray	Fossil fuel combustion
Nitrate (NO_3^-)	-	Mobile source exhaust
Minerals	Erosion and re-entrainment	Fugitive dust from paved/unpaved roads; agriculture; forestry; construction and demolition
Ammonium (NH_4^+)	-	Mobile source exhaust and agriculture
Organic carbon (OC)	Fires	Prescribed burning, wood burning, mobile source exhaust, cooking, tyre wear and industrial processes
Elemental carbon (EC)	Fires	Mobile source exhaust (mainly diesel); wood biomass burning; cooking
Metals	Volcanic activity	Fossil fuel combustion; smelting and other metallurgical processes; brake wear
Biological material	Viruses and bacteria	-

- minor source or no known source of component.

Table 8 Stationary and mobile sources of $PM_{2.5}$ (adapted from USEPA (2009)).

Stationary sources	Mobile sources
Fuel combustion for electrical utilities, residential space heating and cooking	Exhaust emissions from road vehicles
Industrial processes	Fugitive dust from paved and unpaved roads
Construction and demolition	Diesel trains
Metal, mineral, and petrochemical processing	Shipping
Wood products processing	Aircraft
Mills and elevators used in agriculture	Non-road mobile machinery (tractors, generators, construction plant, *etc.*)
Erosion from tilled lands	
Waste disposal and recycling	
Biomass burning[a]	

[a] Biomass combustion encompasses many emission activities including burning of wood for fuel, burning of vegetation to clear land for agriculture and construction, to dispose of agricultural and domestic waste, to control the growth of animal or plant pests, and to manage forest resources (prescribed burning). Wildlands also burn due to lightning strikes and arson, the former being natural, the latter anthropogenic.

3.2 What are the Anthropogenic Combustion Sources of $PM_{2.5}$?

Results of receptor modelling calculations indicate that $PM_{2.5}$ is produced mainly by combustion of fossil fuel, either by stationary sources or by transport. A relatively small number of broadly defined source categories, compared to the total number of chemical species that are typically measured in ambient monitoring source receptor studies, account for the majority of the observed PM mass (USEPA, 2009).

3.2.1 What contribution is made by road-vehicle exhaust to $PM_{2.5}$?

Road transport was responsible for around a quarter of primary $PM_{2.5}$ emissions in the UK in 2008 (NAEI, 2010), with diesel vehicles accounting for a much higher proportion than petrol. Greater concern is often expressed about road transport emissions than other sources, probably on account of the proximity of road vehicles to people and residential or commercial properly.

Combustion-derived particles generally comprise carbonaceous material, onto which a wide range of organic and inorganic compounds may be adsorbed. Exhaust particles are generally in the fine fraction, with an aerodynamic diameter of less than 1.0 µm. Given that there is no physical process occurring in an engine that could produce primary particles as large as 2.5 µm, any coarse particles measured in tests probably result from the sampling system walls and not primary engine exhaust.

The general processes of particle formation during diesel combustion have been described in detail by Heywood (1988). Particle formation begins with the creation of carbonaceous material (soot) in the cylinder. Condensation reactions of gas-phase species such as unsaturated hydrocarbons and polycyclic aromatic hydrocarbons (PAHs) lead to the appearance of the first recognisable solid material (soot spherules) at combustion temperatures of between 1000K and 2800K. A phase of particle growth then follows inside the cylinder that includes surface growth of spherules by the adsorption of gas-phase components, as well as by their coagulation and agglomeration. At temperatures above 500°C the particles are principally clusters of spherules, with individual spherule diameters of 15–30 nm (0.015-0.03 µm). Once these clusters have left the cylinder they are then subject to a further mass addition process as the exhaust gases cool; as temperatures fall below 500°C the particles become coated with adsorbed and condensed high-molecular weight organic compounds. Almost all of the particles found in the exhaust pipe before dilution are present as these carbonaceous agglomerates (accumulation mode particles), with a small amount of metallic ash and adsorbed materials.

Most nucleation mode particles (up to 90%) are thought to originate from the condensation of lower volatility organics in the exhaust gas during exhaust dilution, rather than during combustion. However, nucleation is a non-linear process and the number of particles formed is very sensitive to the nature of the sampling conditions. If a nucleation mode is observed during measurement – and this is not always the case – then the number of particles it contains will greatly exceed the number in the accumulation mode, although the nucleation mode particles contribute little to the total mass.

The latest emission standards for cars and heavy-duty vehicles require large reductions in PM emissions, and these will be achieved through the use of particle filters. Such filters lead to almost complete removal of solid particles, with a reduction in mass of more than three orders of magnitude. However, for a Euro 4 diesel car equipped with a filter, Tzamkiozis *et al.* (2010) have shown that nucleation mode particles can still be emitted under high-speed driving conditions, and have recommended that this be investigated further.

3.2.2 What contribution is made by rail transport to $PM_{2.5}$?

In the UK passenger trains are powered by either diesel or electricity, whereas freight trains are mostly powered by diesel. The nature of the PM emitted from rail transport is dependent upon the type of traction. In the case of diesel locomotives particles are emitted directly into the local environment as result of both combustion (engine exhaust) and abrasion (wheels, track, brakes and catenary system). In the case of electric locomotives the abrasion processes also result in local emissions, but the exhaust emission is effectively transposed to the power stations which are responsible for producing the electricity.

Emissions from diesel locomotives are estimated from fuel consumption estimates using fuel-specific emission factors. Although total UK emissions from railways contribute in a minor way to the total (0.8% of primary $PM_{2.5}$ emissions in 2008), it is likely that they could be more important locally, for example, close to a rail terminus with a large proportion of non-electric trains (AQEG, 2005).

Compared with cars, trains have negligible PM emissions per passenger-km. PM emissions from railway traffic have therefore only been addressed in a small number of studies, most of which have related to either in-train exposure or to measurements in subway systems. However, particle measurements were conducted near a busy railway line in Zurich by Bukowiecki *et al.* (2007). The concentrations of several metals were investigated. It was found that the main elements abraded by railway traffic were manganese and iron originating from steel abrasion, as well as copper abraded from the overhead traction line. More than half of the iron and manganese particles emitted by railway traffic through wheel and track abrasion were found in the coarse mode range, and particles were generally larger for cargo trains than for passenger trains. However, the average mass concentration of all abrasion particles was roughly 1–2 $\mu g/m^3$, which only equated to a minor contribution to total ambient PM_{10} (total = 31 $\mu g/m^3$).

3.2.3 What contribution is made by Shipping to $PM_{2.5}$?

The UK National Atmospheric Emissions Inventory (NAEI) provides emission estimates for coastal shipping, naval shipping and international marine. Detailed information on shipping emission factors is provided by Entec UK Ltd and the EMEP/EEA Air Pollutant Emission Inventory Guidebook. Information on shipping activity at UK ports is provided by Lloyds Marine Intelligence Unit (LMIU).

Shipping within coastal waters (out to 12 miles) is responsible for a significant fraction (10.1% in 2008) of primary $PM_{2.5}$ emissions in the UK[10]. Many ships use old engines or old engine technology, and few exhaust after-treatment devices are used to control particle emissions. The PM emission factor for ship engines (primary and secondary particles) is typically around 1.25 g/kWh (Fridell *et al.*, 2008). This can be compared with the regulated value for a modern truck with Euro 5 standards, which is 0.02 g/kWh. Moreover, shipping also contributes considerably to secondary particle formation, as NO_x and SO_2 emissions from ships are rather high.

The fuel quality used for most ships is poor relative to that used for road vehicles. In particular, the fuel oil used in shipping contains very high levels of sulphur, which also affects PM emissions. Ash content of the fuel is also an important parameter affecting PM emissions (Denier van der Gon and Hulskotte, 2009).

There is little information on the size distribution of PM emissions from ships. Denier van der Gon and Hulskotte (2009) reviewed the methodologies for estimating emissions from shipping in the Netherlands, and concluded that the general assumption that $PM_{2.5}$

---

[10] Emissions from fuels used by vessels of all flags that depart and arrive in the same country

represents 95% of PM_{10} is probably too high. For example, Fridell *et al.* (2008) give values of 66% and 75% for heavy fuel oil and marine diesel oil respectively.

3.2.4 What contribution is made by air transport to $PM_{2.5}$?

Air transport makes a small contribution to national emissions of primary $PM_{2.5}$, accounting for 0.09% in 2008 (NAEI, 2010). These emissions are calculated over the landing/take-off (LTO) cycle, which includes emissions up to 1000 m. A substantial fraction of PM arises from tyre wear during landing; around 22% for PM_{10} at Stansted Airport (BAA, 2008). Most studies have focused on PM_{10}, and there is greater uncertainty as to $PM_{2.5}$ emissions.

While national emissions are very small, the localised nature of the emission means that impacts may be greater in the vicinity of an airport. Heathrow is the largest airport in the UK, and considerable monitoring has been carried out around the airport. One monitoring site is located on the northeast boundary of the airfield, 180 m from the centre of the northern runway, and downwind of the prevailing wind. It has been estimated that PM_{10} concentrations are enhanced at this location by about 0.9 $\mu g/m^3$ due to airport activities (DfT, 2006). The contribution to $PM_{2.5}$ will be smaller, as it will be a fraction of the PM_{10}.

3.2.5 What contribution is made by stationary sources to $PM_{2.5}$?

Stationary combustion sources of $PM_{2.5}$ are dominated by emissions from residential plant, which contributed 13% of primary $PM_{2.5}$ emissions in the UK in 2008. The next most important sector is emissions from power generation. Public utilities contributed 6% of primary $PM_{2.5}$ emissions for in 2008, with manufacturing industries and construction contributing 3.7% (NAEI, 2010). The uneven distribution of these sources means that the local impacts can be much more significant than implied by the small contributions they make to national emissions.

3.2.6 How much does biomass burning contribute to $PM_{2.5}$?

Particle emissions can arise from the burning of biomass in several ways:

- combustion of wood in domestic wood-burning stoves;
- emissions from garden bonfires;
- emissions from accidental fires including forest fires and house fires;
- combustion of biomass-based fuels in large scale boilers for space heating or in industrial facilities and power stations.

The 2007 NAEI gives the following data for PM_{10} emissions from the combustion of wood (NAEI, 2010)

- Domestic combustion 8.5 kilotonnes (10% of total UK emissions of $PM_{2.5}$);
- Other industrial combustion 0.11 kilotonnes (0.1% of total UK emissions of $PM_{2.5}$);
- Power stations 0.66 kilotonnes (0.7% of total UK emissions of $PM_{2.5}$).

These numbers are no doubt open to considerable uncertainty, particularly those associated with domestic combustion. The NAEI does not give data for $PM_{2.5}$ from biomass but inferences can be drawn from the report of the Air Quality Expert Group on *Particulate Matter in the United Kingdom* (AQEG, 2005). That report gives a PM_{10} emission from domestic combustion of wood in 2001 of 7.1 kilotonnes, hence indicating an increase in emissions from this source between 2001 and 2007. The AQEG report also gives estimates of emissions of other size fractions, indicating 70.4% of PM_{10} emissions from domestic wood combustion to be in the $PM_{2.5}$ size range. This leads to an estimate of $PM_{2.5}$ from domestic combustion of wood of 4.9 kilotonnes per year which is slightly larger than emission from coal combustion (4.7 kilotonnes per year) or natural gas combustion (4.1 kilotonnes per year). It is, however, much smaller than the emission of $PM_{2.5}$ from road transport, which amounted to 19 kilotonnes per year in 2008 (NAEI, 2010). Nonetheless, it remains a significant emission, but in the absence of data on its geographic distribution, it is difficult to comment on its likely significance for air quality. The AQEG report indicates uncertainties in the estimation of PM_{10} emissions from domestic wood consumption to be of the order of -50% to +200% in the emission factor and ± 30% in the activity data. Given additional uncertainties in the percentage of $PM_{2.5}$ in PM_{10} the overall uncertainties in $PM_{2.5}$ emissions amount to a factor of around 3. A recent review (Bolling *et al*., 2009) comments on the highly variable combustion conditions in currently available combustion appliances for wood and the fact that these result in large variations in the physio-chemical characteristics of the emitted particles. This dependence on combustion conditions will also influence the emission factors.

Because of the large uncertainties in emission factors, considerable work has been conducted in both North America and mainland Europe to use atmospheric measurements to quantify airborne concentrations of biomass smoke. There are three main methods, all of which require calibration before wood smoke concentrations can be estimated:

- Many workers have used the concentration of levoglucosan, a carbohydrate derived in large abundance from wood burning, to estimate the impact of biomass combustion (*e.g.* Puxbaum *et al.,* 2007; Caseiro *et al.,* 2009).
- Fine particle potassium is a tracer for biomass smoke (Caseiro *et al*., 2009; Puxbaum *et al*., 2007). The potassium concentration needs to be corrected for contributions from both sea salt and soil, adding to the uncertainty.
- Use of a multi-wavelength aethalometer, a device which collects particles on a paper tape and measures their absorption in the visible and ultra-violet wavelengths. This method allows a differentiation of wood smoke components from black carbon (Sandradewi *et al*., 2008; Favez *et al*., 2009).

It is also possible to get indirect but useful information from analysis of carbon-14 in airborne particles (representative of contemporary carbon), and from analysis of the data from aerosol mass spectrometry.

The weakness of the levoglucosan and potassium methods is that the ratio of wood smoke mass to levoglucosan or potassium is not a fixed quantity and varies with combustion conditions. The aethalometer method depends upon calibration against one of the other procedures, as it estimates wood smoke only indirectly and is also sensitive to the chemical nature of the wood smoke. Consequently, these procedures used alone are open to significant uncertainty, although this uncertainty is less than that associated with emissions inventories.

Rather few studies report concentrations of biomass smoke as such. Puxbaum *et al.* (2007) report biomass smoke concentrations in the $PM_{2.5}$ size range (2-hour averages), ranging from 0.05 $\mu g/m^3$ in the Azores to 4.3 $\mu g/m^3$ at Aveiro on the west coast of Portugal. The highest monthly concentrations of up to 14 $\mu g/m^3$ were observed in winter at the Aveiro site. Measurements in Paris (Favez *et al*., 2009) suggested that carbonaceous aerosols originating from residential wood burning represented about 20% of the $PM_{2.5}$ concentration (around 5 $\mu g/m^3$).

The only published data for the United Kingdom (Yin *et al*., 2010) have shown rather lower concentrations. At an urban background site in Birmingham, biomass smoke was estimated to contribute only 0.07 $\mu g/m^3$ out of a total measured $PM_{2.5}$ of 11.6 $\mu g/m^3$. At a rural site, the annual average biomass smoke contribution was estimated at 0.06 $\mu g/m^3$ from a total measured $PM_{2.5}$ of 10.5 $\mu g/m^3$. These data were the aggregate of one sample per month, and in the light of more recent unpublished data from the same group, appear likely to be a significant under-estimate of the wood smoke concentration.

There is an increasing tendency to consider biomass fuels for space heating of both dwellings and commercial/industrial premises, and two reviews have been conducted of the potential impact on air quality from increased biomass use in London (Abbott *et al.*, 2007) and in Scotland (Abbott *et al.,* 2008). The former report examined five scenarios for the use of biomass boilers or biomass combined heat and power plant in London and estimated that in the most extreme scenario PM_{10} concentrations at background locations in London could increase from below 24 µg/m^3 currently to almost 37 µg/m^3, hence leading to an exceedence of the daily mean air quality objective for PM_{10} (the objective is expected to be exceeded when the annual mean exceeds 31.5 µg/m^3). In the case of $PM_{2.5}$, predicted concentrations were set to rise to almost 30 µg/m^3, compared with 17 µg/m^3 for a 'business as usual' scenario. In the second study (Abbott *et al.,* 2008), scenarios based upon local development plans were formulated for Dundee and Edinburgh and indicated a potential increment to PM_{10} of < 0.1 µg/m^3 except in the immediate vicinity of the proposed installations. In a model for 2020, using a higher emission factor, contributions to PM_{10} of 0.5-1 µg/m^3 were predicted across large parts of both cities. The disparity between this and the London study is, no doubt, largely a consequence of the very different scenarios selected.

3.3 What are the Anthropogenic Non-Combustion Sources of $PM_{2.5}$?

3.3.1 What contribution does road-vehicle tyre wear make to $PM_{2.5}$?

Several studies conducted in the 1970s revealed that airborne tyre wear particles tend to be divided into two distinct size groups: ultrafine particles with an aerodynamic diameter of less than 1 µm, and coarse particles larger than around 7 µm (Cardina, 1974; Dannis, 1974; Pierson and Brachaczek, 1974; Cadle and Williams, 1978). These observations were confirmed by Fauser (1999). Plausible mechanisms for the distinction are the volatilisation (the thermal degradation of tyre polymer and the volatilisation of extender oils) and subsequent condensation of material in the ultrafine particle mode, and normal mechanical wear for larger sizes (Cadle and Williams, 1978).

The relative mass contributions of the fine and coarse modes appear to be rather variable, and dependent on the sampling conditions and the metric used. Pierson and Brachaczek (1974) showed that only 10% by mass of all tyre wear particles were smaller than 3 µm. Cadle and Williams (1978) also observed that the larger particles dominate the total mass, but only during dynamic sampling (*i.e.* with a continuous flow of air through the test chamber). Under low air-flow conditions many of the large particles settled before reaching the sampling point, with the result that sub-micron particles dominated the mass distribution. More recent receptor modelling work includes that by Rauterberg-Wulff (1998; 1999), which indicated that tyre wear PM_{10} was only present in the coarse fraction (2.5-10 µm), and that by Moosmüller *et al.* (1998), which indicates that the products of tyre wear are dominated by particles larger than 10µm; these studies appear to confirm the earlier findings. However, Fauser (1999) found that around 90% by mass of tyre wear particles smaller than 20 µm had an aerodynamic diameter of less than 1 µm. Other PM_{10} data from the USEPA (2006) and Berdowski *et al.* (1997) indicate that around 70% by mass of tyre wear PM_{10} can be classified as $PM_{2.5}$, 10% as PM_1, and 8% as $PM_{0.1}$. Dannis (1974) found that mean particle diameter decreases with increasing speed, and this may be one of a number of factors contributing to the differences in the reported findings.

Dahl *et al.* (2006) reported a predominance of sub-micrometre particles generated by abrasion at the road surface/tyre interface in road simulator studies. The particle size distribution and source strength were observed to vary with tyre type. It was therefore suggested that the detected sub-micrometre particles originated from tyre wear as opposed to wear of the road surface material.

The overall picture of the distribution of tyre wear PM between the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ fraction is very unclear. The current emission factors used in the NAEI assume that 70% of the PM_{10} is in the $PM_{2.5}$ size fraction (Murrells, 2010).

3.3.2 What contribution does road-vehicle brake wear make to $PM_{2.5}$?

As with tyre wear, not all of the worn brake material will be emitted as airborne PM, although proportionally more in the case of brake wear, of which a significant proportion is as $PM_{2.5}$ (Wahlin *et al*., 2006; Iijima *et al*., 2007). Garg *et al.* (2000) found that, on average, around 35% of brake wear mass is released as airborne PM. However, this does not take into account sampling losses, and if these were to be included in the study by Garg *et al.* (2000) the airborne fraction would increase to around 64% (Sanders *et al*., 2003). Sanders *et al.* (2003) conducted detailed laboratory tests using state-of-the-art equipment, and observed that, depending on the severity of the braking, between 50% and 90% of the total wear material was in the form of airborne particles. The collection efficiency for brake wear debris was between 90% and 100% of the wear mass.

Whilst the majority of the fine particulate brake dust from disc brakes is released to the environment, small amounts of brake dust can be retained on the vehicle. According to Lohrer and Mierheim (1983), 10% of brake dust is retained in the drum brake enclosure. This value appears to be slightly low compared with the observations made by Sanders *et al.* (2003), whose test track and wind tunnel measurements revealed that typically 50% of the brake wear debris escapes the vehicle and enters the atmosphere. It was also found that 3-30% of brake debris falls on the road, 16-22% is retained on the wheel, and 8-25% is retained on the brake and steering/suspension equipment, but the exact proportions will vary from vehicle to vehicle depending on the design and operating conditions. When low-metallic brake linings were used, 60% of the wear debris was found to originate from the disc and 40% from the linings, a result which could have implications for the interpretation of the results of studies which have only considered the brake linings.

Under controlled laboratory conditions, Cha *et al.* (1983) found that the diameters of airborne particles and deposited dust were generally similar, with a peak in the particle size distribution at 2.1-3.3 μm and around 10% of particles in the sub-micron size range. The percentage of airborne particles was found to increase with vehicle speed. Data from the USEPA (1995) and Berdowski *et al.* (1997) indicated that 98% (by mass) of airborne brake wear particles can be classified as PM_{10}, whilst around 40% of the PM_{10} is $PM_{2.5}$, 10% is PM_1, and 8% is $PM_{0.1}$. Garg *et al.* (2000) recorded airborne brake wear particle mass fractions smaller than 10 μm, 2.5 μm, and 0.1 μm of 88%, 63% and 33% respectively. Sanders *et al.* (2003) give PM_{10} and PM_1 proportions of 80% and 2%. Different size distributions have been obtained elsewhere. Receptor modelling work by Abu-Allaban *et al.* (2003), using PM_{10} and $PM_{2.5}$ measurements, showed that the brake wear contribution was observed mainly in the PM_{10} fraction, and that the $PM_{2.5}$ share of PM_{10} was only 5-17%. The mass mean diameter of brake wear debris reported by Garg *et al.* (2000) varied between 0.7 and 2.5 μm. It is possible that the high temperatures generated during braking can vaporise some of the brake pad material, and Garg *et al.* (2000) suggested that the volatile material may condense during measurement and contribute to the fine particle fraction. For three different types of brake lining, Sanders *et al.* (2003) observed a consistent mass-weighted size mean diameter over an urban driving cycle of around 5-6 μm. Under harsh braking conditions, the mass mean diameter was closer to 10 μm, and it was considered possible than a significant proportion of the wear debris could have been larger than 20 μm in diameter.

The findings of Sanders *et al.* (2003) appear to be strongly supported by the work of Iijima *et al.* (2007). Dynamometer tests were conducted on three non-steel brake lining materials to generate abrasion dusts, across a range of temperatures simulating different driving conditions and braking severity. Number size distributions revealed a peak mode of 1–2 μm in all tests. Mass size distributions were calculated from the number size distributions, and indicated peak particle emissions in the range 3–6 μm. The authors estimated between 74% and 92% of brake wear particles, by number, to be emitted as $PM_{2.5}$, corresponding to 12–36% of particle mass. The highest proportions of $PM_{2.5}$ emissions were found in the lower temperature tests, considered to be representative of urban driving cycles. In addition, brake dust particles were found to consist of, amongst other materials, the elements Ba, Cu, K, Fe, Sb, and Zn. According to Thorpe and Harrison (2008), these findings highlight the great variability that can be anticipated in brake wear emissions under real-world conditions, and the strong dependency on vehicle operating conditions. Differences between the conclusions of Iijima *et al.* (2007) and the earlier work of Garg *et al.* (2000) may be explicable in terms of differences in experimental set-ups and lining materials tested (Thorpe and Harrison, 2008). Mosleh *et al.* (2004) characterised brake wear particles using a laser scattering method, and concluded that wear debris tends to show a bimodal particle number size distribution. A peak in the fine particle fraction around 350 nm was identified in all the brake tests performed, independent of brake pressure or lining material. A second mode was seen in the coarse particle fraction. The peak diameter of this mode was observed to vary with braking pressure. Consistent with the findings of Sanders *et al.* (2003), there was an increased tendency for coarser particles to be emitted as braking pressure increased.

The recent work by Gietl *et al.* (2010) also produced results which were consistent with those of Iijima *et al.* (2007). Size-segregated aerosol samples were collected at three sites in London, one roadside with heavy traffic (Marylebone Road) and two urban background sites (Regent's College and North Kensington). The samples were analysed for metals which were considered as potential candidates for tracers of non-exhaust vehicle emissions. The metals Fe, Ba, Cu and Sb - which have been associated with brake wear in the literature - were clearly increased at the roadside in the size range 3.7 to 7.2 µm. Furthermore, it was considered that the similarity between the size distributions of barium in roadside air and urban background air was strongly suggestive of brake dust being its predominant source. Barium was found to comprise 1.1% of brake wear (PM_{10}) particles from the traffic fleet as a whole, allowing its use as a quantitative tracer of brake wear emissions at other traffic-influenced sites

Based on emission factors for brake wear used in the EMEP/EEA Air Pollutant Emissions Inventory Guidebook (EEA, 2009), Thorpe and Harrison (2008) calculated that PM_{10} accounts for 98% of emitted brake wear particles, whilst $PM_{2.5}$ accounts for 39%. These figures are broadly comparable with the findings of Garg *et al.* (2000) and Sanders *et al.* (2003). However, the proportion of brake wear emissions in $PM_{0.1}$ is quoted as only 8%, somewhat smaller than the 33% stated by Garg *et al.* (2000).

As for tyre wear, the overall picture of the distribution of break wear PM between the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ fraction is very unclear. The current emission factors used in the NAEI assume that 40% of the PM_{10} is in the $PM_{2.5}$ size fraction (Murrells, 2010).

3.3.3 What contribution does road surface wear make to $PM_{2.5}$?

Very little information on the size distribution of road surface wear particles can be found in the literature. In the only study identified for this review, Fauser (1999) found that airborne bitumen particles mainly ranged in size between 0.35µm and 2.8µm, with a mean of around 1 µm.

3.3.4 What contribution does resuspension of road surface dust make to $PM_{2.5}$?

Several studies in the United States have indicated that the resuspension of paved road dust contributes significantly to atmospheric PM_{10} and $PM_{2.5}$ concentrations (Chow *et al.,* 1995; Schauer *et al*., 1996; Kleeman and Cass, 1999). Kleeman and Cass (1999) found that the entrainment of road dust from paved and unpaved roads was responsible for 34% of all PM_{10} and 20% of all $PM_{2.5}$ in Los Angeles. The fact that the same study showed that exhaust emissions from road vehicles were responsible for 5% of all PM_{10} and 14% of all $PM_{2.5}$ illustrates the potential importance of resuspension processes. It was also found that particles emitted from paved road dust sources, and crustal material other than paved road dust, dominated the 24-hr size distribution for particles larger that 1µm. Similarly, Gaffney *et al.* (1995) and Zimmer *et al.* (1992) estimated that the contribution of emissions from paved roads to total PM_{10} might be as high as 30% in California and 40% to 70% in the Denver Metropolitan area respectively. An inventory of fine particulate organic carbon emissions to the Los Angeles area atmosphere compiled by Hildemann *et al.* (1991) indicated that paved road dust was the second largest source of fine aerosol organic carbon particles to the urban atmosphere. Brake linings and tyre wear were found to be the eleventh and thirteenth largest sources respectively.

A study in France showed that resuspension may be three to seven times higher than exhaust emissions from road transport (Jaecker-Voirol and Pelt, 2000). At a range of sites in Germany, PM_{10} emissions due to resuspension have also been found to be up to several times larger in magnitude than those from vehicle exhaust, but with a large range of variation between locations (Pregger and Friedrich, 2002). Investigations in Berlin by Rauterberg-Wulff (1998) showed that more than 50% of traffic-derived PM was in the coarse mode, with this mode consisting of 22% carbonaceous material (elemental and organic carbon), and 78% minerals. This indicated that most of the coarse mode particles are due to resuspension of road dust and tyre and brake wear. Even the fine PM mode contained a considerable proportion of mineral matter.

Resuspension is also a major concern in Nordic countries, especially in relation to the use of studded tyres and road sanding. Johansson (2002) derived emission factors for non-exhaust emissions for $PM_{2.5}$ and PM_{10} which were respectively equal to, and 9 times larger than, the exhaust emission factors. The resuspension of road dust is the most important local source in urban areas of Sweden, contributing 17 to 22 $\mu g/m^3$ to annual mean PM_{10} concentrations at kerbside sites, representing around half the total PM_{10} (Areskoug *et al*., 2004). At sites where traffic is the major source, the coarse particles mass is about 6 times larger than the fine particle mass, although $PM_{2.5}$ can contain a considerable fraction of resuspended particles (Areskoug *et al*., 2004).

In the UK, Harrison *et al.,* (2001) concluded that vehicle-induced resuspension of PM_{10} has a source strength approximately equal to that of exhaust emissions for a location alongside the Marylebone Road in London. This was revised during subsequent work to an estimate of 28% from resuspension at this location (Charron and Harrison, 2005). However, UK emission inventories and models cannot adequately include the resuspension of road dust as a particle source due to the absence of appropriate emission factors. Thorpe *et al.* (2007) derived emission factors for resuspension of road dust at a site in Marylebone Road. The starting assumption was that all the resuspended PM is in the $PM_{2.5\text{-}10}$ fraction, with all the vehicle exhaust in the $PM_{2.5}$ fraction. This is unlikely to be strictly true, and a component of the resuspended PM is likely to be present as $PM_{2.5}$. Indeed, the USEPA (2006) $PM_{2.5}$ emission factor for paved roads is 14% of that for PM_{10}, *i.e.* 14% of the resuspended PM_{10} is in the $PM_{2.5}$ fraction.

Using the work of Charron and Harrison (2005), which showed around 28% of roadside PM_{10} was due to vehicle-induced resuspension, and the USEPA (2006) assumption of 14% of resuspended PM_{10} being in the $PM_{2.5}$ fraction, then the resuspended $PM_{2.5}$ fraction will be around 4% of the $PM_{2.5}$ in exhaust emissions (assuming exhaust emissions are all in the $PM_{2.5}$ fraction).

3.3.5 What contribution is made by stationary sources to non-combustion $PM_{2.5}$?

A wide range of industrial operations can contribute to $PM_{2.5}$ emissions. These include emissions from point sources at industrial installations, such as foundries, through to fugitive releases from materials handling, such as in steel works. These emissions are not well characterised in terms of PM size. The routine monitoring of emissions is generally of total PM, and usually not PM_{10}, let alone $PM_{2.5}$. $PM_{2.5}$ emissions are therefore inferred from the measurements, using results from specific size fractionation studies. Another issue related to these emissions is that measurements of particles within a stack omit those particles that may subsequently be formed in the gas stream, termed 'condensable particles' (Whiting, 2004). The emission inventory for the UK shows individual industrial sectors generally make a small contribution, *e.g.* aluminium production accounted for 0.13% of UK emissions in 2008, while industrial coating application accounted for 1.9% and iron and steel 3.7%. In total though, these industrial sources accounted for 17% of primary non-combustion PM$_{2.5}$ emissions (NAEI, 2010)

3.4 How much Primary PM$_{2.5}$ is Emitted from Different Sources in the UK

Annual emissions of a wide range of air pollutants in the UK are presented in the NAEI. Emissions of primary PM$_{2.5}$ in 2008 are shown in Figure 10. The main sources are also summarised in Table 9. The largest source sector is industry, accounting for 28.9% of the 2008 total primary anthropogenic PM$_{2.5}$ emission. Next is road transport, accounting for 23.5% (including brake and tyre wear). The residential sector and shipping are also important sources (NAEI, 2010). The break down of primary PM$_{2.5}$ emissions from the industrial sector, including power stations is shown in Figure 11.

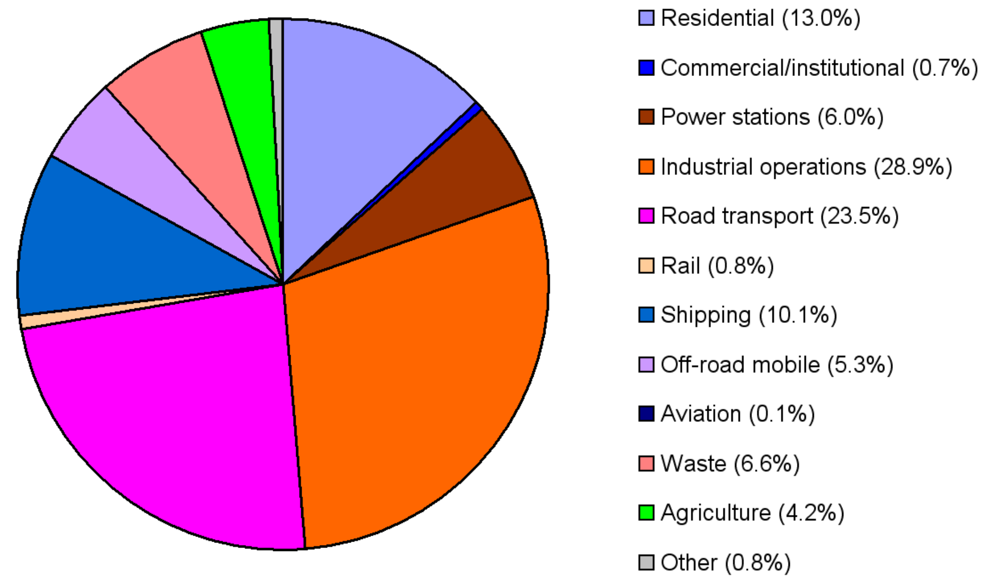

Figure 10 Proportional emissions of primary PM$_{2.5}$ in the UK in 2008. Source: NAEI (2010).

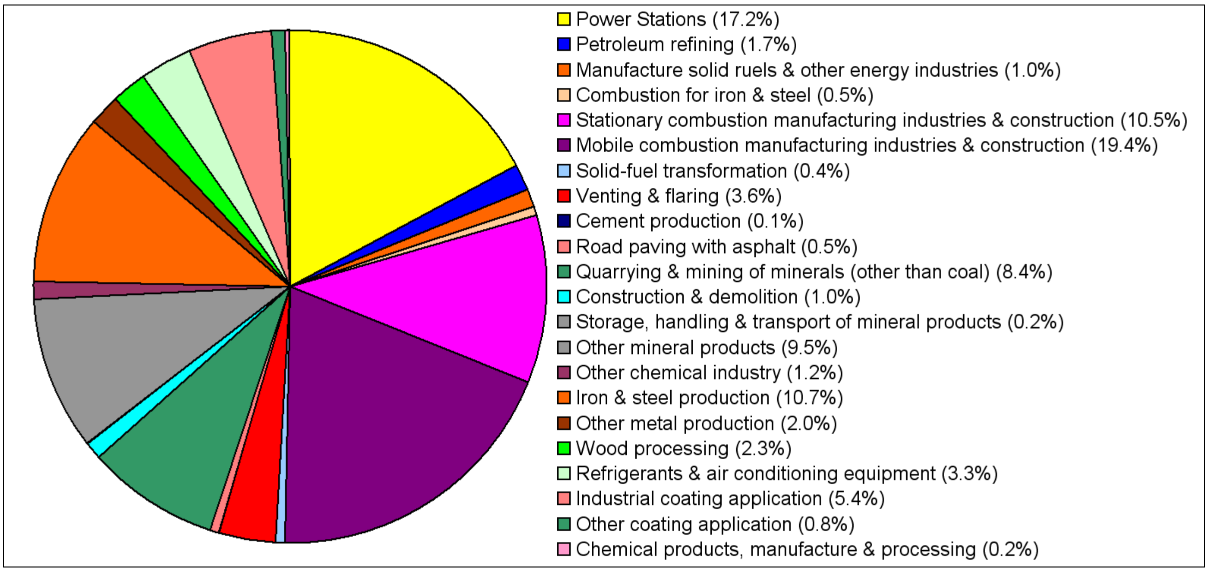

Figure 11 Proportional emissions of primary PM$_{2.5}$ from power stations and the industrial sectors in the UK in 2008. Source: NAEI (2010).

Table 9 UK $PM_{2.5}$ emissions by sector in 2008. Source: NAEI (2010).

Sector	$PM_{2.5}$ (ktonnes)	Proportion of total
Industry	23.5	28.9%
Road transport	19.1	23.5%
Residential	10.6	13.0%
Shipping	8.2	10.1%
Waste	5.3	6.6%
Power stations	4.9	6.0%
Off-road mobile	4.3	5.3%
Agriculture	3.4	4.2%
Rail	0.6	0.8%
Other	0.6	0.8%
Commercial/institutional	0.5	0.7%
Aviation	0.1	0.1%
Total	81.1	100%

The time series of UK $PM_{2.5}$ emissions between 1970 and 2008 is shown in Figure 12. Between 1970 and 2008, emissions of $PM_{2.5}$ decreased by 66%, mainly due to the reduction in coal use. Emissions from the domestic, commercial and institutional sectors have fallen by almost half from 21 ktonnes in 1990 to 11 ktonnes in 2008. Road traffic emissions (including exhaust and brake and tyre wear) have decreased from 34 ktonnes in 1990 to 19 ktonnes in 2008 (NAEI, 2010).

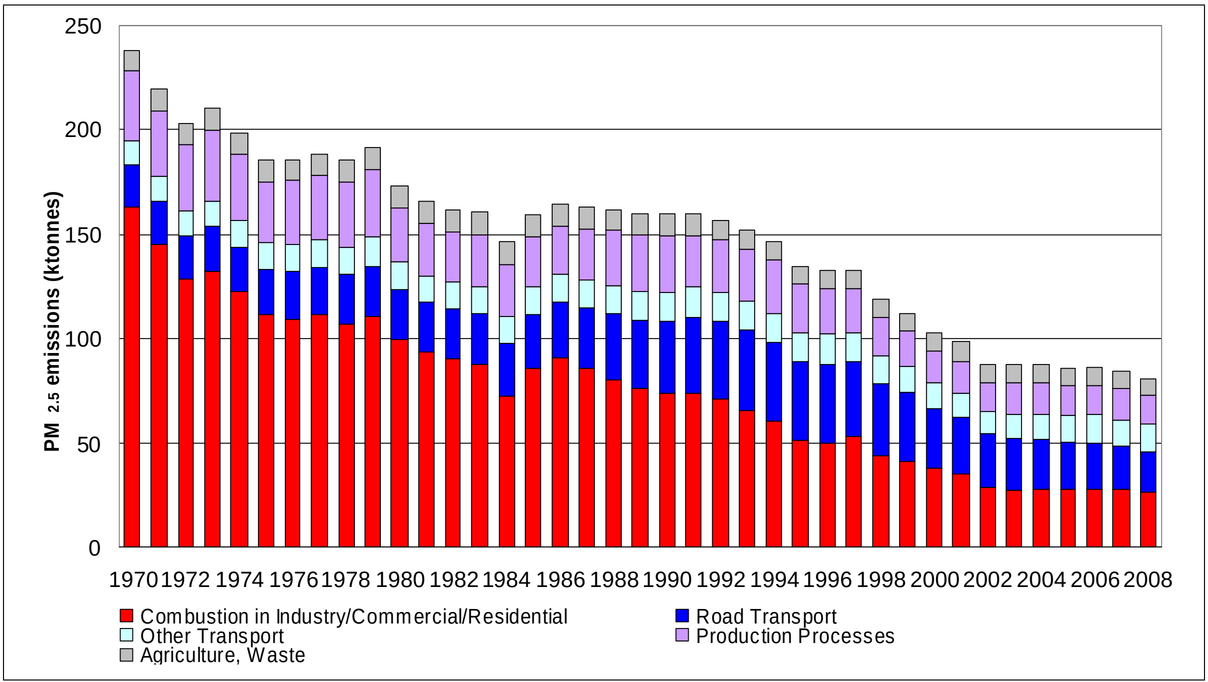

Figure 12 Time series of UK $PM_{2.5}$ emissions. Source: NAEI (2010).

3.5 What are the Natural Sources of Primary $PM_{2.5}$?

Natural sources of primary $PM_{2.5}$ include windblown dust from undisturbed land, sea spray, biological material, and natural forest/grass fires (USEPA, 2009). They can make significant contributions to urban background $PM_{2.5}$. The contribution of windblown dust may be some 5-8% (see Table 5 and Figure 30). Sea salt may contribute some 3-15% to $PM_{2.5}$, depending on proximity to marine sources (see Figure 25 and Figure 26). Primary emissions of biological material are expected to make a minor contribution, while a more significant, but still small, contribution will arise from the formation of secondary biogenic aerosols from emissions of natural organic precursor gases such as terpenes. The cycling of natural biogenic material and PM formation is still poorly understood and not yet well quantified. Natural fires make a small contribution overall, accounting for 0.23 ktonnes per annum in the national inventory (NAEI, 2010), while other natural sources contribute 6.4 ktonnes per annum (NAEI, 2010) (which is around 8% of primary anthropogenic emissions.

3.6 What are the Anthropogenic Precursor Sources of Secondary $PM_{2.5}$ and How are They Changing over Time?

The key precursors of secondary $PM_{2.5}$ are sulphur dioxide, which gives rise to sulphates, nitrogen oxides, which give rise to nitrates, ammonia, which gives rise to ammonium, and NMVOCs, which give rise to organic aerosols. A brief overview is provided of the main sources in the UK and the trends in emissions in both the UK and the EU27 countries. The EU27 countries are considered as they can make a significant contribution, given the regional nature of secondary PM. This Section only deals with anthropogenic sources, and does not include natural sources of these precursors.

Trends in emissions of SO_2, NO_x and NH_3, which relate to the formation of ammonium sulphate and ammonium nitrate particles, and of NMVOCs, which relate to the formation of organic particles, are shown for the UK in Figure 13 to Figure 16. These Figures include projected emissions from the UK for 2010, 2015 and 2020. Trends in the EU27 countries for these same precursor emissions are shown in Figure 17 to Figure 20. The energy and industrial sectors are the dominant source of SO_2, and have shown a substantial decline in the UK over the last 35 years, which looks set to continue (Figure 13). It is notable that the decline in total SO_2 emissions has been faster in the UK than in the EU27 countries since 1990 (Figure 17). A wider range of sources make significant contributions to NO_x emissions (Figure 14), and the decline in emissions in the UK has been slightly faster than in the EU27 as a whole (Figure 18). Emissions from the farming industry, and cattle in particular, dominate NH_3 emissions, and there has only been a slow decline in both the UK and EU27 as a whole (Figure 15 and Figure 19). Emissions of NMVOCs are distributed across a range of sources (Figure 16). The sizeable decline in the UK between 1990 and 2008 has been driven largely by the reduction in emissions from the transport sector. The UK decline has been faster than that in the EU27 as a whole (Figure 16 and Figure 20).

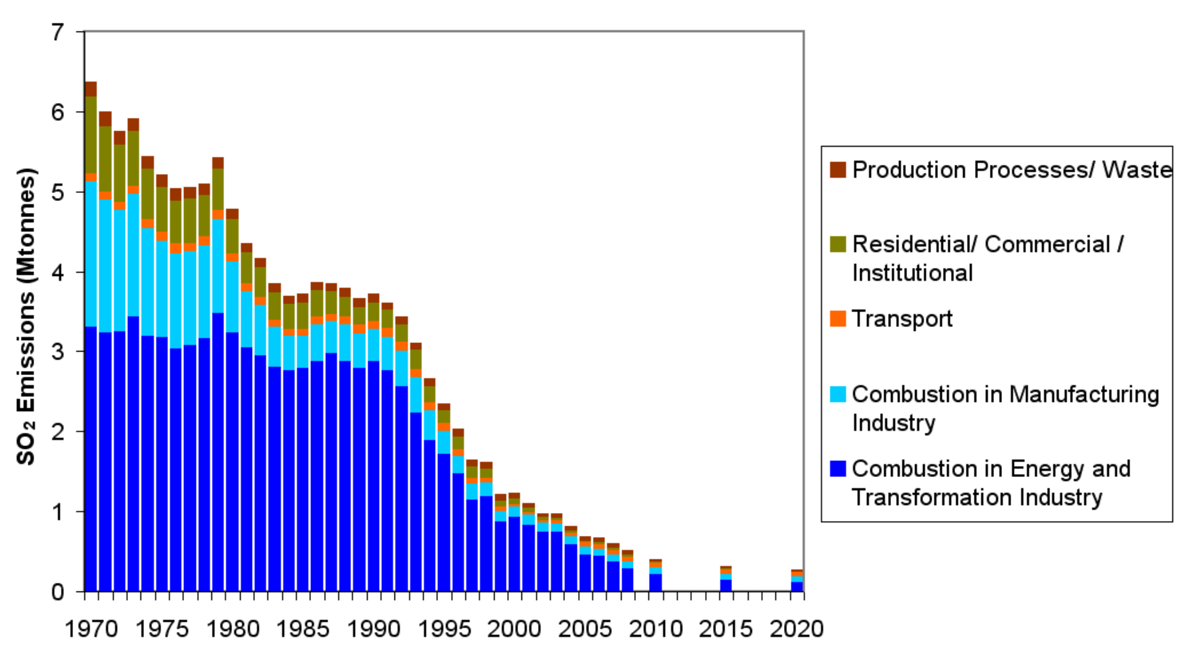

Figure 13 Emissions of SO_2 from UK sources 1970-2020. Source: NAEI (2010).

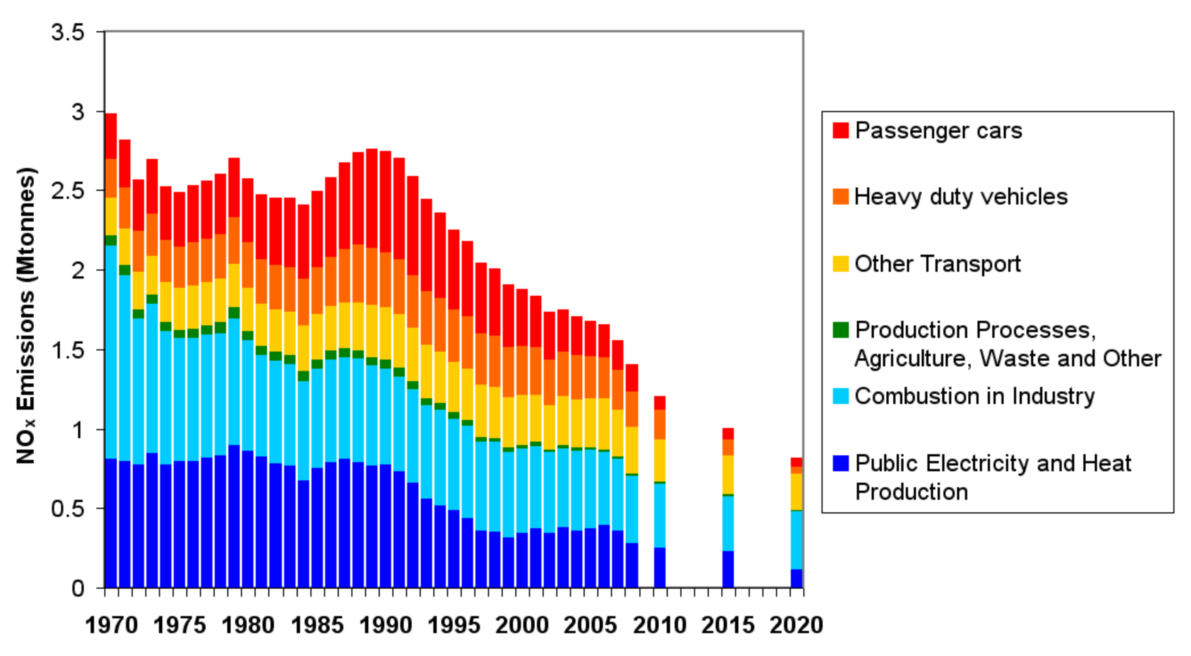

Figure 14 Emissions of NO_x from UK sources 1970-2020. Source: NAEI (2010).

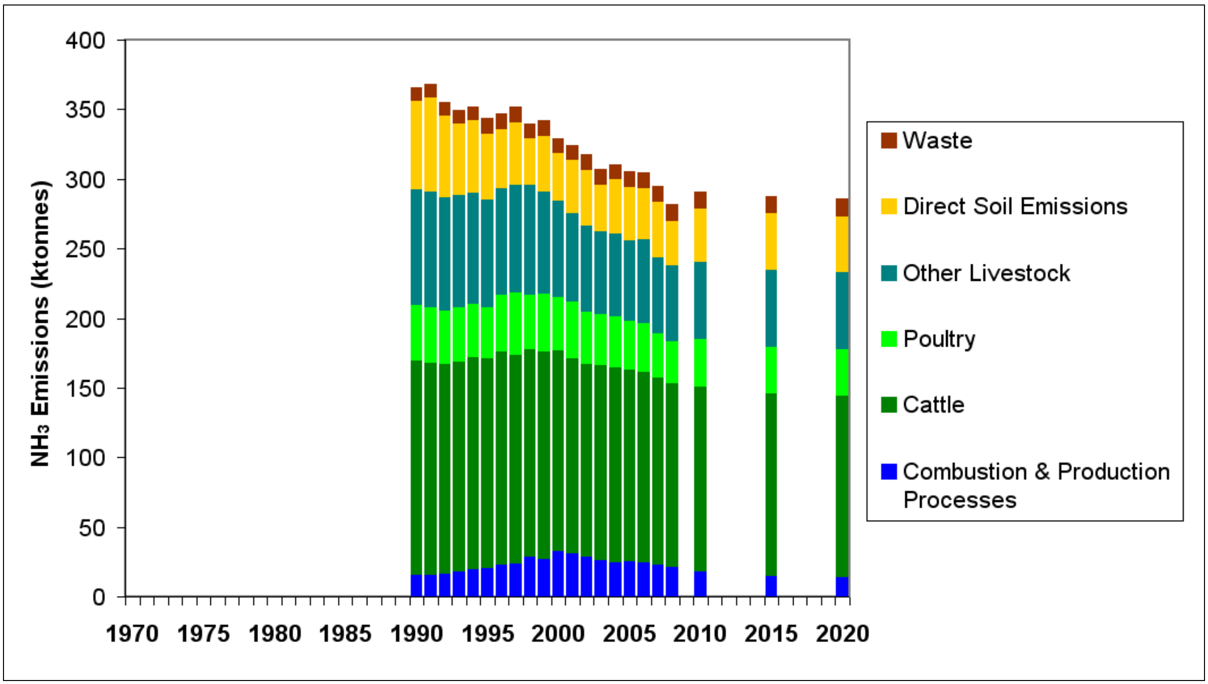

Figure 15 Emissions of NH_3 from UK sources 1990-2020. Source: NAEI (2010).

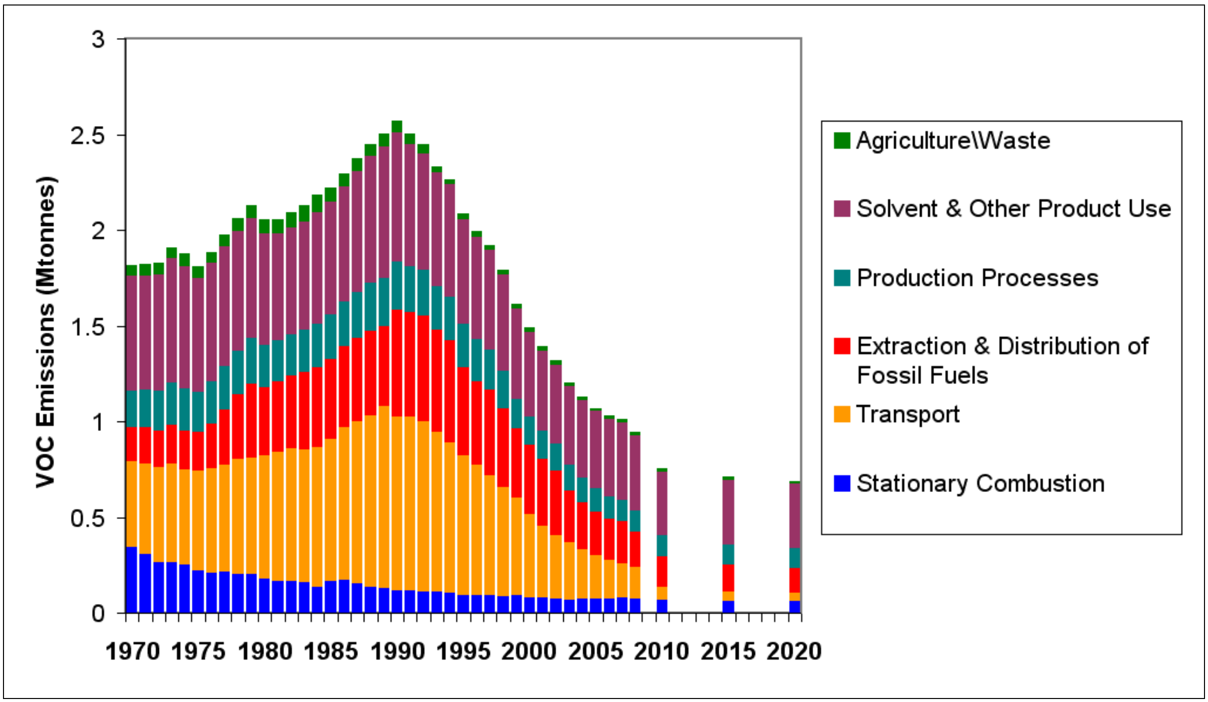

Figure 16 Emissions of NMVOCs from UK sources 1970-2020. Source: NAEI (2010).

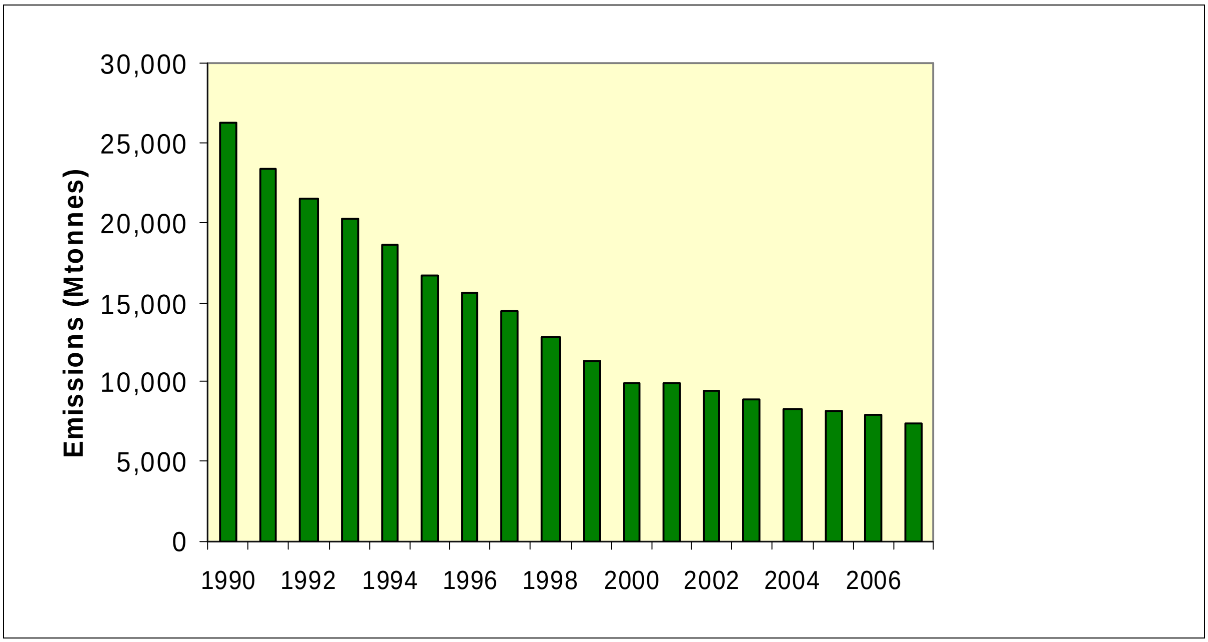

Figure 17 Emissions of SO_2 from EU27 countries 1990-2007. Source: European Environment Agency website http://dataservice.eea.europa.eu.

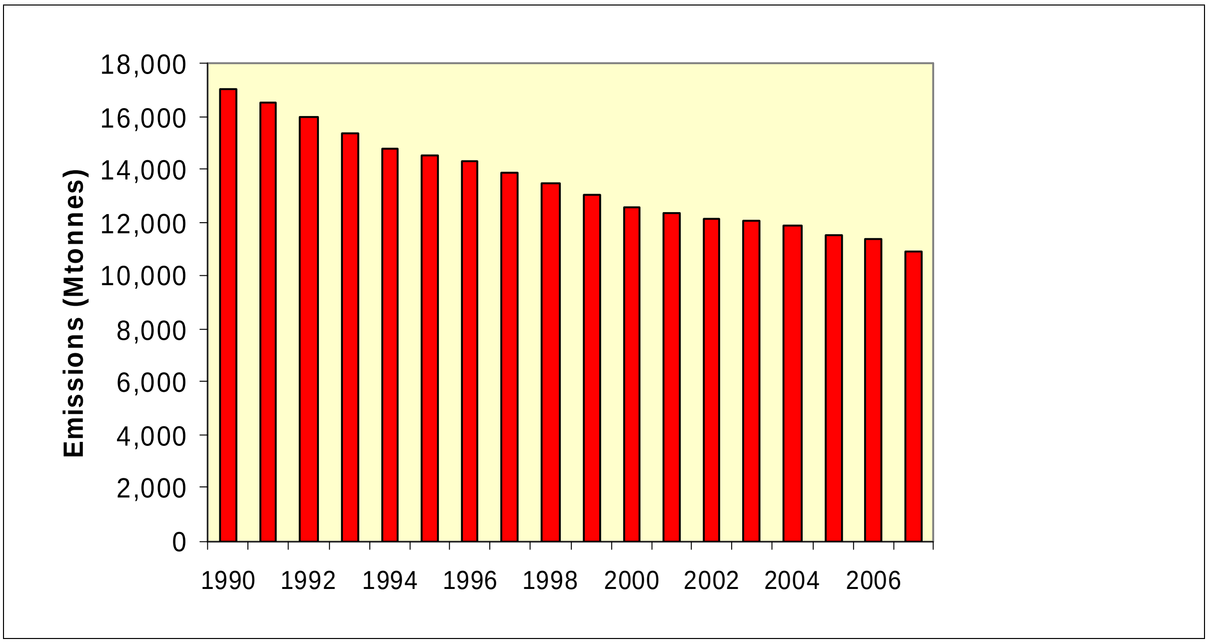

Figure 18 Emissions of NO_x from EU27 countries 1990-2007. Source: European Environment Agency website http://dataservice.eea.europa.eu.

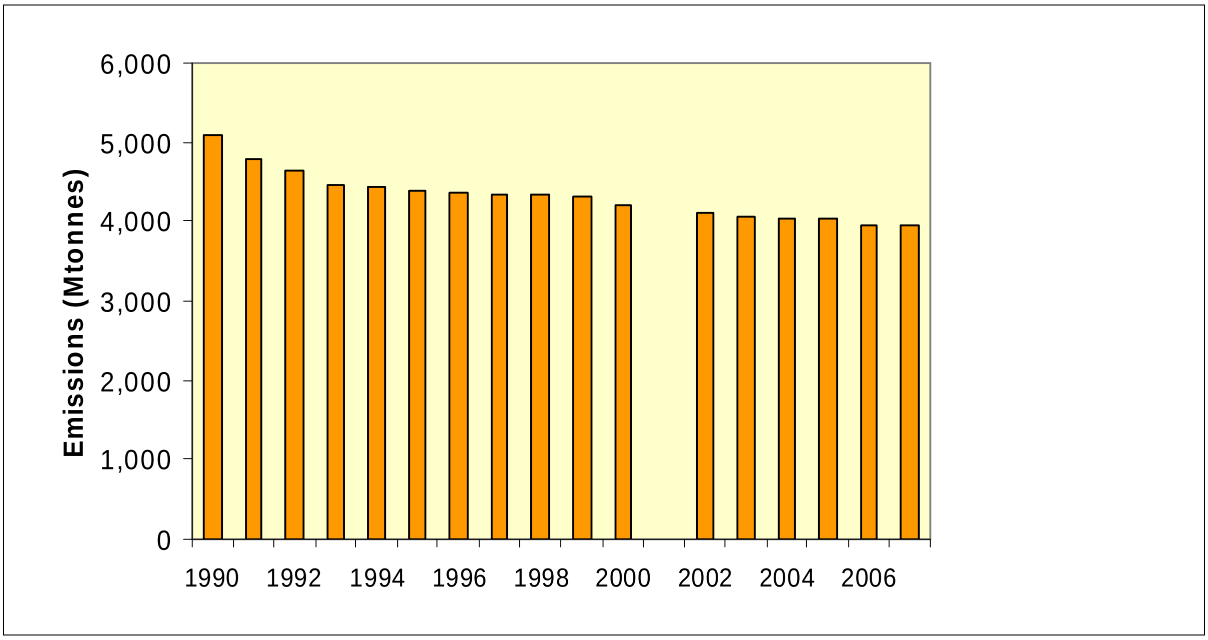

Figure 19 Emissions of NH_3 from EU27 countries 1990-2007. Source: European Environment Agency website http://dataservice.eea.europa.eu.

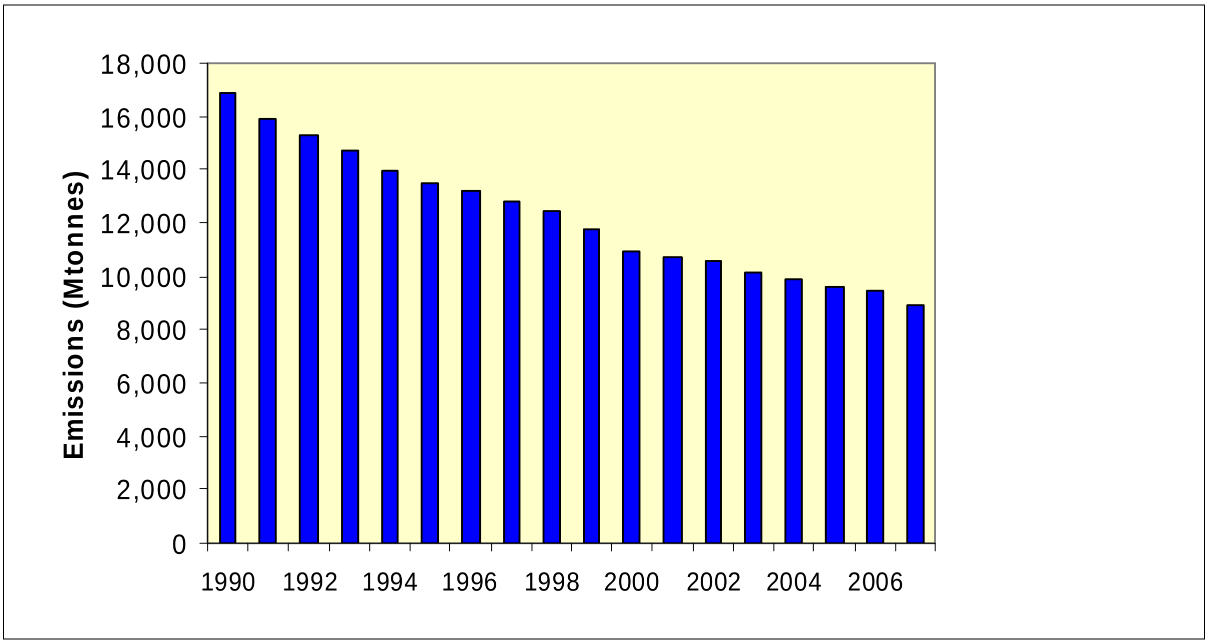

Figure 20 Emissions of NMVOCs from EU27 countries 1990-2007. Source: European Environment Agency website http://dataservice.eea.europa.eu.

3.7 What are the Contributions of Fossil and Non-Fossil Sources to the Carbon Component of $PM_{2.5}$?

Determination of the amount of carbon-14 radioisotope present in the carbon of PM can be used to apportion between fossil-fuel sources of carbon, which are wholly anthropogenic, and modern sources of carbon which comprise both biomass/biofuel combustion and SOA derived from biogenic emissions of VOC (isoprene, pinene, *etc*.). Results indicate that a substantial proportion of PM total carbon is of contemporary origin: ~50–60% for urban background PM_{10} in Zürich (Szidat *et al.*, 2004, 2006), and ~50% for urban background PM_{10} and $PM_{2.5}$ in Göteborg (Szidat *et al.*, 2009). Unpublished data for samples of urban background $PM_{2.5}$ from Birmingham are entirely in line, showing mean contemporary carbon content of 50%, range 27-66% (*n* = 26) (Heal *et al.,* 2010). For samples of $PM_{2.5}$ collected at five remote and rural sites in the European CARBOSOL project, fraction contemporary values were higher still, in the range ~57-82% (Gelencser *et al.*, 2007), presumably reflecting greater contribution of biogenic carbon at these non-urban locations.

More sophisticated approaches determine carbon-14 in the OC and EC fractions of total carbon separately, although this is subject to the same ambiguities in defining OC-EC split as for more conventional analytical methods. Results of this top-down source apportionment of total carbon into five broad source categories for PM in Zürich, Göteborg and Birmingham, averaged over a number of samples, are shown in Figure 21.

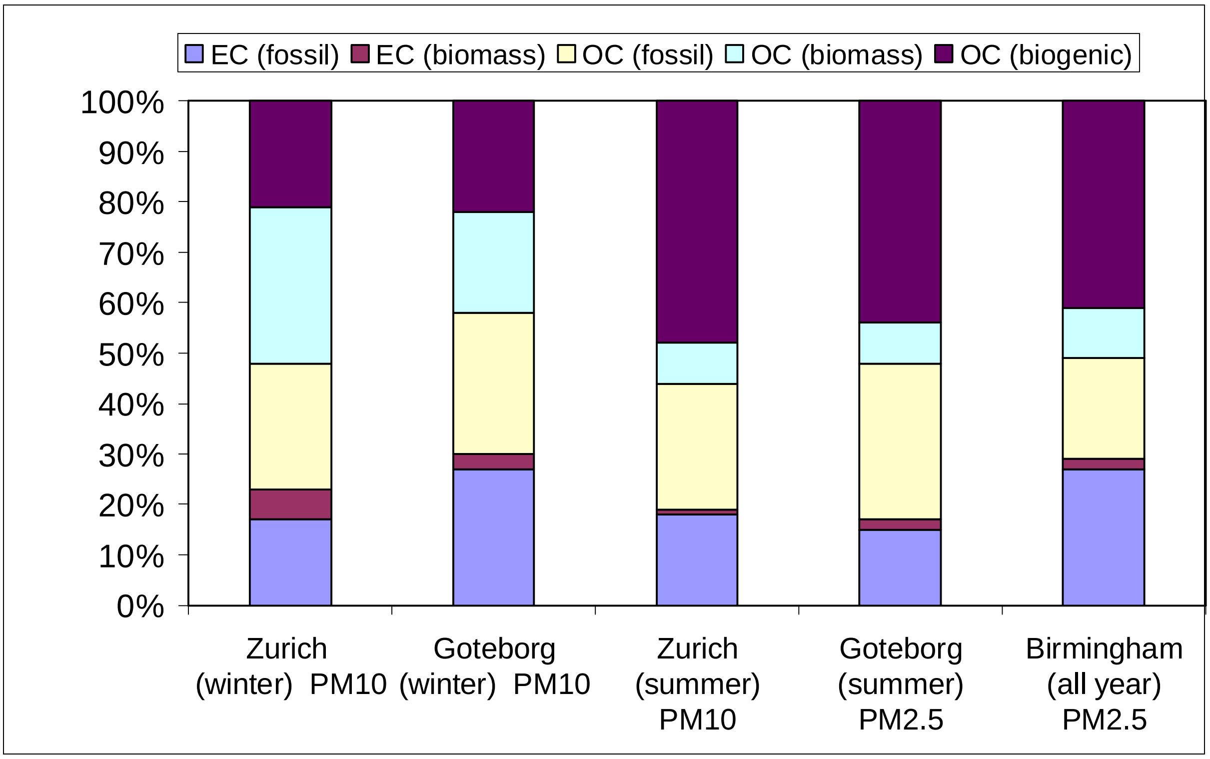

Figure 21 Estimated source apportionment, based on carbon-14 measurements, of the organic and elemental carbon in samples of urban background PM in Europe. Sources of data are given in the text.

The biomass categories (OC & EC) are assumed to derive predominantly from solid-fuel biomass burning, but could include liquid biofuels, and are likely entirely of anthropogenic origin (rather than from natural wild fires). The OC fossil category will comprise a mixture of primary OC directly from combustion sources and secondary organic aerosol derived from anthropogenic emissions of VOC of fossil origin such as toluene and other aromatic compounds. The OC biogenic category is assumed to indicate predominantly secondary organic aerosol derived from emissions of VOC but may also include additional primary material of biological origin such as pollen, spores and small fragments of plant material (carbohydrates, *etc.*) or other sources of contemporary organic carbon PM, such as food cooking or cigarette smoke, not accounted for in the OC biomass category. Nevertheless, even with these uncertainties it is clear there is a significant and ubiquitous presence of biogenic SOC even in urban background $PM_{2.5}$. The apportionments are broadly similar, although the two continental cities are strongly influenced by solid biomass burning in winter, not reflected in samples from Birmingham which do not show seasonality. Samples from Birmingham showed a positive trend for the fraction contemporary carbon and SOC/TC ratio to be higher for air-mass back trajectories arriving in Birmingham from over land, consistent with terrestrial sources contributing to secondary OC.

4. EXPOSURE TO $PM_{2.5}$ IN THE UK

4.1 What are the Background $PM_{2.5}$ Levels to which People are Exposed?

Maps of estimated background $PM_{2.5}$ concentrations across the UK on a 1 x 1 km grid have been produced by Defra and the Devolved Administrations (Defra, 2010a). The methodology for deriving these maps is similar to that used for the PM_{10} maps, as described by Grice *et al.* (2010). They are based on a semi-empirical model, using concentrations measured in 2008, and using projections for all years up to 2020. They provide the average concentration across a grid square derived from all sources, primary and secondary, natural and anthropogenic. Figure 22 shows the estimated background $PM_{2.5}$ concentrations for 2010. Rural concentrations are highest in south-east England (approximately 10-12 µg/m^3) declining in a north-westerly direction to below 6 µg/m^3 in Scotland and Northern Ireland. Slightly higher concentrations occur within the major urban areas, *e.g.* London, Birmingham, Cardiff, Belfast, Edinburgh, Glasgow, *etc.* There is also evidence of higher concentrations associated with the major motorways.

This information on background concentrations has been combined with the population within each grid square to estimate a population-weighted exposure concentration (Stedman, 2010). Clearly this will be higher than the average across all UK grid squares, as the population is biased towards urban areas, where $PM_{2.5}$ concentrations are higher. The values for 2010, 2015 and 2020 are set out in Table 10. The population-weighted mean concentrations in 2010 range from 5.5 µg/m^3 in Scotland to 14.1 µg/m^3 in inner London. There is a projected decline over the decade of around 5-7% assuming a business as usual scenario, *i.e.* one that takes account of reductions due to policies and legislation already in place, including the tightening Euro standards for road vehicles (Stedman, 2010).

These results can be compared with the data presented by De Leeuw and Horálek (2009) for exposure across Europe in 2005. Their analysis involved mapping of background PM_{10} concentrations and application of $PM_{2.5}$:PM_{10} ratios to derive a $PM_{2.5}$ concentration map. Population-weighted and area-weighted concentrations are set out in Table 11 for selected countries. The area-weighted values are generally lower than the population-weighted values, which reflects the fact that more people live in urban areas where concentrations are higher. The result for the UK of 10.2 µg/m^3 in 2005 is very similar to that derived independently by Stedman (2010) for 2010. It is also clear that the levels in the UK are almost the lowest in Europe.

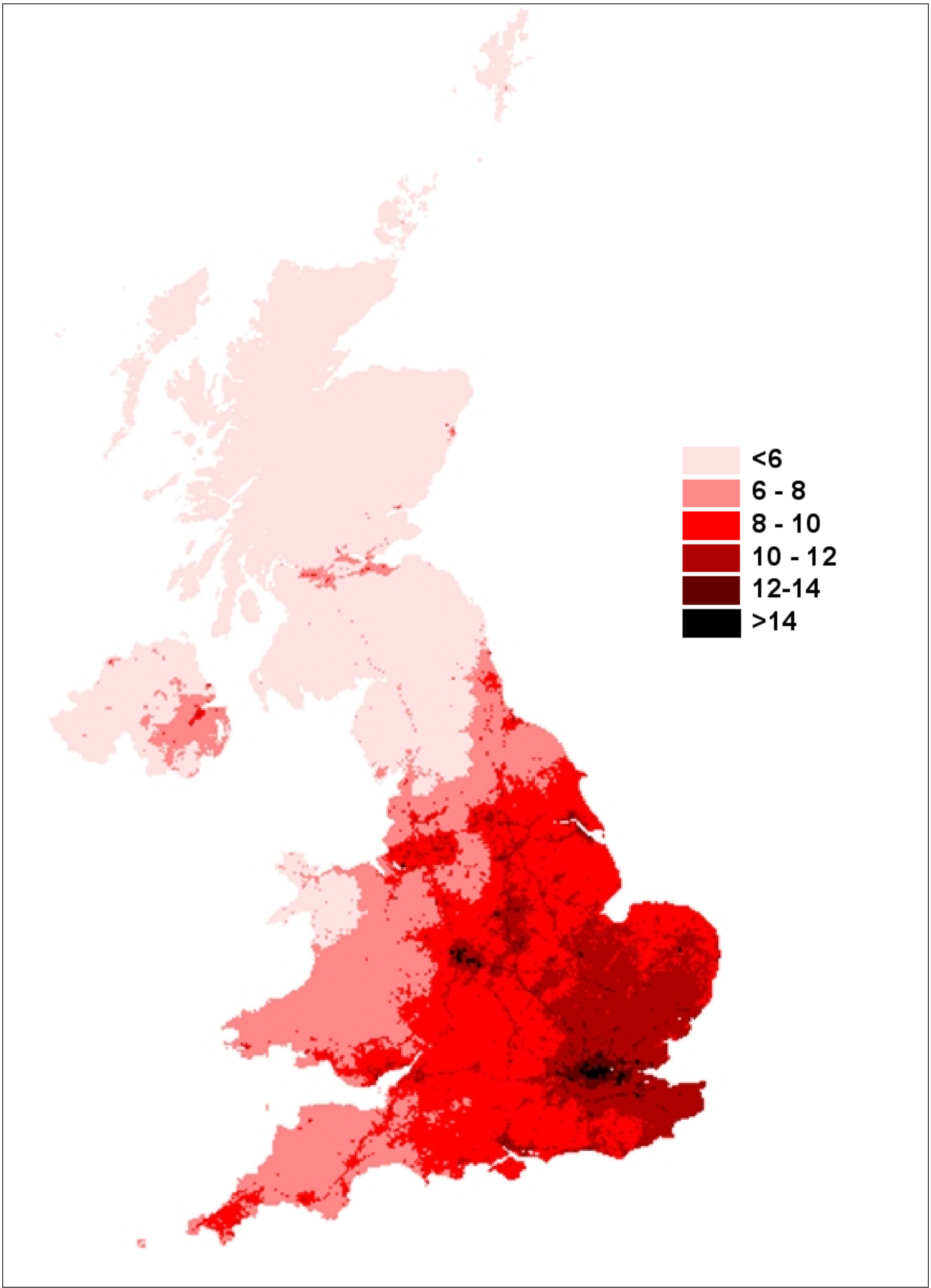

Figure 22 Estimated annual mean background $PM_{2.5}$ concentrations (µg/m^3) in 2010. Source: Drawn from data provided by Defra (2010a)

Table 10 Population-weighted mean[a] PM$_{2.5}$ concentrations (µg/m^3) in 2010, 2015 and 2020. Source: Stedman (2010).

	2010	2015	2020	Percentage Reduction (2010-2020)
Scotland	5.5	5.3	5.1	7.0
Wales	8.3	8.0	7.8	5.0
Northern Ireland	6.4	6.1	6.0	5.6
Inner London	14.1	13.4	13.1	7.3
Outer London	13.4	12.9	12.7	5.7
Rest of England	10.6	10.2	10.0	5.4
UK	10.3	9.9	9.7	5.6

[a] The population-weighted mean concentration cannot be directly compared with the Average Exposure Indicator in the EU Directive, as this is based on an average PM$_{2.5}$ concentration, measured over three years at urban background sites in major conurbations.

Table 11 Population- and area-weighted mean background PM$_{2.5}$ concentrations (µg/m^3) in 2005. Source: De Leeuw and Horálek (2009).

Country	Population-Weighted	Area-Weighted
United Kingdom	10.2	7.0
Ireland	7.6	5.9
Sweden	11.0	8.2
Germany	15.3	14.2
France	13.2	12.4
Austria	18.4	13.6
Czech Republic	23.5	20.4
Spain	16.1	10.2
Italy	23.8	18.0

4.1.1 How many people are exposed to levels above thresholds in background air?

The background maps have been used to define the population exposed to PM$_{2.5}$ concentrations in excess of:

- 25 µg/m^3, the EU limit value from 2015
- 20 µg/m^3, the EU exposure concentration obligation from 2015
- 12 µg/m^3, the annual mean objective in Scotland from 2020

Stedman (2010) has provided tables setting out the populations that exceed these thresholds for 2010, 2015 and 2020 in different parts of the UK. These are provided in Appendix 1 and the findings are summarised below.

The results show:

- No one is exposed to $PM_{2.5}$ concentrations in background air above the 25 µg/m^3 EU limit value, in any year between 2010 and 2020.
- No one is exposed to $PM_{2.5}$ concentrations in background air above the 20 µg/m^3 EU exposure concentration obligation, in any year between 2010 and 2020.
- No one in Scotland is exposed to $PM_{2.5}$ concentrations in background air above the 12 µg/m^3 Scottish objective, in any year between 2010 and 2020

In other words, there are no areas where background $PM_{2.5}$ concentrations are predicted to exceed any of the thresholds that have been established by the EU or UK governments.

It is important to note that these results do not include the higher exposure that can arise alongside busy roads. Stedman (2010) has developed a national model that calculates $PM_{2.5}$ concentrations alongside the national trunk road network. This has shown that there are 16.9 km of road in London in 2010, where the 20 µg/m^3 exposure concentration obligation would be exceeded within 10 m of the edge of the road; however, this is predicted to drop to zero in 2015, when the exposure concentration obligation applies[11].

4.1.2 How much do the different sources contribute to exposure to background $PM_{2.5}$?

The background maps described above have been disaggregated into the different components that contribute to background $PM_{2.5}$. Figure 23 shows the distribution of secondary $PM_{2.5}$ (organic and inorganic) across the UK. Concentrations decline from over 6 µg/m^3 in the southeast of England, to less than 2 µg/m^3 in the northwest of Scotland. This demonstrates the strong transboundary contribution to secondary $PM_{2.5}$ from mainland northern Europe.

---

[11] The exposure concentration obligation is based on the Average Exposure Indicator (AEI) and therefore does not strictly apply at roadside locations.

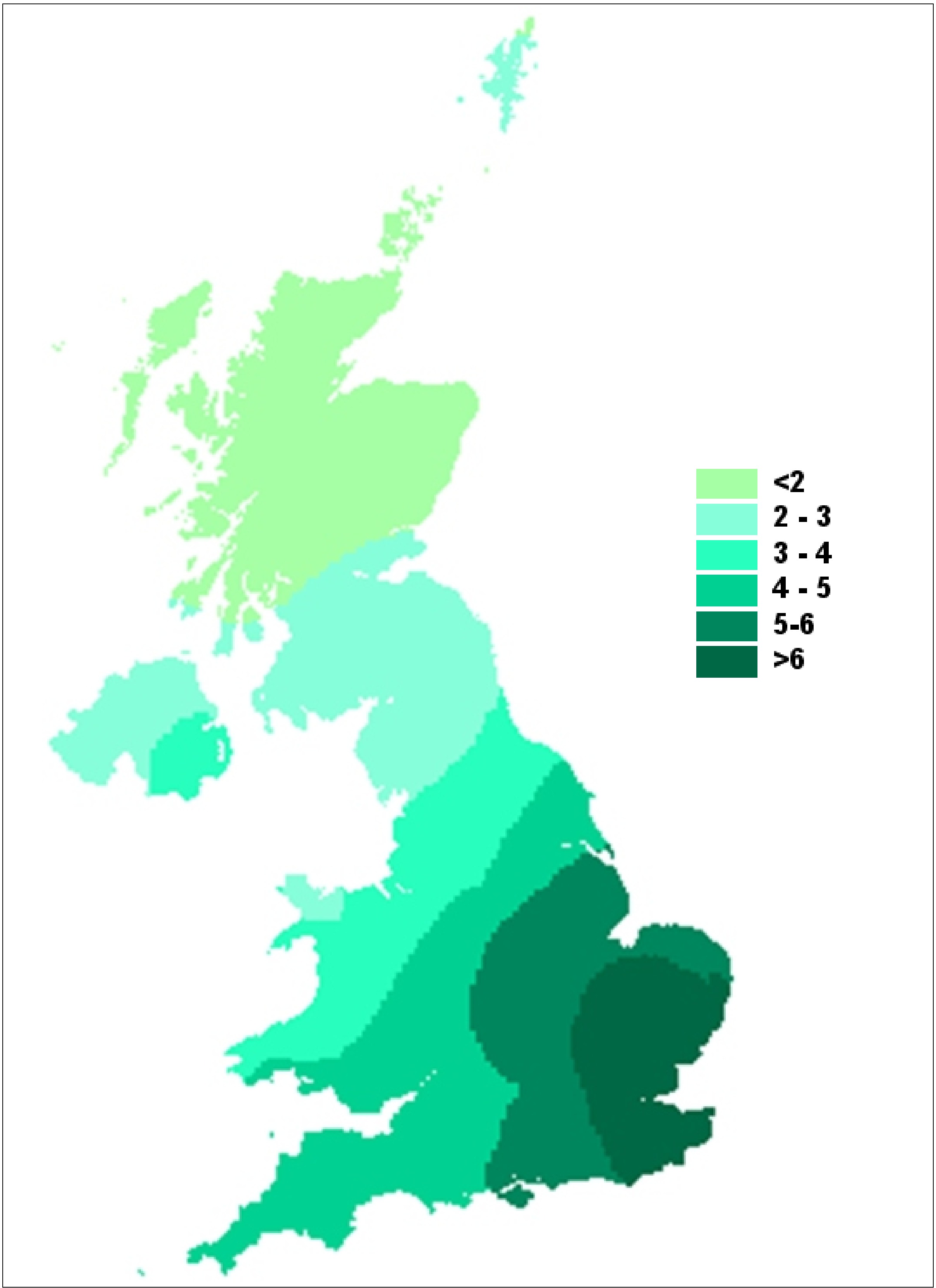

Figure 23 Annual mean secondary $PM_{2.5}$ concentrations (μg/m^3), 2010. Source: Drawn from data provided by Stedman (2010)

To provide a more comprehensive picture of the contributions of different sources, urban and rural areas of the UK have been selected and the results presented as pie-charts. The different areas used are shown in Figure 24 and represent samples for selected, limited geographical areas in the different parts of the UK that are broadly rural or urban. The results are set out in Figure 25 and Figure 26.

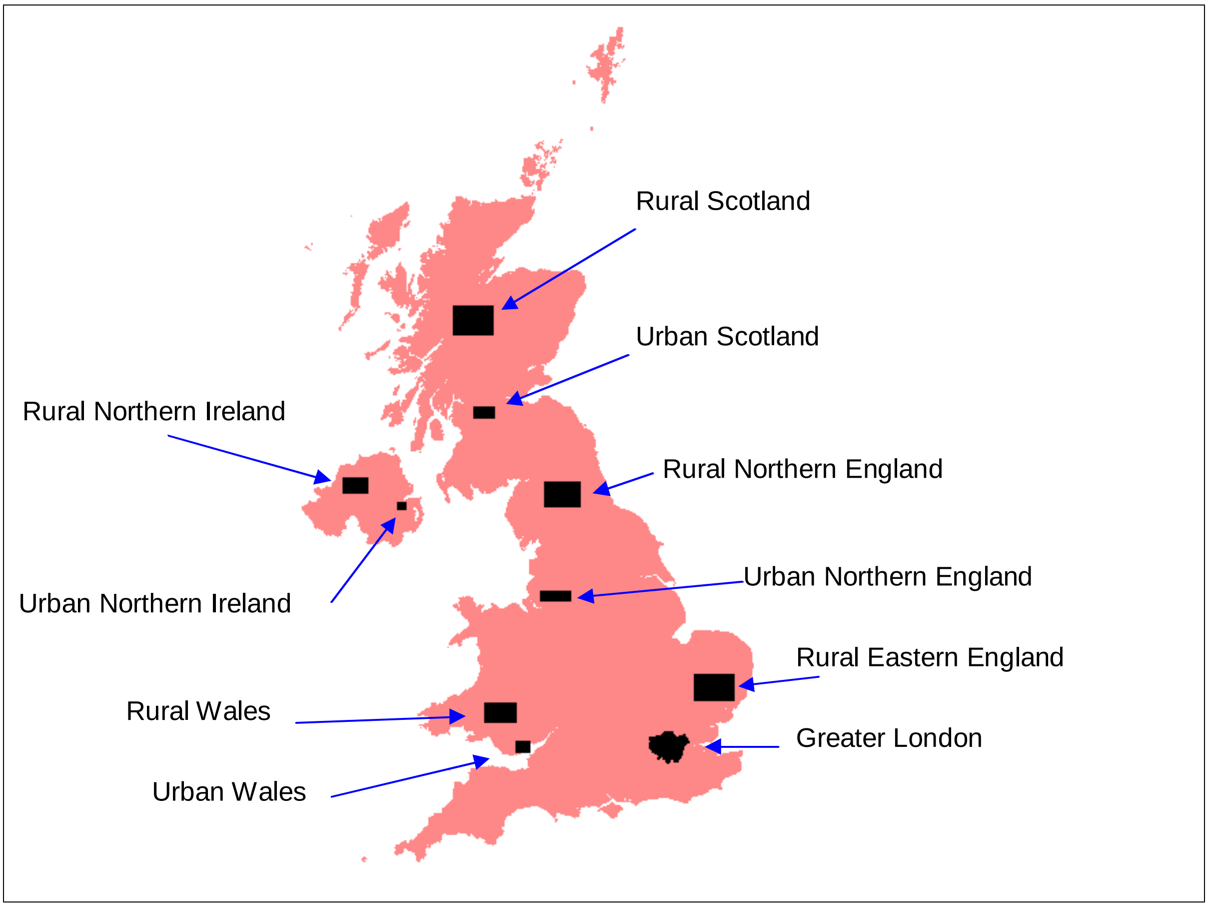

Figure 24 Areas examined for $PM_{2.5}$ source contributions

The source contributions are categorised as follows:

- Vehicle exhausts (exhaust emissions from road traffic on all roads);
- Brake and tyre wear (non-exhaust emissions from road traffic, but does not include any resuspension component);
- Industry (agriculture, combustion in industry, construction, energy production, extraction of fossil fuels, quarries, solvents and waste – treated as area sources);
- Point sources (Part A processes);
- Domestic (domestic, institutional and commercial space heating);
- Secondary (inorganic and organic);
- Sea salt;
- Other (calcium and iron-rich dusts, regional primary PM, other transport sources (*e.g.* aircraft, rail, off-road));
- Unknown residual component (0.75 $\mu g/m^3$ everywhere).

The following conclusions can be drawn:

- The secondary PM contribution is very substantial in all rural and urban areas, accounting for between 31% (in urban Scotland) to almost 60% (in rural south-east England);
- In urban areas, brake and tyre wear emissions (which are not controlled) represent approximately 50% of the exhaust contribution, *i.e.* 33% of the traffic contribution (vehicle exhaust + brake and tyre wear);
- With the exception of London, emissions from industrial sources (industry and point sources) are broadly as important in urban areas as emissions from vehicle exhausts and brake & tyre wear. Within rural areas industrial sources become more important.

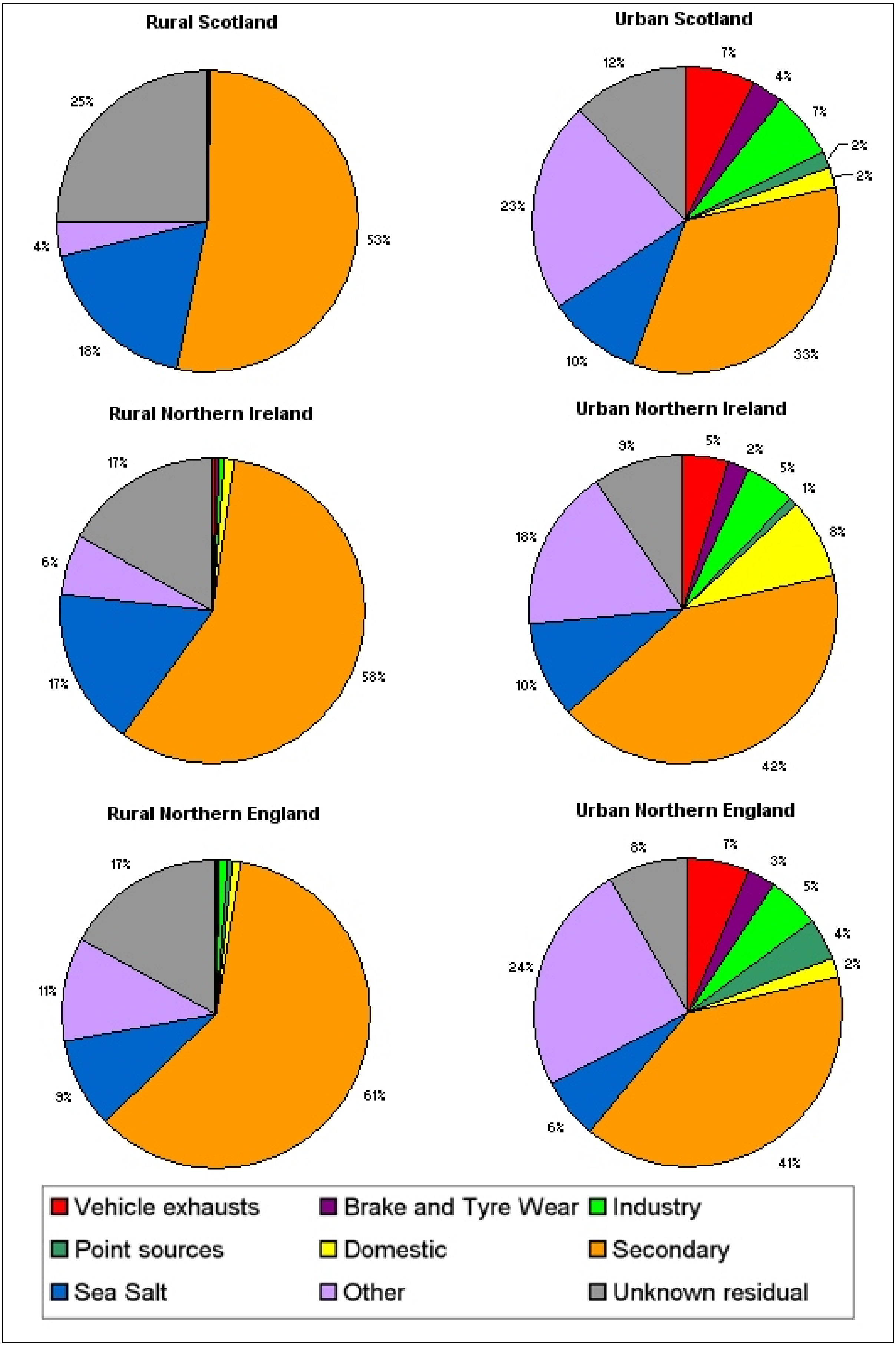

Figure 25 **Source contributions to $PM_{2.5}$ concentrations for different areas of UK from the national mapping exercise in 2010. The areas are shown in Figure 24. Source: Prepared using data provided by Defra (2010a)**

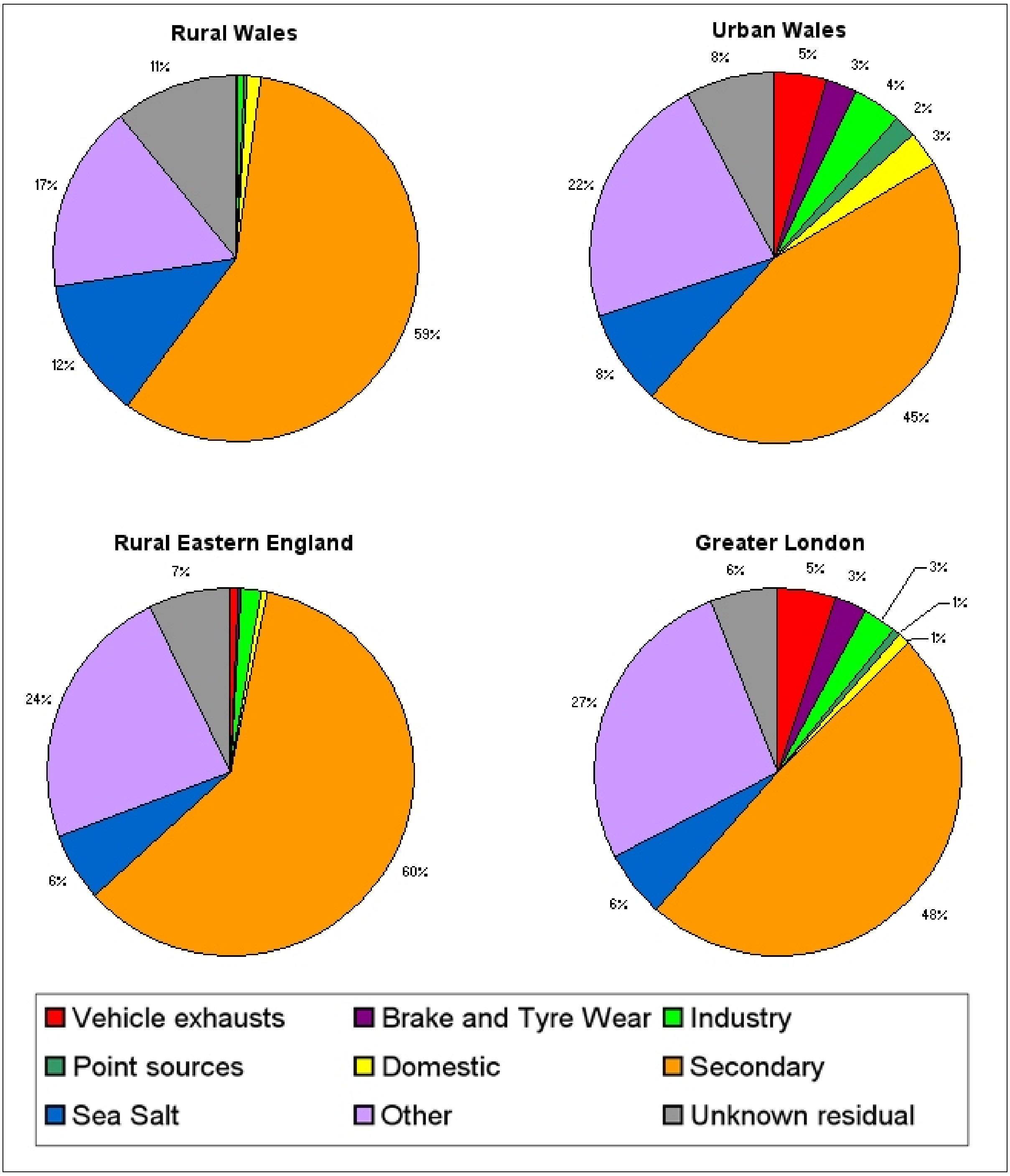

Figure 26 Source contributions to $PM_{2.5}$ concentrations for different areas of UK from the national mapping exercise in 2010. The areas are shown in Figure 24. Source: Prepared using data provided by Defra (2010a)

Yin *et al.* (2010) characterised the chemical composition of particles collected at an urban background site in Birmingham and a rural site 20 km to the west of Birmingham over a 12 month period from May 2007 to April 2008. The results were used within a mass closure model (Harrison *et al.*, 2003), coupled with the US EPA's chemical mass balance model (USEPA, 2010), to identify the key sources. The average concentrations were not very different at the two sites, being 11.2 $\mu g/m^3$ at the urban background site and 10.3 $\mu g/m^3$ at the rural site. The results show that ammonium nitrate and ammonium sulphate dominate, accounting for just under half the $PM_{2.5}$ in both locations (Figure 27). The 'other organic matter' component is also significant, and is considered to be mostly secondary organic aerosol. These results indicate that around 60% of the $PM_{2.5}$ is of secondary origin, while around 10-15% is from diesel and petrol engines (mostly diesel). There is also a significant component, around 10%, assigned to ‘smoking engines’, which is defined as engines emitting atypically large amounts of lubricating oil, which would include off-road engines, such as lawn-mowers and other small engines. It is not clear whether this component is meant to cover some aspect of road vehicles.

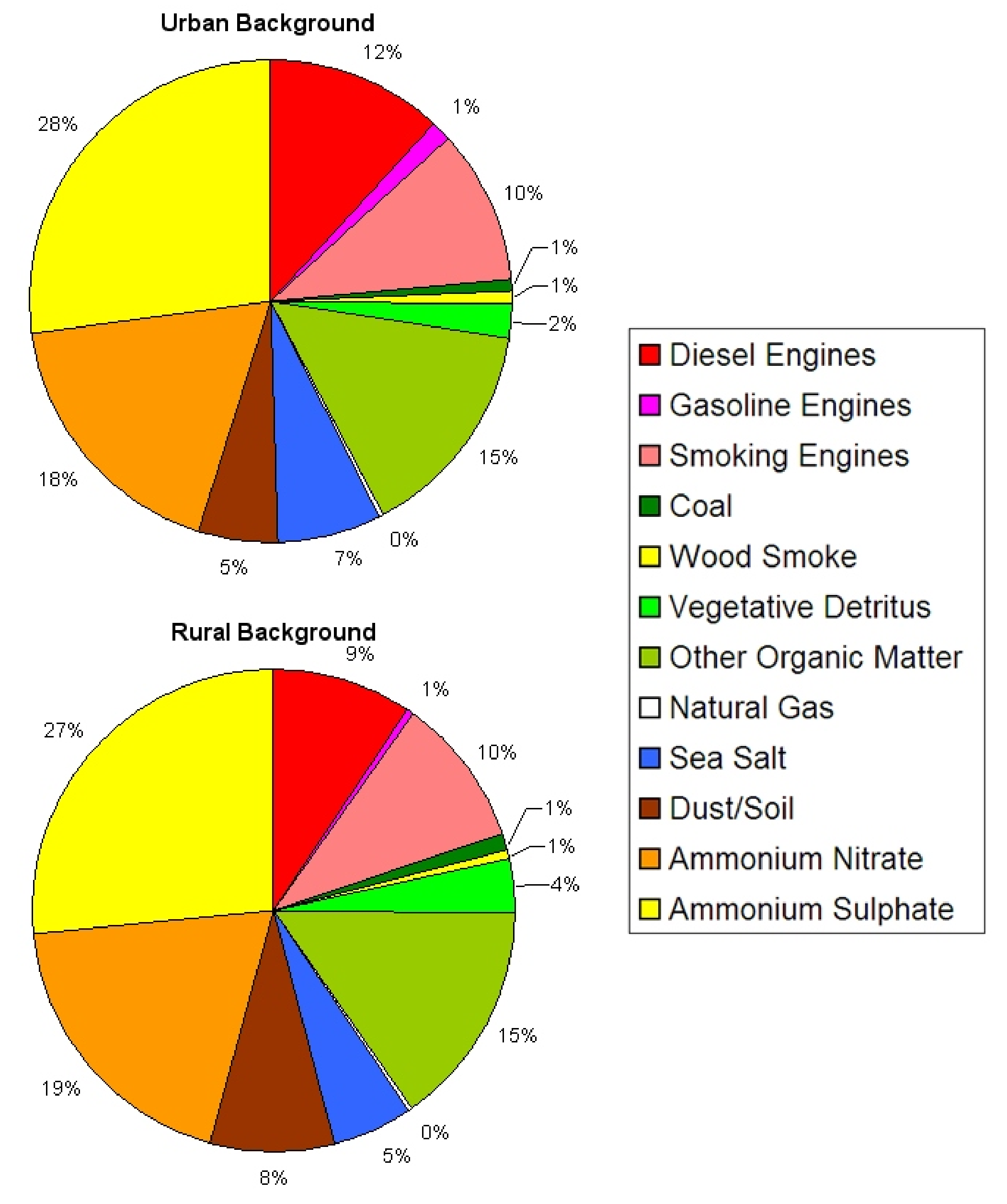

Figure 27 Sources of $PM_{2.5}$ at urban and rural background sites in the West Midlands. Source: Prepared using data from Yin *et al.* (2010).

It is interesting to compare the results from Yin *et al.* (2010) with the source apportionment within the national maps produced by Stedman (2010). Both data sets show reasonable agreement for secondary sources, which account for around half the $PM_{2.5}$, and for road traffic in urban areas, which accounts for around 10% (*cf.* Figure 25, Figure 26 and Figure 27). Yin *et al.* (2010) still show a significant vehicle contribution at their rural site, but the road traffic contributions in the maps of Stedman (2010) are minimal in rural areas. On the other hand, Stedman (2010) shows a small industrial/point source component of primary $PM_{2.5}$ that is essentially absent in the Yin *et al.* (2010) results, as is the brake and tyre wear component. Yin *et al.* (2010), on the other hand, have a dust/soil component that is not explicitly identified in the Stedman (2010) results (although it is within the ‘other’ component). Notably, resuspension of $PM_{2.5}$ from roads is not explicitly included in either data set. The indications are that this resuspension component of $PM_{2.5}$ may be around 4% of the vehicle exhaust component (see Section 3.3.4). There are clearly differences between the two approaches which deserve further investigation.

Allan *et al.* (2010) have analysed the organic fraction of urban aerosol using mass spectrometric analysis, to determine source contributions to primary organic aerosol (POA). They sampled urban background air in London (October 2006 and October/November 2007) and in Manchester (January/February 2007) and applied positive matrix factorisation to determine sources, showing that during the winter months, an average of 40% of the POA was from traffic, 34% from cooking and 26% from solid fuel used for space heating. The solid fuel component was not definable in the results for the warmer October 2006 period. When it was present, the solid fuel component primarily occurred in the late evening and in the middle of the night (20:00 h to 03:00 h). The POA from cooking was associated with aerosol generated from oils and was most prominent during the evening, peaking between about 20:00 h and 22:00 h. The study found that that POA accounted for 47-72% of the organic aerosol, while secondary organic aerosol (SOA) accounted for 28-53%. The high POA contribution will relate to the time of year the samples were collected, with SOA likely to dominate during the summer months.

Using carbon-14 determinations in samples of urban background $PM_{2.5}$ from Birmingham, Heal *et al.* (2010) estimated contributions to the total carbon content, on average, of 27% fossil EC, 20% fossil OC, 2% biomass EC, 10% biomass OC, and 41% biogenic OC (see Section 3.7). The proportion of secondary organic carbon and the fraction of contemporary carbon were higher for air-mass back trajectories arriving in Birmingham from over land, consistent with contributions from terrestrial sources.

4.2 How Much $PM_{2.5}$ is Imported into the UK?

Modelling studies have been used to estimate the contribution of different sources to secondary sulphate and nitrate at different locations in the UK. Results for 2002 as reported by AQEG (2005) are summarised in Figure 28 and Figure 29 (current proportions should only be marginally different, as emissions of precursor gases have declined in both the UK and Europe - see Section 3.6 – and furthermore, this change will be tempered by the non-proportional nature of the relationship between precursor emissions and PM concentrations – see Section 8.13.4). Emissions of sulphur dioxide in UK and Ireland are the predominant source of sulphate aerosol in Scotland and Northern Ireland, while in the South East of England, emissions of sulphur dioxide arising from other northern European countries (France, Benelux, Germany, Denmark, Norway and Sweden), play a significant role. In the case of nitrate aerosol, UK sources are predominant throughout, reflecting the shorter time-frame for formation of nitrates from nitrogen oxides emissions, although there is still a contribution from other northern European countries.

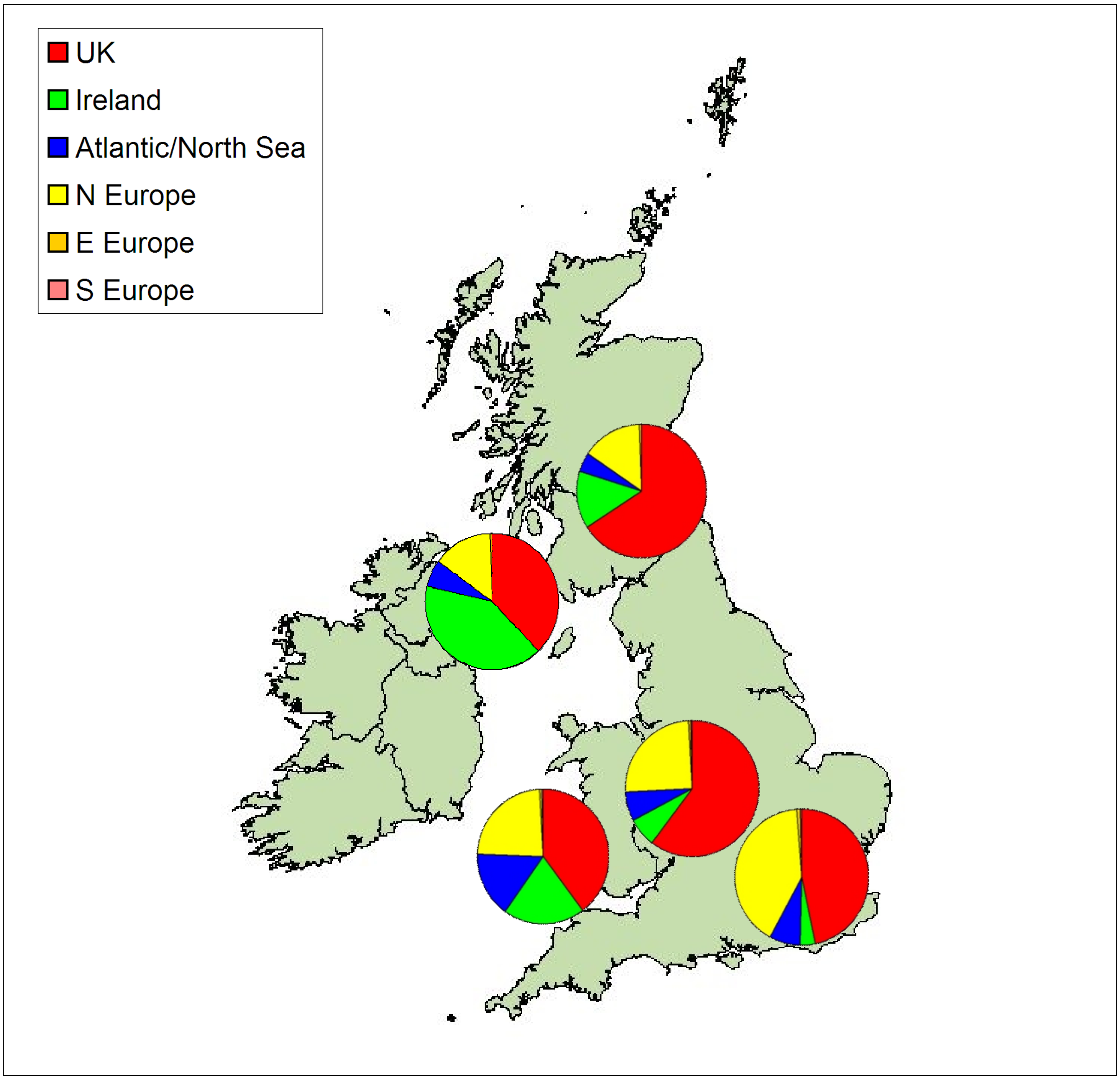

Figure 28 Sources of sulphate in 2002. Source: Prepared using data from AQEG (2005).

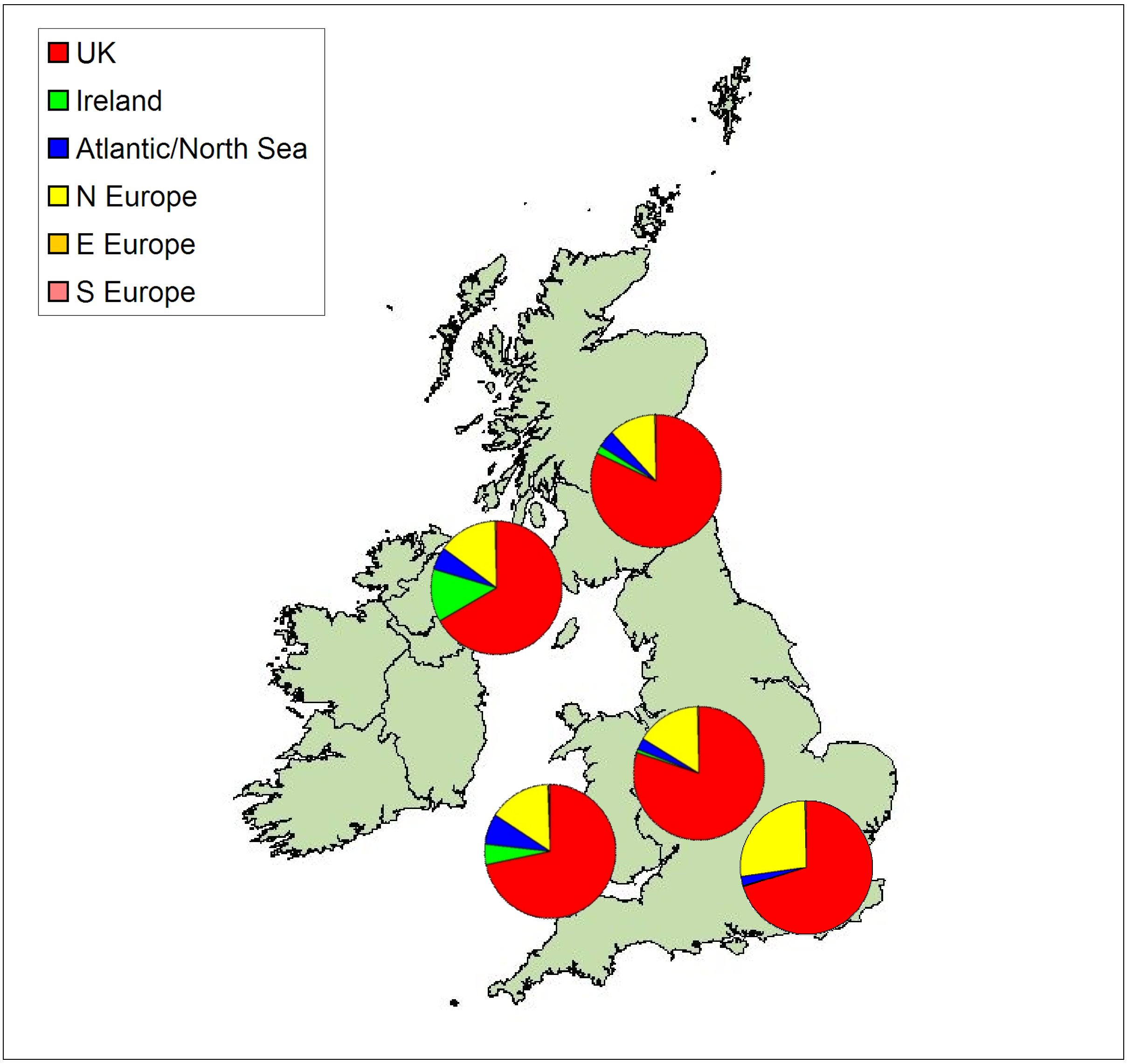

Figure 29 Sources of nitrate in 2002. Source: Prepared using data from AQEG (2005).

Sea salt is a natural source imported to the UK that contributes to PM$_{2.5}$. The work of Grice *et al* (2010) summarises the approach to mapping the sea salt contribution across the UK. This is based on measurements at 28 rural monitoring stations, with the assumption that 27% of the sea salt mass falls in the PM$_{2.5}$ fraction. A map showing the calculated sea salt concentrations across the UK in 2008 is shown in Figure 30. The highest contributions are on the western extremities of the UK, nearest to the Atlantic, declining towards the east. The contributions range from 0.37 to 1.57 µg/m^3.

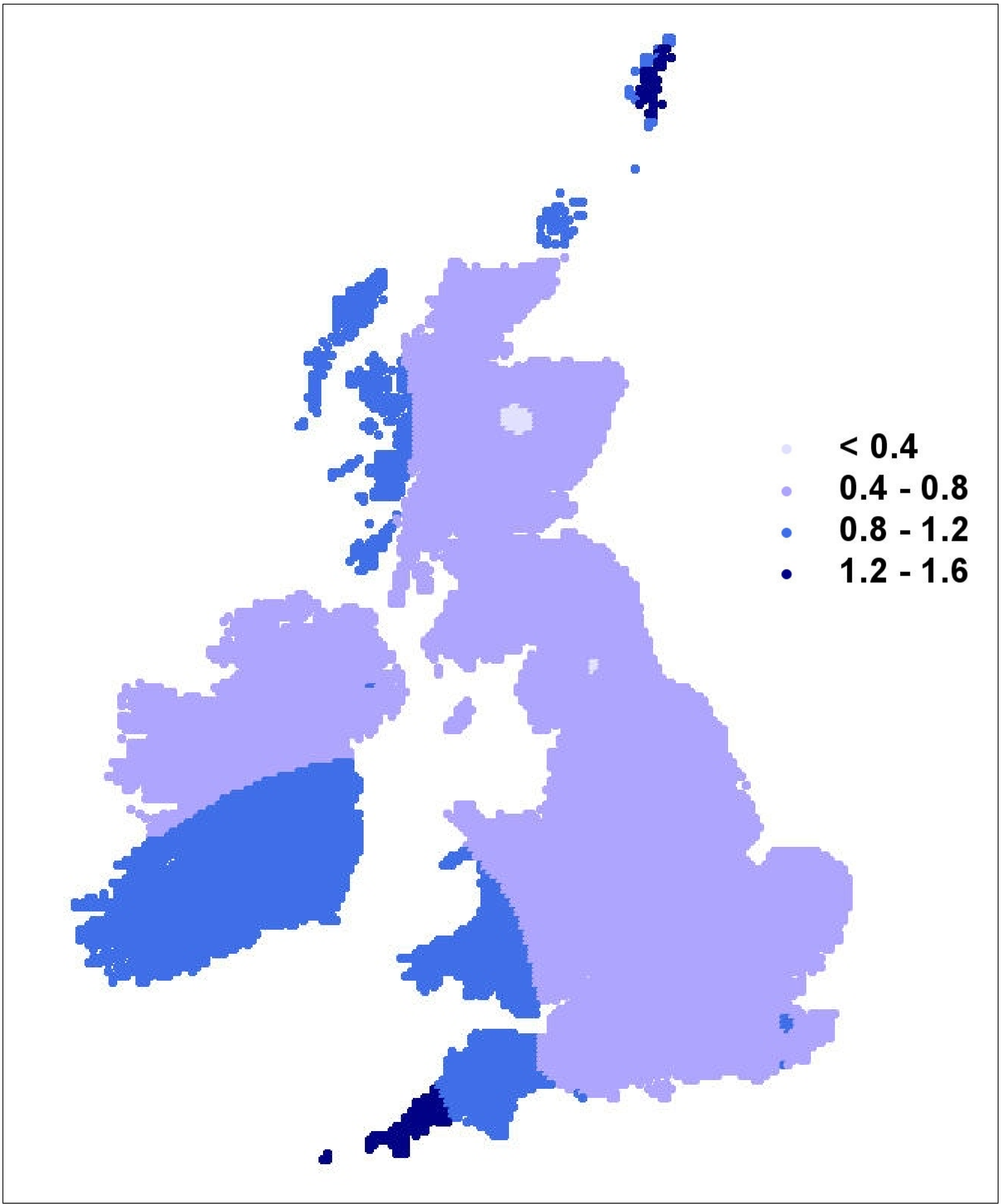

Figure 30 Sea salt concentrations (µg/m^3) in 2008. Source: Drawn from data provided by Stedman (2010).

There is currently no evidence available to describe the contribution of primary anthropogenic sources outside the UK to background concentrations of PM$_{2.5}$, although it is likely to be very small.

5. WHAT ARE THE HEALTH EFFECTS OF $PM_{2.5}$

5.1 What are the Effects of $PM_{2.5}$ on Human Health

5.1.1 Summary answer

Day-to-day changes in $PM_{2.5}$ concentrations are associated with hospital admissions and mortality from respiratory and cardiovascular diseases. This is manifested as increased hospital admissions and numbers of deaths on days of high $PM_{2.5}$ pollution and those immediately following. It is likely that many people suffer less severe effects such as the exacerbation of asthma symptoms or simply feeling unwell (having a reduced activity day) but these outcomes are much more difficult to quantify. Long-term exposure to $PM_{2.5}$ is associated with a shortening of life expectancy. It has been estimated that this amounts to an average of around six months for each person in the UK population, for lifetime exposure at current levels of $PM_{2.5}$, but with considerable variation between individuals. The expert view is that the above associations represent causality.

5.1.2 Extended answer

Our knowledge of the effects of $PM_{2.5}$ exposure on human populations derives mainly from two kinds of epidemiological studies. The first kind of study examines the impact of day-to-day changes in $PM_{2.5}$ concentrations upon hospital admissions and death rates, typically the day following or within a few days of the elevated $PM_{2.5}$ concentrations. Most of the available information comes from studies of PM_{10}, and as $PM_{2.5}$ concentrations typically correlate very highly with PM_{10} concentrations, it is difficult to know for certain how much of the effect is attributable to the $PM_{2.5}$ fraction. The first five lines of Table 12 indicate the estimates of increases in daily mortality (all-cause, respiratory and cardiovascular) for a 10 µg/m^3 increase in PM_{10} concentrations taken from the report of the World Health Organisation (WHO, 2005). The table is accompanied by the comment that:

"*use of these risk relationships in any particular setting is subject to uncertainty relating to their generalisability; on the other hand, there is not yet sufficient evidence to focus on specific PM fractions, whether defined by physical or chemical properties, for the purpose of standard setting.*"

Dominicci *et al.* (2006) examined the effects of $PM_{2.5}$ on hospital admissions for cardiovascular and respiratory diseases in the United States, finding the largest association for heart failure which had a 1.28% increase in risk per 10 µg/m^3 increase in same-day $PM_{2.5}$, but effects were seen for both cardiovascular and respiratory diseases. Analysing data from a number of European countries, Atkinson *et al.* (2001) found positive associations between PM_{10} exposures and hospital admissions for asthma and chronic obstructive pulmonary disease (COPD). In a study of the UK West Midlands, Anderson *et al.* (2001) found significant positive associations between $PM_{2.5}$ concentrations (and black smoke concentrations) and respiratory hospital admissions in the warm season but not the cold season. There was also a positive association of both $PM_{2.5}$ and black smoke with cardiovascular hospital admissions, but this was not statistically significant. Daily mortality was also significantly positively associated with $PM_{2.5}$ concentrations in the warm season of the year. Peters *et al.* (2001) interviewed patients following a recent myocardial infarction (heart attack) and found that the risk of myocardial infarction onset increased in association with elevated concentrations of $PM_{2.5}$, both within the previous two-hour period and in the 24-hour period one day before the onset. Koenig *et al.* (1993) studied elementary school children, finding that an increase in fine particulate air pollution was associated with a decline in the pulmonary function of asthmatic children.

Table 12 Risk estimates for PM exposure, for the metrics specified. Source: WHO (2005).

Outcome	Source	Original Reference	Estimate	95% Confidence Limit
Daily mortality (all-cause)	WHO meta-analysis	WHO (2004)	0.6%/10 µg/m^3 (PM_{10})	0.4 – 0.8
Daily mortality (respiratory)	WHO meta-analysis	WHO (2004)	1.3%/10 µg/m^3 (PM_{10})	0.5 – 2.09
Daily mortality (cardiovascular)	WHO meta-analysis	WHO (2004)	0.9%/10 µg/m^3 (PM_{10})	0.5 – 1.3
Daily mortality (all-causes)	NMMAPS [a] revised	Health Effects Institute (2003)	0.21%/10 µg/m^3 (PM_{10})	0.09 – 0.33
Daily mortality (cardiovascular)	NMMAPS [a] revised	Health Effects Institute (2003)	0.31%/10 µg/m^3 (PM_{10})	0.13 – 0.49
Long-term mortality (all-cause)	ACS CPS II [b] 1979 – 1983	Pope *et al.* (2002)	4%/10 µg/m^3 ($PM_{2.5}$)	1 - 8
Long-term mortality (cardiopulmonary)	ACS CPS II [b] 1979 – 1983	Pope *et al.* (2002)	6%/10 µg/m^3 ($PM_{2.5}$)	2 – 10

[a] NMMAPS = National Morbidity, Mortality and Air Pollution Study

[b] ACS SPS II = American Cancer Society Cancer Prevention Study II

The second kind of epidemiological study compares health outcomes over the long term for populations living in areas with different exposures to PM. Seminal studies from North America (Dockery *et al*., 1993; Pope *et al*., 1995; 2002) have shown higher death rates in cities with higher concentrations of airborne particles. In the case of the very large American Cancer Society Study of Pope *et al.* (1995; 2002) highly significant correlations were found between death rates and concentrations of $PM_{2.5}$ and airborne sulphate. These studies demonstrated an increased risk of all cause, cardiopulmonary and lung cancer mortality. The bottom two lines of Table 12 indicate estimates of 4% and 6% increases in long-term risk from all-cause and cardiopulmonary mortality, respectively, for a 10 µg/m^3 increase in $PM_{2.5}$.

In the UK, a review by the Committee on the Medical Effects of Air Pollutants (COMEAP) has recommended a relative risk factor for all cause mortality associated with each 10 µg/m^3 exposure to $PM_{2.5}$ of 1.06 (*i.e.* 6%/10 µg/m^3) with a 95% confidence interval (CI) of 1.02 to 1.11 (COMEAP, 2009).

Conversion of a long-term risk estimate into loss of life expectancy requires application of a complex life-table approach. However, the chronic effects of long-term exposure to $PM_{2.5}$ are generally considered to be of greater public health concern than the short-term (acute) effects on daily hospital admissions and mortality. In the UK, the estimated mean loss of life expectancy in the range 176–182 days (~6 months) for people born in 2008 and exposed to the 2008 population-weighted anthropogenic $PM_{2.5}$ concentration of 10.39 µg/m^3 for the duration of their lives (Defra, 2010b). It is expected that impacts on life expectancy will vary widely between individuals (see Sections 5.2 and 5.4); so if, for example, only one in ten individuals were affected, estimated mean loss of life expectancy for the affected individuals would be in the range 1,760–1,820 days (~5 years) . The estimated total life years lost across the UK population at year 2008 $PM_{2.5}$ concentrations (over a 100 year period, including new birth cohorts) for this risk estimate is 18.2-32.4 million life years. (For context, the total life years lived over the quantification period is about 5,000 million.) The ranges in values reflect different assumptions about the extent of lag, if any, between exposure and mortality effect. The view expressed by COMEAP is that "*a noteworthy proportion of the total effect is likely to appear within the first five years*." (COMEAP, 2009). The corollary of this observation is that the greatest health benefit from PM concentration reduction will occur in the first few years after the reduction.

It is not straightforward to compare the health risks associated with exposure to $PM_{2.5}$ with other risks, but the Department of Health commissioned work from the Institute of Occupational Medicine to compare the benefits of reducing $PM_{2.5}$ exposure by 10 µg/m^3 (considered equivalent to eliminating the man-made contribution to exposure in 2005) with the elimination of motor vehicle traffic accidents and the elimination of exposure to passive smoking. The results were reported by the Environmental Audit Commission (2010). Since then, Defra (2010b) has reduced the expected gain in life expectancy from the value cited by the Environmental Audit Commission. The expected gains in life expectancy for elimination of anthropogenic $PM_{2.5}$, road traffic accidents and passive smoking are set out below:

- 6 months for elimination of exposure to anthropogenic $PM_{2.5}$;
- 1-3 months for elimination of road traffic accidents; and
- 2-3 months for elimination of exposure to passive smoking.

5.2 Is there a Safe Level for Exposure to $PM_{2.5}$?

5.2.1 Summary answer

None has so far been identified.

5.2.2 Extended answer

The epidemiological studies establish a relationship between exposure, determined using fixed monitoring sites, and rates of a health outcome (generally hospital admissions or mortality). Within the range of concentrations studied (which is the range of concentrations found in the ambient air of the cities studied) all studies to date have shown an approximately linear increase in risk with exposure concentration, and no demonstrable threshold. In this context, the term threshold refers to a concentration below which no effects are seen. In most studies, there will have been occasions with very low concentrations but these are relatively infrequent and therefore conclusions based upon them have a low level of confidence. It is therefore difficult to be sure that there is no threshold, but the prevailing expert view is that a threshold is unlikely. This opinion is based on the fact that there is a wide range of individual susceptibilities within the population and, while the majority of the population will be unaffected by typical ambient pollution levels (at least in the short-term), there will be a long-term impact on their life expectancy, and those most susceptible by virtue of their age, infirmity or genetic make-up may be affected by extremely low concentrations in the short term.

5.3 Are the Consequences of Short-Term Exposure Different from Long-Term Exposure?

5.3.1 Summary answer

The consequences are very similar in that both short-term and long-term particle exposures are associated with an exacerbation of respiratory and cardiovascular diseases including, for long-term exposures, lung cancer and, possibly, neurodegenerative disease and incident type 2 diabetes. For those affected, short-term exposures can result in health events within a few days of exposure while the long-term exposures manifest themselves as chronic morbidity and reduction in life expectancy.

5.3.2 Extended answer

Zanobetti *et al.* (2003) studied the time period over which short-term PM_{10} exposure effects manifest themselves. They found that effects persist for up to a month and are much larger than those normally evaluated over a few days. Pope (2007) concluded that short-term exposure studies capture only a small amount of the overall health effects of long-term exposure to PM. Longer-term exposures have larger, more persistent cumulative effects than short-term exposures, including associations with development of lung cancer. Some evidence also exists for adverse impact of long-term exposure to PM on neural and cognitive function (Calderon-Garciduenos *et al*., 2004; 2009) and incidence of type 2 diabetes (Kramer *et al*., 2010).

5.4 Does $PM_{2.5}$ Only Affect the Health of Particularly Sensitive Individuals? Does this Differ for Short and Long-Term Exposures?

5.4.1 Summary answer

There is limited evidence on this point, but the acute effects of $PM_{2.5}$ exposure are expected to be predominantly amongst those with pre-existing disease, and in particular amongst children and the very elderly. Long-term effects of $PM_{2.5}$ exposure upon mortality appear to be greatest in men and those in the lowest educational category.

5.4.2 Extended answer

Rather few epidemiological studies on $PM_{2.5}$ have sought to study effects on susceptible groups. This is primarily because in some kinds of study it is not possible to identify such groups and more generally, by limiting the population numbers under study, the statistical power is reduced and it becomes more difficult to establish statistical significance of findings. Some of the studies of short-term effects have looked at different age groups and a number (*e.g.* Goldberg *et al*., 2001; Ostro *et al*., 2006) have found greater effects in the over 65 age group. The study of the UK West Midlands, which found adverse effects of $PM_{2.5}$ exposure during the warm season, also found greater effects upon respiratory admissions in the over 65 age group than in the all age population (Anderson *et al*., 2001). In their study of primary school children, Koenig *et al.* (1993) found pulmonary function of asthmatic children to decline with increased levels of $PM_{2.5}$. Ko *et al.* (2007) also showed greater risk of hospital admission for asthma with increased concentrations of $PM_{2.5}$ in Hong Kong, for children aged 14 years and under, compared with adults.

The extended analysis of the American Cancer Society Study (Pope *et al*., 2002) examined the effects of a number of co-variates on the impact of $PM_{2.5}$ exposure upon life expectancy. Those that came through as showing significant positive effects were: a) men showed greater susceptibility than women and b) a relationship to educational status, with those having less than high school education being the most susceptible sub-group of the population.

5.5 Is $PM_{2.5}$ More Toxic than PM_{10}?

5.5.1 Summary answer

$PM_{2.5}$ is a sub-component of PM_{10}, frequently comprising about 70% of the total, thus the distinction between these two metrics in epidemiological studies is not easy. However, there are indications in the literature that $PM_{2.5}$ may be slightly more toxic per unit mass than PM_{10}. Regarding the effects of long-term exposure, the stronger relationships are with $PM_{2.5}$ concentrations but this may be an artefact of the study design.

5.5.2 Extended answer

Studies that have included both $PM_{2.5}$ and PM_{10} are relatively few in number but some such as the UK West Midlands Study (Anderson *et al*., 2001) show slightly greater relative risk factor for $PM_{2.5}$ than PM_{10} indicating that $PM_{2.5}$ may be slightly more toxic than PM_{10} per unit mass. The more recent study conducted in London by Atkinson *et al.* (2010) is rather more equivocal giving little indication of a clear difference in relatively toxicity of $PM_{2.5}$ and PM_{10}.

Regarding the effects of long-term exposure, Pope *et al.* (2002) based their analysis primarily upon $PM_{2.5}$ exposures but comment that:
"*weaker less consistent mortality association were observed with* PM_{10} *and* PM_{15}."

The comment regarding study design is elaborated upon in the answer to the question in the next Section.

5.6 Is the $PM_{2.5\text{-}10}$ Fraction Toxic?

5.6.1 Summary answer

Epidemiological studies on the short-term health effects of the coarse particle fraction, $PM_{2.5\text{-}10}$, generally indicate lower toxicity for this fraction than for $PM_{2.5}$ which is consistent with the lower coefficients for PM_{10} than $PM_{2.5}$ in the health effects studies referred to above. On the other hand, many toxicological studies demonstrate toxicity inherent to the coarse particle fraction. There are insufficient epidemiological studies to address the issue of long-term health effects of the coarse fraction. Overall, however, it is likely that the $PM_{2.5\text{-}10}$ fraction is responsible for some adverse health outcomes.

5.6.2 Extended answer

A number of studies have looked at the health effects of the coarse particle fraction referred to as $PM_{2.5\text{-}10}$. Almost without exception, the studies on short-term effects have indicated generally lower toxicity for this fraction than for $PM_{2.5}$ and this would be consistent with the lower coefficients for PM_{10} than $PM_{2.5}$ in the health effects studies referred to above. In the UK studies (Anderson *et al*., 2001; Atkinson *et al*., 2010) there are not statistically significant associations between coarse particle exposure and the evaluated heath outcomes. However, Brunekreef and Forsberg (2005) carried out a systematic review of studies which analysed fine and coarse PM jointly and examined the epidemiological evidence for effects of coarse particles on health. They conclude that:

"*In studies of chronic or obstructive pulmonary disease, asthma and respiratory admissions, coarse PM has a stronger or as strong short-term effect as fine PM, suggesting that coarse PM may lead to adverse responses in the lungs triggering processes leading to hospital admissions. There is also support for an association between coarse PM and cardiovascular admissions.*"

The recent review by the USEPA (2009) concluded that:

"A growing body of evidence both from epidemiological and toxicological studies… supports the general conclusion that $PM_{2.5}$ *(or one or more* $PM_{2.5}$ *components), acting alone and/or in combination with gaseous co-pollutants, are likely causally related to cardiovascular and respiratory mortality and morbidity."*

"A much more limited body of evidence is suggestive of associations between short-term (but not long-term) exposures to ambient coarse-fraction thoracic particles… and various mortality and morbidity effects observed at times in some locations. This suggests that $PM_{2.5\text{-}10}$*, or some constituent component(s) of* $PM_{2.5\text{-}10}$*, may contribute under some circumstances to increased human health risks… with somewhat stronger evidence for… associations with morbidity (especially respiratory) endpoints than for mortality".*

They summarised the evidence as set out in Table 13.

Table 13 Summary of evidence of causality for fine and coarse PM by exposure duration and health outcome. Source: USEPA (2009).

Size Fraction	Exposure	Outcome	Causality
$PM_{2.5}$	Short-term	Cardiovascular effects	Causal
		Respiratory effects	Likely to be causal
		Central nervous system	Inadequate
		Mortality	Causal
	Long-term	Cardiovascular effects	Causal
		Respiratory effects	Likely to be causal
		Mortality	Causal
		Reproductive and developmental	Suggestive
		Cancer, mutagenicity, genotoxicity	Suggestive
$PM_{2.5-10}$	Short-term	Cardiovascular effects	Suggestive
		Respiratory effects	Suggestive
		Central nervous system	Inadequate
		Mortality	Suggestive
	Long-term	Cardiovascular effects	Inadequate
		Respiratory effects	Inadequate
		Mortality	Inadequate
		Reproductive and developmental	Inadequate
		Cancer, mutagenicity, genotoxicity	Inadequate

There are reasons to believe that coarse particles may indeed have a substantial inherent toxicity. This comes from two lines of investigation. Wilson and Suh (1997) examined the spatial distribution of coarse particle concentrations, finding it to be much less homogeneous than that of $PM_{2.5}$ concentrations. They argued that since in most epidemiological studies a small number of fixed point monitors were used to describe the exposure of large populations, the greater spatial variability of coarse particle concentrations would lead to greater exposure misclassification which tends to bias results towards the null, *i.e.* it leads to an underestimation of the impacts of coarse particle exposure.

Studies in the laboratory of the *in vitro* toxicity of coarse and fine particles paint a mixed picture. Diociaiuti *et al.* (2001) found greater toxic effects in the study of human blood cells caused by fine particles than coarse particles, while Jalava *et al.* (2007) found the coarse fraction of PM samples collected across six different sites in Europe to exhibit greater inflammatory activity on mouse macrophages than the fine fraction. Studies investigating inflammation induced by instillation of PM into animal models (*e.g.* Schins *et al.*, 2004, Gerlofs-Nijland *et al.*, 2009) also report much of the toxicity to reside in the coarse particle fraction most probably because of the enrichment of a number of trace elements known to have higher oxidative potential within this fraction.

Concerning the long-term effects of particle exposure, Pope *et al.* (2002) used $PM_{15\text{-}2.5}$ as a measure of the coarse particle fraction and failed to find a consistent association with mortality. However, their study design is particularly susceptible to the problems of exposure misclassification referred to above. In their study of Californian Seventh Day Adventists, McDonnell *et al.* (2000) found associations of long-term concentrations of PM_{10} with mortality but concluded that:
"*previously observed associations of long-term ambient PM_{10} concentration with mortality for males were best explained by a relationship with the fine fraction of PM_{10}* (*i.e. $PM_{2.5}$*) *rather than with the coarse fraction of PM_{10}.*"

This study would also have been influenced by the spatial inhomogeneity of coarse particles.

5.7 Are Some Specific Components of $PM_{2.5}$ Responsible for its Toxicity, and What are Their Mechanisms of Toxicity?

5.7.1 Summary answer

The literature gives some indication that combustion particles from sources such as road traffic and oil combustion may be more toxic per unit mass than particles from other sources. There are also indications that the nanoparticle fraction, *i.e.* particles less than 100 nanometres diameter, may have an enhanced toxicity. However, any such conclusions are currently provisional. There are a number of mechanisms for the toxicity of PM which explain effects both on the respiratory system and the cardiovascular system.

5.7.2 Extended answer

From the perspective of developing policy, the most valuable answers to this question derive from studies on human subjects or human populations. Human challenge studies in which human subjects are exposed to controlled levels of pollutants in the laboratory accompanied by monitoring of their physiological and biochemical responses have not given clear answers to this question, probably because ethical constraints severely limit the exposures that can be administered. Thus, whilst Mills *et al.* (2005) showed that inhalation of dilute diesel exhaust impairs important aspects of vascular function in healthy human volunteers, Routledge *et al.* (2006) in a study of the effects of carbon particles and sulphur dioxide on heart rate variability failed to find a significant effect from the particles, although the sulphur dioxide did exert an impact.

A number of epidemiological studies have sought to link either individual chemical constituents of PM, individual size ranges of PM, or individual sources of PM, with adverse effects on health. Laden *et al.* (2000) applied multi-variate statistics to multi-element particle concentration data to identify factors classified as soil and crustal material, motor vehicle exhaust and coal combustion in ambient particles. In a study of short-term effects of $PM_{2.5}$, they found that a 10 $\mu g/m^3$ increase in $PM_{2.5}$ from mobile sources accounted for a 3.4% increase in daily mortality, while the equivalent increase in fine particles from coal combustion sources accounted for a 1.1% increase, and $PM_{2.5}$ crustal particles were not associated with daily mortality. This study was, however, based upon a very old dataset from a time when lead additives were used extensively in motor fuels. In a more recent study, Sarnat *et al.* (2008) also applied source apportionment methods and found consistent associations between $PM_{2.5}$ from mobile sources and biomass burning with both cardiovascular and respiratory emergency department visits, and between sulphate-rich secondary $PM_{2.5}$ and respiratory visits. Peng *et al.* (2009) studied the association between hospital admissions for cardiovascular disease and respiratory disease with the chemical components of $PM_{2.5}$ in the United States. They concluded that ambient levels of elemental carbon and organic carbonaceous matter, which are generated primarily from vehicle emissions, diesel and wood burning, were associated with the largest risks of emergency hospitalisation across the major chemical constituents of $PM_{2.5}$. Tsai *et al.* (2000) also applied source apportionment methods to airborne PM and found significant associations between mortality and several PM sources including oil burning, industry, sulphate aerosol and motor vehicles. Burnett *et al.* (2000) examined the effects of $PM_{2.5}$ upon daily mortality rates in Canada, finding the strongest associations with sulphate, iron, nickel and zinc. The three metallic components (iron, nickel and zinc) are most probably indicative of emissions from road traffic and oil combustion. In their study of the UK West Midlands, Anderson *et al.* (2001) found positive associations of both black smoke (indicative of combustion sources, mainly diesel traffic) and sulphate with both respiratory and cardiovascular health outcomes.

In a study of daily mortality and hospital admissions in London, Atkinson *et al.* (2010) used the traditional mass and chemical component metrics but also examined associations with particle number count. The latter metric is a reflection of ultrafine particles, *i.e.* particles of less than 100 nanometres diameter, which, in the atmosphere of London, derive largely from diesel vehicle emissions. They concluded that:

"particle number concentrations were associated with daily mortality and admissions, particularly for cardiovascular diseases lagged one day ...... Secondary pollutants especially for non-primary $PM_{2.5}$ and nitrate and sulphate were more important for respiratory outcomes."

Wichmann and Peters (2000) also investigated associations between ultrafine particle number and daily mortality in Erfurt, Germany. Ultrafine particle number had comparable effects to fine particle mass, although the latter had more immediate effects whilst the former showed effects that were delayed by a few days. The immediate effects were clearer in respiratory cases, whereas delayed effects were clearer in cardiovascular cases.

Some studies have also sought to elucidate the effects of specific chemical components in the long-term impacts of $PM_{2.5}$ exposure. Bell *et al.* (2009) used long-term and short-term average concentrations of $PM_{2.5}$ chemical components for 2005 from the US concluding that communities with higher $PM_{2.5}$ content of nickel, vanadium and elemental carbon and/or their related sources were found to have higher risk of hospitalisations associated with short-term exposure to $PM_{2.5}$. This indicts oil burning (a source of nickel and vanadium) and diesel vehicles (the main source of elemental carbon). After examining the survival of a cohort of US military veterans, Lipfert *et al.* (2006) concluded that traffic density appears to be the most important predictor of survival, but potential contributions were also seen for nitrogen dioxide, nitrate, elemental carbon, nickel and vanadium, highly consistent with the results of the short-term effect studies. Chen and Lippmann (2009) carried out both human and laboratory animal exposures to concentrated ambient particles and performed analyses of the results of human population studies, including both the long-term and short-term effects studies. They found that in the large US study of daily mortality (NMMAPS) the differences in mortality risk coefficients for fine PM components were highest for nickel, vanadium and elemental carbon. Only for the former two were they statistically significant, pointing to oil burning emissions as playing a particular role in particle toxicity. In a critical review of the health impact of common inorganic components of $PM_{2.5}$ in ambient air, Schlesinger (2007) highlights the large database for sulphate but points to certain inconsistencies across epidemiological studies and a lack of coherence with toxicology studies, which is explored in the answer to the question in Section 5.9. Schlesinger concludes that the limited data for nitrate suggest little or no adverse effects at current levels and that crustal components of $PM_{2.5}$ are not likely by themselves to produce significant health risks, particularly since these components do not have unequivocal biological plausibility from toxicological studies.

There is also evidence from *in vitro* studies and animal models that specific particle sources, sizes or composition may exert higher toxicities than others. There have been many relevant studies, including the following. Gavett *et al.* (1997) investigated the effect of composition of two residual fly ash particle samples by intratracheal installation in rats. They concluded that the composition of soluble metals and sulphate leached from the fly ash is critical in the development of airway hyper-reactivity and lung injury. Happo *et al.* (2008) investigated the association between chemical composition and sources of different size fractions of PM from six European cities and inflammatory activity via instillation in the mouse lung. They concluded that local sources of incomplete combustion and resuspended road dust were most important. Cho *et al.* (2009) studied size-fractionated PM obtained at different distances from a highway on acute cardiopulmonary toxicity in mice. The results indicated that on a comparative mass basis, the coarse and ultrafine PM affected the lung and heart respectively. In a multi-centre European study, Kunzli *et al.* (2006) examined the capacity of $PM_{2.5}$ samples to generate hydroxyl radicals in the presence of hydrogen peroxide, as well as their capacity to deplete anti-oxidants from a synthetic model of respiratory tract lining fluid. They found that PM oxidative activity varied significantly among European sampling sites, but that correlations between oxidative activity and all other characteristics of PM were low both within centres (temporal correlation) and across communities (annual mean). Other studies have, however, attached considerable importance to the transition metal content of particles. Wessels *et al.* (2010) deployed a range of *in vitro* oxidant generation and toxicity tests to size-fractionated particles collected in Great Britain and Ireland at sites with a wide range of pollutant loadings. They found that PM collected at high traffic locations generally showed the strongest oxidant capacity and toxicity and that significant correlations were observed between the oxidant generating potential and all toxicological endpoints investigated. Trace metal enrichment at the traffic polluted sites appears to have been an important factor.

There are number of mechanistic explanations of these observations. The most pervasive hypothesis potentially explaining both respiratory and cardiovascular effects is that particles depositing in the human body exert oxidative stress which, in turn, generates inflammation. This results in a cascade of physiological processes (Donaldson *et al*., 2005). In terms of a respiratory condition such as asthma, oxidative stress on its own would appear to be a sufficient mechanism to provoke a narrowing of the airways leading to a worsening of symptoms. It has, however, proved more challenging to understand how the inhalation of airborne particles can affect the cardiovascular system. A breakthrough was provided by the hypothesis put forward by Seaton *et al.* (1995), which proposed that ultrafine particles were able to penetrate the lung wall, depositing in the pulmonary interstitium between the lung and the bloodstream. In doing so, they set up an inflammatory response resulting in a cascade of clotting factors leading to an increased risk of a cardiac event. Subsequent additional hypotheses have led to the suggestion that ultrafine (nano) particles can penetrate into the bloodstream causing a destabilisation of atheromous plaques on the arterial walls hence provoking a cardiac event. An alternative suggestion for which there has been less evidence is that particles depositing in the respiratory system affect the autonomic nervous system leading to a reduction in heart rate variability, which is a known risk factor for a fatal dysrhythmia (Donaldson *et al*., 2005). Mills *et al.* (2008) have reviewed the adverse cardiovascular effects of air pollution. They conclude that the main arbiter of cardiovascular effects including hospital admissions with angina, myocardial infarction and heart failure is combustion-generated nanoparticles that incorporate reactive organic and transition metal components. They argue that inhalation of this PM leads to pulmonary inflammation with secondary systemic effects or, after translocation from the lung into the circulation, to direct toxic cardiovascular effects.

The studies of the long-term effects of $PM_{2.5}$ have demonstrated an association with lung cancer in non-smokers. Harrison *et al.* (2004) examined whether this association could be explained through exposure to known chemical carcinogens in the atmosphere, or whether $PM_{2.5}$ itself could exert carcinogenic activity irrespective of the presence of chemical carcinogens. The study found that, accounting for likely latency periods, concentrations of known chemical carcinogens could plausibly account for the carcinogenic effects of $PM_{2.5}$ exposure.

5.8 Are Traffic Particles Especially Important?

5.8.1 Summary answer

The work described above concerning the toxicity of specific components of PM_{10} indicates that traffic particles may have a higher toxicity than some other components and given their important contribution to $PM_{2.5}$ concentrations, can be regarded as having a special importance.

5.8.2 Extended answer

A number of the aforementioned studies (*e.g.* Laden *et al*., 2000; Sarnat *et al*., 2008; Lipfert *et al*. 2006; Wessels *et al*., 2010) have provided evidence through epidemiological or toxicology studies that road traffic particles are a major contributor to the adverse effects of PM exposure. Supporting evidence comes from a number of other studies. The Dutch NLCS-AIR cohort study (Beelen *et al*., 2008; Brunekreef *et al*., 2009) involved an examination of many risk factors, including point of residence. Significant associations between natural cause and respiratory mortality were found for nitrogen dioxide and black smoke, the latter being a strong indicator of diesel traffic. Grahame and Schlesinger (2007; 2010) examined the impacts of different source types upon health outcomes. The former paper concluded that public health was likely to be better protected by reduction of various vehicular emissions than by continued regulation of the total mass of $PM_{2.5}$ as if all PM in this mode is equitoxic. The latter paper deals specifically with cardiovascular health, reporting numerous studies that found significantly higher risks of cardiovascular morbidity and mortality for people living in close proximity to major roadways versus those living further away.

Janssen *et al.* (2003) studied the respiratory health of children attending schools located close to busy motorways. They found that respiratory symptoms increased near motorways with high truck but not high car traffic counts. Sensitisation to pollen also increased in relation to truck but not car traffic counts. Mills *et al.* (2005) exposed 30 healthy men to diluted diesel exhaust or air for one hour during intermittent exercise. They found that at levels encountered in an urban environment, inhalation of dilute diesel exhaust impaired the regulation of vascular tone and endogenous fibrinolysis providing a potential mechanism linking air pollution to the pathogenesis of atherothrombosis and acute myocardial infarction. Gerlofs-Nijland (2007) exposed rats by intratracheal installation to samples of coarse and fine PM. They found a trend that suggests that samples from high traffic sites were the most toxic.

5.9 Are Primary Components of $PM_{2.5}$ More Toxic than Secondary Components?

5.9.1 Summary answer

The evidence is inconclusive. The fact that particles from road traffic and fuel oil combustion are indicated by many studies to be especially toxic suggests a positive answer. However, sulphate, which is a secondary component, features strongly in the positive results of many epidemiological studies, rendering a simple answer to this question impossible.

5.9.2 Extended answer

Studies of the short-term effects of $PM_{2.5}$ including for example the UK West Midlands study (Anderson *et al*., 2001) and the study by Sarnat *et al.* (2008) which have included chemical markers of combustion or traffic sources (*e.g.* black smoke or elemental carbon) and of secondary particles (*e.g.* sulphate) have typically found associations of adverse health outcomes with both chemical metrics. This suggests that both primary and secondary particles are of appreciable toxicity. There is, however, a conundrum highlighted by Grahame and Schlesinger (2005) that, while sulphate regularly appears as an apparent causal factor in epidemiological studies, toxicological studies with pure sulphate suggests that they pose little health risk. An explanation may lie in the results of studies such as Laden *et al.* (2000), who associated toxicity with a component that they associated with coal combustion sources which was rich in sulphate but also a number of trace metals. As mentioned above, Chen and Lippmann (2009), using both toxicological and epidemiological approaches, attach particular importance to nickel and vanadium which are emitted together with sulphur dioxide (the precursor of sulphate) in fuel oil combustion. Grahame and Schlesinger (2005) point out that sulphate might also act as a surrogate for the possible effects of secondary organic aerosols that might be the product of acid catalysis due to sulphur dioxide oxidation products. The other major inorganic secondary component, nitrate, is not, however, generally indicated as having high toxicity. Consequently, it seems very plausible that secondary components, or at least inorganic secondary components are of lower toxicity than some primary components of $PM_{2.5}$ but the evidence is as yet inconclusive. This point was considered in depth by COMEAP (2009). Reiss *et al.* (2007) also reviewed the available literature, highlighting the considerable knowledge gaps relating to sulphate, and (especially) nitrate. They conclude that for both species, evidence for a causal link with adverse health outcomes is weak.

5.10 Is the Toxicity of $PM_{2.5}$ Enhanced by the Presence of Other Pollutants?

5.10.1 Summary answer

Few epidemiological studies have addressed interactions of PM with other pollutants. Some toxicological and human exposure studies have shown additive and in some cases synergistic effects, especially for combinations of particles and ozone, and of particles and allergens.

5.10.2 Extended answer

Synergistic and antagonistic interactions are difficult to estimate in epidemiological studies because large studies are required for statistical confidence, and these do not exist for $PM_{2.5}$. The large pan-European APHEA2 study found that PM_{10} effects on mortality were stronger in areas with high NO_2 (Katsouyanni *et al.*, 2001), but this finding, although pointing to positive interaction, has also been interpreted as PM_{10} containing more harmful components in areas with high NO_2 than in areas with low NO_2.

Evidence of synergy is clearer from experimental studies, especially for interactions between PM and O_3. Ozone has been found to increase lung permeability in both animals and human, as well as to increase bronchial hyper-responsiveness (WHO, 2003). This may be expected to lead to synergistic effect in co-exposures to PM with O_3, as demonstrated in a number of animal studies for diesel exhaust particles (Madden *et al.*, 2000) and model ultrafine particles (Oberdorster *et al.*, 1995; Elder *et al.*, 2000). Interactions of particles and allergens have been studied in controlled human and animal exposure studies. Adjuvant effects of diesel exhaust particles have been demonstrated in mice (Granum *et al.*, 2001) and humans (Diaz-Sanchez *et al.*, 1999).

It is also possible that some pollutant interactions might be adaptive. For example, pre-exposure to ozone could up-regulate antioxidant enzymes and thus partially protect against oxidative injury elicited by particles; or chronic exposure to SO_2 could increase mucous secretion and airway narrowing potentially providing a thicker protective mucous barrier and more central particle deposition and more rapid particle clearance (WHO, 2003).

5.11 What are the Health Benefits of Abatement of $PM_{2.5}$?

5.11.1 Summary answer

The benefits are calculable from the results of the epidemiological studies, but the best evidence of benefits comes from the ‘intervention’ studies which demonstrate tangible reductions in morbidity and mortality following actual decreases in ambient levels of $PM_{2.5}$.

5.11.2 Extended answer

Using the risk factors in the research papers on short-term and long-term effects on $PM_{2.5}$ exposure on health, it is possible to calculate the benefits of a reduction in $PM_{2.5}$ exposures. For example, using the COMEAP (2009) all-cause mortality health risk factor of 6% per 10 $\mu g/m^3$ long-term exposure to $PM_{2.5}$, the UK IGCB estimate 6.4 – 12.2 million life years saved in the UK for the predicted decrease in $PM_{2.5}$ levels between 2005 and 2020 estimated to arise from implementation of pollution control measures already in place (IGCB, 2006). These figures are relative to those given in the answer to the question in Section 5.1 above. As before, the range in estimated life years saved reflects assumptions about extent of lag between exposure and health impact. The view of COMEAP is that the greatest health benefit from $PM_{2.5}$ concentration reduction will occur in the first few years after the reduction.

The above estimates are, however, rather theoretical, and there has been much interest in ‘intervention’ studies in which the benefits of genuine reductions in atmospheric concentration are evaluated. Clancy *et al.* (2002) examined the reduction in death rates after a ban on coal sales in Dublin, finding that both respiratory and cardiovascular standardised death rates fell coincident with the ban on coal sales. About 116 fewer respiratory deaths and 243 fewer cardiovascular deaths were seen per year in Dublin after the ban. Hedley *et al.* (2002) carried out a study of the effect of a reduction in the sulphur content of fuel oil in Hong Kong. They found a significant decline in the annual average trend in deaths from respiratory and cardiovascular diseases, but not from other causes. The average gain in life expectancy per year of exposure to the lower pollutant concentrations was 20 days (females) to 41 days (males). They made no attempt to disaggregate the beneficial impacts of a reduction in sulphur dioxide from that of a reduction in primary and secondary sulphate and the associated trace metal emissions from fuel oil combustion. In a follow-up of the Harvard Six Cities Study, Laden *et al.* (2006) compared death rates in the Six Cities Study cohort for eight years in a period of reduced air pollution concentrations. They found a highly significant reduction in overall mortality associated with decreased mean $PM_{2.5}$. Pope *et al.* (2009) investigated the changes in life expectancy accompanying the changes in fine PM air pollution in 51 US metropolitan areas over the ~20 year period of the 1980s and 1990s. After adjustment for other factors associated with change in life expectancy over the period they found a decrease of 10 $\mu g/m^3$ in concentration of $PM_{2.5}$ was significantly associated with an increase in mean life expectancy of 0.61 years.

The 2007 Air Quality Strategy (Defra, 2007) estimated the economic costs of the health impacts arising from exposure to anthropogenic PM (largely associated with $PM_{2.5}$) to be between £8.5 and £20.2 billion a year in 2005. The Environmental Audit Commission (2010) report notes that these costs are likely to be an under-estimate as no account was taken of NHS costs incurred by increased morbidity.

The cost-benefit analysis carried out at a European level to support the CAFE Directive (Holland *et al.,* 2005) estimated the annualised costs in 2020 for the proposed CAFE strategy to be €776 million compared with health benefits ranging from €5.9-16.8 billion, generating ratios of benefits/costs of between 7.6 and 21.7.

5.12 Which Sources Should be Controlled to Provide the Greatest Health Benefits?

5.12.1 Summary answer

There are two possible but linked approaches. If one assumes that particles from all sources are equally toxic, then the abatement measures which reduce the overall population-weighted exposures by the greatest margin are the most beneficial. If one takes into account possible differential toxicity of particles, it becomes necessary to carry out a more sophisticated study taking account not only of exposures but relative particle toxicity.

5.12.2 Extended answer

The emphasis in European law upon meeting limit values has led to an approach of controlling pollution in local hot-spots. However, the recent CAFE Directive includes both a limit value and an exposure-reduction approach for PM. The latter involves widespread measures which may reduce pollutant concentrations by only a small margin, but provides substantial public health benefit because of the large numbers of people receiving a reduced exposure. De Leeuw and Horalek (2009) compared sensitivity cases in which the limit value was met everywhere or the exposure-reduction target had been met by all countries. They concluded that the exposure-reduction approach results in a larger reduction in the burden of disease than meeting the limit values. If relative toxicity is taken into account, then reduction of pollution from traffic sources may prove especially beneficial. This is demonstrated by Tainio *et al.* (2010) in a sensitivity study of the cumulative health impacts of different sources of primary $PM_{2.5}$ in Finland, using ‘expert elicitation’ of potential differential $PM_{2.5}$ toxicity. However, a full cost benefit analysis is the preferred way of ranking abatement policies.

6. WHAT LEGISLATION IS USED TO CONTROL EXPOSURE TO $PM_{2.5}$

This Section summarises the various legislative controls at the international, European, national and local scales to control emissions of $PM_{2.5}$ from various sectors. Whilst the focus is upon the control of primary $PM_{2.5}$ emissions, consideration must also be given to the important role that emissions of precursor pollutants have in the formation of secondary organic and inorganic particles. Nitrogen oxides (NO_x), sulphur dioxide (SO_2), NMVOCs and ammonia (NH_3) all act as precursors to the formation of secondary PM (AQEG, 2005). A description of the legislative controls for these precursor pollutants is thus also included.

It is important to note that legislation to control emissions of PM does not specifically focus on $PM_{2.5}$, and emissions standards for both industry and transport are expressed in terms of total PM or 'dust'. Nonetheless, controls to limit total PM will also reduce emissions of $PM_{2.5}$, and in many cases PM emissions will be predominantly within the $PM_{2.5}$ fraction (*e.g.* tailpipe emissions from road vehicles).

6.1 How are Emissions from the Industrial Sector Controlled?

6.1.1 What International legislation is used to control emissions to air?

The United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) provides a policy framework to tackle air pollution problems that require international interventions. The Protocol to Abate Acidification, Eutrophication and Ground Level Ozone (the 'Multi-Effect' or 'Gothenburg' Protocol), which has been ratified by both the European Commission and the UK, sets national emissions ceilings for sulphur dioxide, nitrogen oxides, VOCs and ammonia. The requirements of the Protocol have been largely incorporated by the European Commission into the National Emissions Ceilings Directive (see below). The Protocol plays an important role in reducing important precursor emissions of both secondary organic and inorganic aerosols. The revision to the Protocol is currently being developed and it is expected that it will be extended to cover primary $PM_{2.5}$ emissions.

The Convention's Protocol on Heavy Metals (the 'Aarhus Protocol') sets emission limits for cadmium, lead and mercury. Whilst the Protocol does not specifically target PM, it suggests best available techniques (BAT) for industrial sources, such as filters and scrubbers, which will also be effective in reducing $PM_{2.5}$ emissions.

Emissions from shipping are controlled under the International Convention for the Prevention of Pollution from Ships (MARPOL). Annex VI to the MARPOL Convention came into force in 2005. The Annex imposes limits on the maximum sulphur content of bunker fuel, and sets NO_x emissions limits for diesel engines. Revisions to Annex VI (IMO, 2008), which come into force on 1 July 2010, set out increasingly stringent controls on both SO_2 and NO_x emissions up to 2020.

Agreed standards for aircraft engine emissions are published by the International Civil Aviation Organisation (ICAO) through the Committee on Aviation Environmental Protection (CAEP). Emissions standards for many current aircraft in service were agreed at the CAEP/4 meeting in 1998; new engines, certified after 31 December 2007 have to meet the more stringent CAEP/6 standards (CAEP, 2004).

6.1.2 What European legislation is used to control emissions to air?

The European Commission formally adopted the Clean Air for Europe (CAFÉ) programme in May 2001. This programme is intended to integrate the various components of air quality polices within the 6th Environmental Action Programme (EAP), and specifically to provide a Thematic Strategy within which various measures such as the National Emissions Ceilings Directive, the AutoOil Programme, and the ambient air Directives could be coordinated. The Thematic Strategy was published in 2005 and provides the framework for air quality control in Europe.

National Emission Ceilings Directive

The National Emission Ceilings Directive (NECD) (2001/81/EC) sets national emissions ceilings for four pollutants (sulphur dioxide, nitrogen oxides, VOCs and ammonia). The current Directive requires Member States to draw up programmes for reducing emissions from all sources (energy, transport (excluding aviation and shipping), industry and agriculture) up until the end of 2010, and to submit annual inventories.

A revision of the NECD forms part of the implementation of the Thematic Strategy, and proposals are still under consideration. The revision will set new emissions ceilings for 2020 for the four pollutants already regulated, and for primary emissions of $PM_{2.5}$.

Integrated Pollution Prevention and Control (IPPC) Directive

The *IPPC Directive* (2008/1/EC) is intended to minimise pollutant emissions from various industrial installations throughout the European Union. Operators of installations that fall under the Directive (energy, metals production and processing, minerals, chemical, waste management, and other smaller sectors including paper, tanneries and poultry farms) must obtain an environmental permit from the appropriate competent authority (regulator). Emissions limits are fixed for dust, and other PM precursor pollutants such as nitrogen oxides, sulphur dioxide, VOCs and heavy metals.

Operators must demonstrate that they are using best available techniques (BAT) to prevent or reduce pollution. Requirements above BAT must be imposed where the installation causes an exceedence of a quality standard (*e.g.* a breach of the air quality limit values). BAT Reference (BREF) notes are produced by the European IPPC Bureau to help identify best available techniques for different industrial processes (http://eippcb.jrc.es)

Solvent Emissions Directive (SED)

The *Solvent Emissions Directive* (1999/13/EC) is intended to limit emissions of VOCs arising from the use of organic solvents in a range of activities, including printing, surface cleaning, vehicle coating, dry cleaning and manufacture of footwear, and pharmaceutical products. The Directive requires relevant installations to comply with emissions limit values or with the requirements of the ‘reduction scheme’.

VOC Paints Directive

Directive 2004/42/EC limits emissions of VOCs from the use of organic solvents in paints and varnishes.

Large Combustion Plant (LCP) Directive

The *Large Combustion Plant Directive* (2001/80/EC) controls emissions of nitrogen oxides, sulphur dioxide and dust from large combustion plant (those with a rated thermal input of 50 MWth, or above. Installations licensed in the UK before 1 July 1987 were given three options; to meet the emissions limit values, to participate in the UK National Emissions Reduction Plan (NERP) (a trading scheme for mass emission based allowances, or accept a limited hours derogation. New installations are required to meet the emissions limit values as set out in the Directive or in accordance with the relevant PPC Regulations.

Continuous measurements of dust, SO_2 and NO_x are normally required to demonstrate compliance with the emissions limits; where they are not, discontinuous measurements are required on a routine (*e.g.* 6-monthly) basis. As an alternative, appropriate determination procedures, which must be verified and approved by the competent authority, may be used to evaluate performance. Measurements and/or other evaluation procedures must be based on European Committee for Standardisation (CEN) standards (where available) or suitable International Standards Organisation (ISO), international or national standards as appropriate.

Waste Incineration Directive (WID)

The Directive (2000/76/EC) on waste incineration covers any mobile or stationary plant that are used for the thermal treatment of waste, with or without heat recovery mechanisms. It covers both hazardous and non-hazardous wastes and sets emissions limits for dust, nitrogen oxides, sulphur dioxide, and heavy metals.

As with the LCP Directive (see above), measurement regimes are required to demonstrate compliance with the emissions limits.

Proposed Industrial Emissions Directive (IE(IPPC)D)

A review of the IPPC Directive was undertaken by the Commission between 2005 and 2007, with the objective of improving its' functionality and effectiveness, and how it links to other legislation related to industrial emissions. The review concluded with a proposal for a new Industrial Emissions Directive (the 'IE(IPPC)D'), which is intended to harmonise and consolidate the existing IPPC, SEC, WID and LPC Directives. The Proposal recasts[12] seven existing Directives into a single, legislative document. The Proposal is still moving through the co-decision process, with an anticipated entry into force at the beginning of 2011, and transposition by mid-2012.

6.1.3 What National legislation is used to control emissions to air?

National legislation to control emissions to air from industrial processes operates differently in England and Wales, compared with Scotland and Northern Ireland. The enabling legislation is the *Pollution Prevention and Control Act 1999*, which implements the IPPC Directive in England, Scotland and Wales, and the *Environment* (*Northern Ireland) Order 2002*, which fulfils the same function in Northern Ireland.

The *Environmental Permitting* (*England and Wales) Regulations 2007* (EPR) bring together a single set of rules covering IPPC and waste management planning. The Environment Agency acts as the regulator for Part A1 installations and most waste

---

[12] The term "recasting" infers that substantive changes and amendments are made to the original acts. The new act is subject to the full legislative process and then repeals all of the acts that have been recast.

operations. The regulation of Part A2 and Part B installations is carried out by local authorities.

In Scotland, the *Pollution Prevention and Control* (*Scotland) Regulations 2000* apply to all Part A (IPPC) and Part B (LAPC) installations, with the Scottish Environment Protection Agency acting as the regulating authority. In Northern Ireland, the Environment Agency Northern Ireland regulates both Part A and Part B installations under the *Pollution Prevention and Control* (*Northern Ireland) Regulations 2003.* Part C installations, for local air pollution control, are regulated by the district councils.

All installations operating under the above regimes must obtain a permit from the appropriate Regulator. In granting the permit, the Regulator may include conditions with the aim of achieving a high level of environmental protection, taking into account Best Available Techniques. Any such conditions must ensure compliance with relevant Directive requirements, including achievement of the ambient air limit values (see below).

Local authorities are also responsible for the regulation and control of local air pollution under the *Clean Air Acts*. Sections of the Acts are related to:

- the prohibition of dark smoke from chimneys;
- the control of smoke, grit dust and fumes from furnaces (including chimney heights);
- the designation of smoke control areas; and
- the requirement to approve the chimney height for a furnace of more than 0.366MWt.

Fugitive Dusts

Fugitive dust emissions are generally less well regulated. They may be controlled by legislation related to statutory nuisance. Section 79(1) of the Environmental Protection Act defines statutory nuisances including:
Any dust, steam, smell or other effluvia arising on industrial, trade or business premises and being prejudicial to health or a nuisance.

The Environment Agency in England and Wales, the Scottish Environment Protection Agency in Scotland and the Environment Agency Northern Ireland have responsibility for regulating fugitive emissions from industrial and waste management sites and from regulated pig and poultry farms. This can be through the conditions applied during the permitting process.

Local authority planning departments control fugitive dust emissions from construction sites thought the imposition of conditions as part of the planning process.

6.2 How are Emissions from the Transport Sector Controlled?

Emission standards for various pollutants are defined for most applications of internal combustion engines, including:

- Road transport
  - Heavy-duty vehicles.
  - Motorcycles.
- Non-road mobile machinery (NRMM)
  - Construction equipment (bulldozers, excavators, road rollers, cranes, *etc*.).
  - Fork-lift trucks, airport ground-support equipment, combine harvesters, snow-ploughs.
  - Machinery that uses constant-speed engines, such as compressors, generators, sweepers and refrigeration units.
  - Marine diesel engines.
  - Inland waterway vessels.
  - Engines for recreational boats and watercraft such as jet-skis.

- Railway locomotives and railcars.
- Small equipment such as lawn mowers and chain saws.
- Engines for agricultural and forestry tractors.

It is important to note that there are no standards that currently limit emissions from brake and tyre wear, or from the 'resuspension' of particles from the road surface. As exhaust emissions are subject to increasingly stringent controls, then the proportional contribution of these 'non-exhaust' PM sources will become more important.

6.2.1 How are road transport emissions controlled?

Pollutants are emitted from road vehicles as a result of fuel combustion and other processes. 'Hot' exhaust emissions of many different pollutants are produced by a vehicle when its engine and exhaust after-treatment system are at their normal operational temperatures. The exhaust emissions produced during the vehicle warm-up phase are often referred to as 'cold-start' emissions. Evaporation from petrol vehicle fuel systems makes a significant contribution to emissions of volatile organic compounds (VOCs).

The regulation of emissions from road vehicles takes three main forms:

- **Type approval legislation:** In the regulation of exhaust particles, practically all worldwide emission standards are expressed in terms of total PM - as determined by gravimetric methods. In some cases diesel smoke opacity is measured using optical methods, and particle number emissions are regulated in the latest European legislation. There is no differentiation by particle size, and so no legislation addresses $PM_{2.5}$ specifically. Mass-based standards are also stated in type approval emission legislation for NO_x and NMVOCs. In addition, the latest European legislation stipulates an exhaust concentration limit for NH_3 applicable to heavy-duty diesel engines.
- **In-service inspection:** In-service inspection represents a further level of legislative control by which authorities can ensure that vehicles conform to regulations when in use. However, whilst type approval checks are relatively detailed and involve the use of specialist and expensive laboratory equipment, a lower level of sophistication applies to the in-service inspection test.
- **Fuel legislation (indirectly):** The emissions from a particular vehicle are affected by the properties of the fuel on which it is running. Engine and vehicle technologies normally achieve their best emissions performance with higher category fuels (ACEA *et al*., 2006). One property on which a great deal of attention has focused is the sulphur content. Fuel sulphur has an adverse effect on emissions due to catalyst inhibition, as it competes strongly with exhaust pollutants for space on the active catalyst surface, and also adversely affects exhaust gas oxygen sensors. The introduction of 'sulphur-free' fuels should enable advanced engine and exhaust after-treatment technologies - such as lean burn GDI vehicles, particle traps and regenerative NO_x storage systems - to meet the increasingly stringent exhaust emissions regulations for road vehicles. Reductions in fuel sulphur also generally lead to reductions in PM emissions (ACEA *et al*., 2006) as well as a proportional reduction in SO_2 emissions.

These three approaches are described in more detail below.

Type approval legislation

Light-duty vehicles

Since the early 1970s in Europe, emission limits have applied to road vehicles and engines, and the methods of measurement have been standardised. The emission limits apply to total hydrocarbons (THC), NO_x and PM from the vehicle exhaust, as well as evaporative emissions of VOCs.

All Member States are subject to the emission limits for road vehicles and engines, and methods of measurement are standardised. In the UK the Vehicle Certification Agency (VCA) is the designated Approval Authority and Technical Service for type approval to all automotive European Community (EC) Directives and the equivalent United Nations Economic Community for Europe (UN/ECE) Regulations.

The emission regulations for new light-duty vehicles - passenger cars (M1, M2) and light commercial vehicles (N1, N2) - were formerly specified in Directive 70/220/EEC. This Directive was amended a number of times, most notably to introduce new emission standards. The emission standards for passenger cars are shown in Appendix 2 (the emission standards for light commercial vehicles are not shown).

The last amendment, Directive 2002/80/EC addressed Euro 3/4 vehicles. Subsequent changes have been introduced in the form of EC Regulations. Whereas Directives had to be transposed into the national legislation of each Member State, in Regulations the detailed technical provisions are directly applicable to manufacturers, approval authorities and technical services, and they can be updated in a more efficient manner than Directives. It is intended that the Regulations will consolidate all previous Directives, which will then be repealed. Regulation (EC) No. 715/2007 introduced the Euro 5 and Euro 6 emission requirements (with compliance required by 2009 and 2014 respectively). This was followed in 2008 by Regulation (EC) No. 692/2008 addressing the implementation and amendment of 715/2007.

Attention generally focuses on the exhaust emissions test, as this addresses the largest contribution to emissions during the lifetime of a vehicle. In this test emissions are measured over the New European Driving Cycle (NEDC), which is composed of an Urban Driving Cycle (UDC), and an Extra-Urban Driving Cycle (EUDC). The UDC is characterised by low vehicle speed, low engine load, and low exhaust gas temperature. The EUDC includes more aggressive driving modes and higher loads.

Heavy-duty vehicles

The European emission standards for heavy-duty vehicles (HDVs) apply to all motor vehicles with a ‘technically permissible maximum laden mass’ of more than 3,500 kg, equipped with compression ignition, positive ignition natural gas or liquefied petroleum gas engines. The responsibility for compliance with emissions regulation is borne by the engine manufacturer.

The first Directive applicable to HDV diesel engines was a restriction on visible smoke (Directive 72/306/EEC). This was determined using a ‘free acceleration’ smoke test. Limits on mass emissions of gaseous pollutants were introduced by Directive 88/77/EEC, which set standards for total hydrocarbons (THC) and NO_x based on the ECE-R49 test.

In October 2000, for the certification of Euro III engines, the European Stationary Cycle (ESC), European Transient Cycle (ETC) and the European Load Response (ELR) test were introduced. The ESC replaced the ECE-R49 test. Different driving conditions are represented by three parts of the ETC cycle. Part one represents city driving with a maximum speed of 50 km/h, and includes frequent starts, stops, and idling periods. Part two, representing rural driving, begins with a steep acceleration segment and has an average speed of 72 km/h. Part three represents motorway driving with an average speed of 88 km/h. The ELR engine test, which consists of a sequence of load steps at constant engine speeds, was introduced for the purpose of smoke opacity measurement.

The emission standards for heavy-duty diesel engines are shown in Appendix 2. Separate limit values are applicable to the ETC (not shown).

For the Euro VI stage the legislation has been simplified and, as with passenger cars, there has been a transition from Directives to Regulations. The Euro VI emission standards were introduced by Regulation 595/2009 in July 2009 (with a correction in July 2009). The implementing Regulation is due to be introduced in 2010.

EU Member States are also allowed to use financial incentives in order to speed up the marketing of vehicles meeting new standards ahead of the regulatory deadlines. Euro VI incentives can also be given for scrapping existing vehicles or retrofitting them with emission controls in order to meet Euro VI limits.

An inadvertent side effect of tightening legislation for PM emissions has been that some of the technologies employed have resulted in much higher ratios of NO_2 to NO_x in the exhaust. This has led to higher roadside NO_2 concentrations than might otherwise have been the case (AQEG, 2007). It has arisen because primary NO_2 emissions are not currently regulated.

Two-wheel vehicles

There have been fewer changes in the legislation relating to two-wheel vehicles (Appendix 2). There are no emission limits for PM in the legislation. Stage 1 (‘Euro 1’) of Directive 97/24/EC, which became effective in 1999, introduced more stringent limits than the existing ECE R40 Regulation. In 2003, stage 2 (Euro 2) of 97/24/EC entered into force. This reduced the limits again without changing the type approval test cycle. For 2006 (Euro 3) the emission limits are lower still, and the type approval test cycle has been changed.

Future directions

In relation to the Euro 6/VI standards, the implementing Regulations for heavy-duty engines are due to be published in 2010, and a particle number emission limit for light-duty petrol vehicles is to be defined by 1 September 2014.

The future of European emission legislation post-Euro 6/VI is somewhat uncertain. In legislative terms, the emphasis at present appears to be on definition of appropriate procedures for the type approval of hybrid-electric, full electric vehicles and hydrogen vehicles.

In-service inspection

European legislation

Directive 96/96/EC of 20 December 1996 mandates all Member States to carry out periodic safety and emission inspections for most types of motor vehicle. It also sets minimum requirements for inspections and their intervals - for light commercial vehicles (up to 3,500 kg) and private cars (up to 8 seats) the first inspection shall occur no later than four years, and at a frequency of no more than two years thereafter. All other types of vehicles are subject to yearly inspection (buses, vans, trucks, trailers, taxis, ambulances, coaches).

UK legislation

Under the UK Motor Vehicle (Tests) Regulations (1981), light-duty motor vehicles are subject to an annual roadworthiness inspection (MoT), including an exhaust emissions test. This inspection is compulsory for all vehicles older than three years. As noted earlier, in-service tests are based on simplified operations of the vehicle (the engine is not placed under load) and relatively low-cost equipment. For such reasons PM and NO_x are not included in the test.

Fuel legislation

The quality of automotive fuels in the European Union is specified by standards developed by the European Standards Organisation (CEN). The standards are periodically updated to reflect changes in specifications, such as the mandatory reductions in sulphur content.

The sulphur content of fuel in the European Union has reduced considerably in recent years. For diesel fuel a maximum sulphur limit of 50 parts per million (ppm) – known as 'ultra-low sulphur' (ULS) - was introduced on 1 January 2005 by Directive 2003/17/EC. The UK introduced this earlier. However, although small improvements in emissions have been achieved by reducing sulphur to these low levels, several emission-control technologies are intolerant to sulphur and require that levels are reduced even further. Directive 2003/17/EC therefore also required 'sulphur-free' petrol and diesel fuels - with a limit of 10 ppm - to be available by 1 January 2005. All UK road diesel has had a sulphur level of less than 50 ppm since 1999 (since 2001 for petrol), and since 1 January 2009 all road fuel has had less than 10 ppm sulphur.

6.2.2 How are non-road mobile machinery sources controlled?

European and UK legislation

Directive 97/68/EC - Emission of Gaseous and Particulate Pollutants from Internal Combustion Engines to be installed in Non-Road Mobile Machinery (NRMM) - was adopted in 1997. The regulations were introduced in two stages: Stage I, which was implemented in 1999, and Stage II which was implemented between 2001 and 2004, depending on the engine power output. The Directive was implemented into UK law by the Non-Road Mobile Machinery (Emission of Gaseous and Particulate Pollutants) Regulations 1999. Directive 2002/88/EC amended Directive 97/68/EC by adding emission standards for small, petrol-fuelled utility engines below 19 kW.

The equipment covered by Stages I and II included industrial drilling rigs, compressors, construction wheel loaders, bulldozers, non-road trucks, highway excavators, forklift trucks, road maintenance equipment, snow ploughs, ground support equipment in airports, aerial lifts and mobile cranes. Agricultural and forestry tractors were subject to the same emission standards but different implementation dates. Engines used in ships, railway locomotives, aircraft, and generating sets were not covered by the Stage I/II standards.

The Stage III and IV emission standards were adopted in 2004 (Directive 2004/26/EC), and for agricultural and forestry tractors in 2005 (Directive 2005/13/EC). The Stage III standards are phased-in from 2006 to 2013, and Stage IV enters into force in 2014. The Stage III/IV standards do cover railway locomotive engines and engines used for inland waterway vessels. The Stage III/IV legislation applies only to new vehicles and equipment. Replacement engines (except for railcars, locomotives and inland waterway vessels) should comply with the original limit values for the engine. The Stage IIIA standards also cover engines used in inland waterway vessels.

International standards for railway locomotives

Emission standards for diesel engines used in railway locomotives have been established by the International Union of Railways (Union Internationale des Chemins de Fer, UIC), an association of European railway companies (UIC, 2006). The standards apply to all new engines used in new vehicles or for repowering of existing locomotives, and are binding to UIC members. The UIC Stage III standards are harmonised with the EU Stage IIIA standards for non-road engines.

6.3 What Legislation Exists to Control Ambient Concentrations of $PM_{2.5}$?

This Section summarises the legislation at the European, national and local scales with respect to concentrations of $PM_{2.5}$. Whilst the focus is upon $PM_{2.5}$, measures that are introduced to reduce PM_{10} concentrations will, in most case, also reduce $PM_{2.5}$. Consideration is therefore also given to legislation related to PM_{10}.

6.3.1 What role does European legislation play?

The *Ambient Air Quality and Cleaner Air Directive* (often referred to as the 'CAFÉ Directive') entered into force on 11 June 2008. The Directive consolidates the previous framework Directive and three Daughter Directives, and retains the existing limit values for PM_{10}. The Directive also strengthens the provisions that require Member States to implement plans and programmes to achieve the limit values, and provides for extensions to the deadlines for compliance where it can be proved that all reasonable steps have been taken. In the case of the PM_{10} limit values (that were to have been attained by 2005), an extension to the deadline may be permitted up until 11 June 2011. The UK is seeking such a Time Extension Notice (TEN) for PM_{10}.

The Directive also introduces new obligations for Member States with regard to $PM_{2.5}$. This includes a limit value (25 $\mu g/m^3$) and an exposure concentration obligation (20 $\mu g/m^3$), to be achieved by 2015, and a national exposure-reduction target of up to 20% reduction from 2010 concentrations by 2020[13].

Article 20 of the Directive allows Member States to subtract the contribution of natural sources from any reported exceedences. Within its Time Extension Notice to the Commission, the UK has therefore subtracted the sea salt component from the data presented. However, it is important to note that Defra has advised that the subtraction of the sea salt component is not to be widely applied in other applications *e.g.* compliance of industrial emissions with the limit values cannot subtract natural sources from measured or modelled concentrations.

6.3.2 What role does the UK's Air Quality Strategy play?

The *Air Quality Strategy for England, Scotland, Wales and Northern Ireland* (Defra, 2007) sets out the air quality objectives and policies of the UK Government and the Devolved Administrations. The Strategy sets out air quality objectives for both PM_{10} and $PM_{2.5}$. For England, Wales and Northern Ireland, these are effectively the same as the limit values, targets and obligations defined in the Directive. The Scottish Government has set more challenging targets for both PM_{10} and $PM_{2.5}$ (as detailed in Section 1, Table 1)

6.3.3 What role does Local Air Quality Management play?

The *Environment Act 1995* and the *Environment* (*Northern Ireland) Order 2002* introduced the system of Local Air Quality Management (LAQM). This requires local authorities to periodically review and assess air quality in their areas, and determine whether the air quality objectives, as prescribed in Regulations, are likely to be achieved. Where it is concluded that the objectives will not be met, the authority must declare an Air Quality Management Area (AQMA) and prepare an Air Quality Action Plan setting out the measures it intends to take in pursuit of the objectives. There is currently no statutory obligation on authorities to achieve the objectives, and there is currently no obligation related to $PM_{2.5}$.

---

[13] The exposure concentration obligation and the exposure-reduction target are based on the Average Exposure Indicator (AEI) which is a three calendar year running annual mean averaged across urban background monitoring stations in the UK. The exposure-reduction target will vary from country to country depending on the absolute concentrations.

6.3.4 What is the basis of the new exposure-reduction standard for $PM_{2.5}$?

The need for a new exposure-reduction approach for PM has been widely recognised and supported. This need arose because there is no known threshold for exposure to PM below which health effects do not occur. The limit value (and objective) approach focuses control of exposure at hotspots (where exceedences of the limit values occur), requiring no additional measures to reduce exposure where concentrations are below the criteria. The European Commission (EC) Working Group on Particulate Matter (EC, 2004) concluded that "*while it is recognised that limit values have an important role to play in helping drive down exposure, the Working Group recommends that the Commission consider the use of alternative approaches, such as gap closure, emissions ceilings or targets, to supplement* (*but not replace) the use of limit values*". The Air Quality Expert Group (AQEG, 2005) supported this view stating that "*for pollutants such as PM, where there is no evidence of a threshold concentration below which health effects do not occur, a strategy based on reducing exposure to the largest population would seem to offer improved benefits to public health*".

Various approaches to exposure-reduction were investigated by Defra and used to advise the Commission (Laxen and Moorcroft, 2005). Variants based on modelling, and monitoring with implicit or explicit population weighting were investigated. Whilst modelling could easily generate population-weighted concentrations, it was concluded that this would be reliant on high quality emissions inventories for PM (which do not exist across Europe) and compliance checking would be difficult to enforce. It was thus concluded that any new exposure-reduction standard should be based on monitoring.

Consideration was also given to the explicit population-weighting of the measured PM concentrations[14]. It was concluded that it would be difficult to accurately define the population-weighted exposure represented by an individual monitoring station, and difficulties would arise if monitoring stations closed, or failed to meet acceptable data capture standards.

Important considerations were then:
- how to define exposure;
- whether to subtract a natural or secondary component; and
- how to determine compliance,

and these are considered in more detail below.

Defining exposure: The exposure-reduction approach was aimed at reducing the overall exposure of the population to PM. The focus was therefore on the highly populated areas, *i.e.* urban centres, where significant numbers would be exposed to higher concentrations. The most practical and simple option identified to define this overall exposure to PM concentrations was to use annual mean concentrations averaged across a carefully specified set of urban background monitoring stations. It was also recognised that the uncertainty in defining the UK average reduces with an increasing number of monitoring stations. It was therefore recommended that an approach based on one sampling point per million inhabitants summed over agglomerations with a population greater than 100,000 should be adopted. This was consistent with the Environmental Noise Directive, which already defined the agglomerations having a population of over 100,000.

Subtraction of rural or secondary background: A number of scientific benefits were associated with the subtraction of either the rural or secondary background component, in that it would provide a measure of the concentration that more closely represents the proportion of PM that is locally controllable, and would produce a higher figure for the percentage reduction. This option was, however, discounted on the basis that it would

---

[14] This explicitly weights the measured PM concentrations according to an estimate of the population exposed to that concentration.

require a substantial increase in the number of rural sites, and would lead to increased uncertainty in the average exposure concentration (unless more urban sites were included). It was also considered that the secondary component (predominantly sulphates and nitrates) is best controlled via the National Emissions Ceilings.

How to determine compliance: In order to determine compliance with the exposure-reduction target it was necessary to set the Base Year and Compliance Year, and to take account of varying meteorological conditions. The latter was extremely important, as the measured change in PM concentrations would not accurately reflect the long-term change in PM exposure if the Base (or Compliance) Year(s) was an unusually high or low PM pollution year(s). A solution to this problem was to define compliance on the basis of a running-average PM concentration. The greater the number of years used to determine the running average, the less any particular year influences the result, but for practical reasons a three year running average was considered to offer the best option.

The exposure-reduction approach as set out in the Ambient Air Quality Directive (2008/50/EC) is based on an Average Exposure Indicator (AEI). The AEI is based on measurements of $PM_{2.5}$ concentrations in urban background agglomerations and zones with populations greater than 100,000 people, and is assessed as a three-calendar year running annual mean averaged over all of the sampling points. In the UK this 3-year average AEI will be for the years 2009-2011. A single 3-year AEI will be developed for each Member State. Reduction targets are then set for the AEI over a 10-year period, with the target being more challenging the higher the initial AEI (Table 14).

Table 14 Exposure-reduction target for $PM_{2.5}$ as a function of initial AEI.

Initial AEI (µg/m^3)	Target reduction 2010-2020
≤8.5	0%
8.5 – <13	10%
13 – <18	15%
18 - <22	20%
>22	All appropriate measures to achieve 18 µg/m^3

7. MONITORING FOR $PM_{2.5}$

7.1 How are Concentrations of Airborne PM measured?

The basic principle for monitoring concentrations of airborne PM is to draw air through a pre-weighed filter for a known length of time and at a known flow rate, and then to re-weigh the filter to determine how much material has been collected. Although simple in principle, the practice of measuring PM concentrations has proved to be far from straightforward. All the measurements discussed here relate to the determination of PM as a concentration in mass per unit volume, in µg/m^3. It is also important to note that both the UK objectives and EU limit values define the sampling volume for the determination of both PM_{10} and $PM_{2.5}$ concentrations as that measured in ambient conditions in terms of temperature and atmospheric pressure[15].

In addition to measuring the PM concentration, there are a variety of techniques that can be used to speciate PM, for example to determine the metals, polyaromatic hydrocarbons (PAHs), major ions, and elemental/organic carbon (EC/OC) components. Measurements of speciated PM have traditionally been carried out by subsequent chemical analysis of the exposed filters, but an increasing number of direct-reading analysers are now available, which allow continuous measurements to be carried out. A detailed review of such techniques is beyond the scope of this report.

There is also some interest in the measurement of particle number concentrations. Such measurements focus on the ultrafine particles, which contribute very little to the particle mass (*e.g.* to the $PM_{2.5}$ concentration) but significantly to the total number of particles. A detailed review of such measurement techniques for particle number concentrations is not relevant to a report that is focused on $PM_{2.5}$ measured on a mass basis.

7.1.1 What are the difficulties associated with the measurement of airborne PM?

Airborne PM includes a very wide range of particle sizes, shapes, and different chemical constituents (hence densities). The shapes and densities will influence exactly which particles penetrated a size selective inlet. Also, some of these particles are semi-volatile (most notably ammonium nitrate and secondary organic compounds) and move between the particle and vapour phases under normal ambient conditions. Other particles are hygroscopic and attract water vapour during sampling and/or analysis.

Due to the complex nature of airborne PM, it is not possible to prepare standards that are traceable to first metrological principles in order to calibrate the instruments (for example in the way that gas standards can be prepared to calibrate sulphur dioxide instruments). For this reason, the method that is used for the collection and subsequent determination of the particle mass influences the ultimate particle mass concentration that is reported.

7.1.2 What monitoring methods are used for PM?

There are a variety of methods that are used to measure airborne concentrations of PM (see Table 15). The principal difference between the methods used for the determination PM_{10} and $PM_{2.5}$ concentrations is the size-selective inlet that is used to exclude the unwanted, larger particles, before the measurement is carried out[16].

---

[15] In contrast, for gaseous pollutants, the sampling volume is standardised at a temperature of 293K and an atmospheric pressure of 101.3kPA

[16] PM_{10} and $PM_{2.5}$ are defined in the CEN reference methods (EN 12341 and EN 14907) as particulate matter which passes through a size-selective inlet with a 50% efficiency cut off at 10 and 2.5 µm aerodynamic diameter, respectively.

There are, however, some inherent additional difficulties with measuring $PM_{2.5}$ concentrations, as opposed to PM_{10}.

- The particle mass that is collected is lower, and quantification will therefore be subject to greater uncertainty;
- The proportion of semi-volatile components is higher in the $PM_{2.5}$ fraction, and any losses will have a greater effect.

PM monitoring in the UK has historically been founded on the use of the Tapered Element Oscillating Microbalance (TEOM) analyser. This instrument uses an oscillating microbalance within the instrument to weigh the mass of PM collected, thus differing from the samplers that collect PM on filters that are returned to the laboratory for weighing. A variant of the TEOM, the Filter Dynamics Measurement System (FDMS) analyser, has now been introduced to replace the TEOM.

Table 15 Summary of PM concentration sampling and analysis methods (based on AQEG (2005)).

Method	Advantages	Disadvantages
Filter-based gravimetric samplers	The European reference sampler for PM_{10} and $PM_{2.5}$. Sequential samplers are available that allow up to 14 days monitoring without site attendance.	Considerable care needs to be taken with filter selection, storage and handling, and with QA/QC procedures for filter weighing. High operating costs. Time resolution of measurement limited to 24h, and results not available in real-time.
TEOM analysers	Provide continuous real-time data with < 1-hour time resolution.	Pre-heated air stream causes losses of semi-volatile components. Method demonstrated to not be equivalent to the European reference sampler (corrections can be applied to approximate reference concentrations using the Volatile Correction Model (VCM)). High capital cost.
FDMS analysers	Provide continuous real-time data with < 1-hour time resolution. Demonstrated to be equivalent to the reference method (certain versions only).	High capital cost. Experience has demonstrated that additional care (compared with the TEOM) needs to be taken with instrument operation and subsequent data ratification.
Beta-attenuation analysers	Provide continuous real-time data with < 1-hour time resolution. Some instruments (non-heated inlets) have been demonstrated to be equivalent to the European reference sampler.	If instruments with heated inlets are used, there may be substantial loss of semi-volatile particles.
Optical analysers	Ability to measure several size fractions simultaneously. Relatively lightweight, portable and may be battery operated.	Calculation of PM concentrations is based upon assumptions about particle characteristics which may vary from place to place and from time to time.
Black smoke	Relatively cheap. Provides a link to a long-term database of measurements. Provide an indication of carbon.	Provides 24-hour concentrations. Cannot be directly related to a particular size fraction or to a particular component of PM.

European Reference Methods

The reference methods for the determination of concentrations of PM_{10} and $PM_{2.5}$ for comparison with the limit values are described in European Standards EN12341 and EN14907 respectively (CEN, 1999 and 2005). The approaches are founded on filter-based gravimetric measurements, in which PM is sampled by drawing air through a filter. The filter is subsequently weighed in a laboratory, after conditioning at a specified temperature and humidity, to determine the PM mass. Any losses of semi-volatile PM, or any artefacts associated with particle-bound water vapour, are deemed to be zero by convention.

The reference methods for both PM_{10} and $PM_{2.5}$ are currently under revision by CEN. The intent is that the methods will be unified into a single standard, the only difference between PM_{10} and $PM_{2.5}$ being the specification of the size-selective inlet. CEN is also preparing an Automated Measurement Standard, which will address the use of continuous analysers for both PM_{10} and $PM_{2.5}$ monitoring.

Other Monitoring Methods

For a variety of practical reasons, the European reference samplers (or other filter-based gravimetric samplers) have not been widely used in the UK. Their use is labour-intensive (requiring pre- and post-sampling weighing of filters) and they only provide information on 24-hour mean PM concentrations. In addition, they cannot provide real-time information on PM concentrations to the public. The TEOM analyser was seen as a way around these limitations, providing real-time hourly mean data, however, it was soon recognised that this analyser generally recorded appreciably lower PM_{10} concentrations than the European reference sampler, due to the loss of semi-volatiles in the heated inlet. As an interim measure a default 'correction factor' of 1.3 was applied to the measured concentrations[17], whilst further studies were carried out. The use of a simple correction factors is no longer recommended for TEOM data (see Section 7.1.3).

The FDMS analyser is an airborne PM monitor based on the TEOM technology, but with the ability to measure both 'core' and volatile fractions of particles. The analyser draws ambient air through a size-selective inlet (PM_{10} or $PM_{2.5}$). This air then passes through a drier to remove water, before entering the sensor unit where the PM is collected onto a filter held at 30°C, and weighed. The analyser samples in this 'base cycle' mode for 6 minutes, during which there will be losses of volatile particles. The sample flow is then switched, so that it passes through a cooled chamber, held at 4°C, and then through a filter which removes all of the PM in the airstream. This cooled, scrubbed air is then returned to the sensor unit. During this 'reference' or 'purge' cycle (which also runs for 6 minutes) volatile particles will continue to evaporate from the sensor unit filter, such that the average PM concentration measured during the purge cycle will normally be negative (it can occasionally be positive if gases in the purge flow airstream are absorbed by the PM collected on the sensor filter). This reference, or purge cycle concentration provides an estimate for the volatile particle concentration that is being lost, which can then be added onto the base concentration to give an overall PM mass concentration. The total PM concentration for each 12 minute cycle is thus equal to the base concentration minus the purge concentration (the latter is normally a negative value and so the purge concentration is in effect added). It should be recognised that the loss of volatile particles occurs relatively slowly, thus the loss during a particular cycle will include volatile material collected in previous cycles. This is manifest as a time shift of a few hours in the purge concentration (see discussion in Section 7.5.1).

---

[17] For Scottish authorities, the Scottish Executive issued additional interim advice for local authorities in Scotland, based on local intercomparison tests. For the annual mean objectives, it was recommended that authorities should correct TEOM PM_{10} concentrations using both a 1.3 factor and a 1.14 factor.

The FDMS analyser has now been widely deployed into the UK networks to measure both PM_{10} and $PM_{2.5}$ concentrations. At the time of drafting this report, only the Model B/C FDMS analyser had been demonstrated to be 'reference equivalent' (see Section 7.1.3).

Other instruments in common use include beta attenuation analysers. These devices sample air onto a paper tape, and the reduction in the transmission of beta particles from the start to the end of the sampling period is recorded to determine the PM concentration. These instruments can have both heated or unheated inlets, which perform very differently (see below). The Met-One BAM (unheated) is used by a number of local authorities.

There are also a number of optical particle monitors that rely on the interaction between airborne particles and visible or infrared laser light. The instruments that utilise light scattering have the advantage that they can report concentrations for a range of particle sizes (total particles, PM_{10}, $PM_{2.5}$ and PM_1) and they are often portable (or semi-portable) and can be battery-operated; they are also relatively small and lightweight and can be attached to lampposts, fences, etc. The principal disadvantage is that they rely on a range of assumptions to calculate the PM mass concentration, the validity of which may vary both spatially and temporally. These optical monitors are often used for screening surveys, to supplement other monitoring programmes (*e.g.* FDMS or beta attenuation analysers), or to identify potential issues surrounding industrial operations (*e.g.* mineral extraction processes) or construction works.

There has been a long history of monitoring black smoke in the UK, with some monitoring sites still operational that date back to the early 1960s. Until 2008 black smoke was sampled using an 8-port sampler, which had an effective size cut off of around 4 µm. These samplers collected PM on filters over 24 hours, then used a reflectometer to measure the reduction in reflectance of the filter, which was converted to an equivalent black smoke concentration using standard calibration curves. Since 2008 the national network has changed over to the use of aethalometers. Aethalometers quantify 'black carbon' on filter samples based on the transmission of light through the filter. The sample passes a $PM_{2.5}$ cyclone and 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. A calibration to convert black carbon to black smoke is being developed (Quincey, 2007; 2010). Black smoke has also been used as a measure of exposure in epidemiological studies.

7.1.3 What methods are equivalent to the reference method?

Member States can use methods for the determination of concentrations of PM_{10} and $PM_{2.5}$ other than the reference method, provided they can demonstrate that they display a consistent relationship to the reference method (*i.e.* that they are 'equivalent'). Guidance on how to demonstrate equivalence has been published (EC Working Group, 2010). Equivalence is defined in relation to the 'data quality objectives' set out in Directive 2008/50EC. This defines the uncertainty for both PM_{10} and $PM_{2.5}$ as 25%[18].

In 2006, Defra and the Devolved Administrations published the results of a study to determine the equivalence of a range of instruments in common use in the UK (Harrison, 2006). The outcome of this study is summarised in Table 16.

---

[18] The stated uncertainty refers to the individual daily measurements averaged over the period considered by the limit value for a 95% confidence interval.

Table 16 Summary of UK equivalence tests. Source: Harrison (2006).

Instrument	Outcome of Test
TEOM (PM_{10})	Fails the equivalence criteria
FDMS 'Model B' (PM_{10})[1]	Meets the equivalence criteria
FDMS 'Model B' ($PM_{2.5}$)[1]	Meets the equivalence criteria
Partisol 2025 (PM_{10})[2]	Meets the equivalence criteria
OPSIS SM200 (PM_{10})[3]	Beta - Meets the equivalence criteria Mass - Meets the equivalence criteria with correction for slope and intercept
Met-One BAM (unheated) (PM_{10})	Meets the equivalence criteria with correction for slope

Notes:

1. The 'Model B' FDMS is no longer available. The UK networks currently use the 'Model B/C' FDMS which is a hybrid instrument incorporating the 'Model B' drier. The 'Model C' FDMS is undergoing equivalence trials.
2. The Partisol 2025 was operated with PTFE-coated glass fibre filters.
3. The OPSIS SM200 instrument can perform dual measurements by direct mass determination (by collecting PM onto a filter for weighing) and by beta-attenuation.

A series of equivalence tests is ongoing, coordinated by Defra and the Devolved Administrations, but with the field trials funded by the instrument manufacturers. This is being carried out as a joint TUV/MCERTS programme[19].

As part of the above study, the dual channel (PM_{10} and $PM_{2.5}$) beta-attenuation analyser manufactured by FAI Instruments (Model SWAM 5a)[20] has achieved both TUV and MCERTS certification which effectively means it is equivalent for use in the UK; it is expected that the $PM_{2.5}$ Smart BAM will achieve MCERTS certification in 2010 (Harrison, 2010).

It is important to note that the TEOM analyser failed the equivalence criteria, even when the 'default' correction factor of 1.3 was applied. A new approach to correcting TEOM data has been introduced involving the Volatile Correction Model (VCM) developed by King's College. The approach is based on the assumption that the volatile component of PM_{10} lost during the heated sampling with a standard TEOM is consistent across a defined geographical area, such that measurements of this component at one location may be used to correct measurements elsewhere. Thus, it is assumed that the volatile component is constant across a region, with the implication that local sources do not contribute to volatile PM. The approach uses the FDMS 'purge measurement' as an indicator of the volatile component that will have been lost by the TEOM. PM_{10} concentrations measured by a TEOM may be corrected to a concentration that is equivalent to the European reference sampler using the following equation:

$$\text{TEOM}_{\text{VCM}}\ \text{PM}_{10} = \text{TEOM PM}_{10} + (1.87 \times \text{Regional FDMS PM}_{10\ \text{purge}})$$

---

[19] TUV is a commercial organisation that provides certification for various international standards. The MCERTS programme is operated by the Environment Agency. Both TUV and MCERTS provide testing and certification of ambient air quality monitoring systems.

[20] **http://www.fai-instruments.it/english/interna.php/pagina=SWAM%20Dual%20Channel/id=2/show=sottocat1**

A VCM web portal (http://laqm1.defra.gov.uk/review/tools/vcm.php) is available which allows users to download geographically-specific correction factors to apply to TEOM PM_{10} measurements on either a 1-hour or 24-hour basis.

The VCM approach is recommended to local authorities and other users of TEOM analysers, but the method is not deemed to be 'equivalent' to EN12341, and it is not used within the UK networks. It is also important to note that it is not currently possible to correct $PM_{2.5}$ concentrations measured using the TEOM in this way.

7.1.4 Quality assurance / quality control procedures for PM monitoring?

QA/QC procedures that should be applied for PM monitoring have been previously documented[21] (Defra, 2009). In particular, the procedures that need to be applied if filter-based gravimetric sampling is carried out are very important. These include the selection of filter material, filter conditioning and weighing, and the use of field blanks.

There are a number of different filter types that can be used for sampling, including PTFE, quartz fibre, glass fibre, and PTFE-coated glass fibre. A study carried out to support the UK equivalence trials identified important differences between these filters, in terms of their friability, collection efficiency, water absorption characteristics, and static interference (Brown *et al.*, 2005). The study concluded that PTFE-coated glass fibre filters performed the best, and these have subsequently been used in all UK equivalence trials.

The procedures set out in both EN12341 and EN14907 require pre- and post-sampling conditioning of filters at 20±1°C and 50±5% relative humidity. However, experience has shown that some filter materials, and the sampled PM itself, exhibits very strong hysteresis effects, *i.e.* water vapour is absorbed from the atmosphere, but is then very hard to remove during filter conditioning at the specified requirements. The effect is that some types of filters may not stabilise mass for many days, or even weeks, at the required weighing room conditions. Further experimental work is currently being carried out by CEN to investigate these effects, which may be reflected in future revisions to the standards.

A variety of terminology is in current use as set out in Table 17.

---

[21] **Appendix 1, pages A1-39 to A1.41**

Table 17 Terminology for PM monitoring.

Term	Description
Reference method	Measurement methodology, which by definition gives the reference value of the determinand (in this case, either PM_{10} or $PM_{2.5}$) for comparison with the limit value. The reference methods are described in EN12341 and EN14907 respectively.
Equivalent method	A measurement method, other than the reference method, for which equivalence has been demonstrated, *i.e.* the method has been shown to conform to the data quality objectives as defined in Directive 2008/50/EC.
Gravimetric equivalent	A term that was commonly applied to TEOM concentrations that had been adjusted using the default factor of 1.3. These values are now known not to be 'reference equivalent'.
Reference equivalent	A term used to describe PM concentrations that have been measured using an Equivalent Method.
Purge concentration	Mass PM concentration obtained from an FDMS analyser at 30°C with the sample having passed through a dryer and a 4°C chilled filter. It is taken to represent the volatile fraction of the PM.
VCM correction	The volatile correction model takes the purge concentrations from sites within the FDMS monitoring network and uses these to estimate the volatile PM lost from the TEOM instruments.

7.1.5 What are the implications for equivalence tests of the lower PM objectives in Scotland?

Scotland has retained the lower 2010 objectives for PM_{10} (50 µg/m^3 to be exceeded no more than seven times per year, and 18 µg/m^3 as annual mean). It has also introduced a lower annual mean objective[22] for $PM_{2.5}$ of 12 µg/m^3. It is useful to consider whether the adoption of these more stringent objectives has any implications to the validity of the equivalence tests that have been carried out in respect of the limit values.

The tests for equivalence are applied to the daily mean measurements (the annual mean is calculated from the daily means). There are therefore no implications associated with the lower PM_{10} objectives in Scotland, as, whilst the permissible number of exceedence days is lower (*i.e.* seven, compared with 35 elsewhere in the UK), the limit value (50 µg/m^3) is unchanged.

For $PM_{2.5}$, there are however potential implications if the strict requirements of the equivalence tests were to be applied. In the absence of a daily mean limit value for $PM_{2.5}$, it is necessary to assume one (as the test is applied to daily mean measurements). The convention applied is to assume a daily mean limit value of 30 µg/m^3.

The uncertainty between the reference method and the candidate method is applied to the complete dataset, and to two datasets, obtained by splitting the whole dataset into above or below 50% of the limit value, *i.e.* for datasets above and below 15 µg/m^3. If the lower

---

[22] This objective has not yet been included in Regulations

annual mean objective in Scotland is considered, a daily mean objective of 15 µg/m^3 would be assumed[23], and the tests then applied to datasets above and below 7.5 µg/m^3.

A further, and potentially more significant issue, is that the calculation of the relative standard uncertainty (W_{CM}) of the candidate method, is a function of the assumed limit value, which is a denominator. The equation takes the form:

$$W^2_{CM} = u^2_{xi=L} / L^2$$

where, L = the limit value, and $u_{xi=L}$ is the uncertainty at the relevant PM limit value.

It is thus apparent that reducing the assumed limit value from 30 µg/m^3 to 15 µg/m^3 will substantially increase the calculated uncertainty, and makes the demonstration of equivalence increasingly more stringent.

This is likely to present different results and it would be expected to be harder to achieve equivalence for measurements in Scotland, due to the greater uncertainty at low concentrations. However, it is not necessary to demonstrate equivalence for measurements made in relation to the Scottish objectives. It will therefore be reasonable to use instruments that are equivalent in the UK context for measurements made in Scotland.

7.2 How are Measurements of $PM_{2.5}$ Stack Emissions Made?

There is currently no requirement to measure emissions of $PM_{2.5}$ from industrial installations. Permit requirements are set in terms of total dust or PM, with no requirement to size select, even for PM_{10}. The principal method for sampling total PM is to draw off air from the stack isokinetically and then collect the PM on a filter for subsequent weighing. Continuous measurement methods are also used, involving the use of opacity meters. However, it is understood that the concentrations derived in this way will be less than the total from filter-based samplers.

A standard reference method for the measurement of PM_{10} and $PM_{2.5}$ in efflux gases has been developed (ISO 23210:2009) (ISO, 2009). It is claimed to be especially suitable for measurements of mass concentrations below 40 mg/m^3, as half-hourly averages in the flue gas of different installations, such as cement and steel production plants, as well as combustion processes. It is not applicable to the sampling of flue gases that are saturated with water vapour, nor where the majority of the particles are likely to exceed PM_{10}, such as in the case of raw gases or plant operating failures. ISO 23210:2009 describes the design, use and theory of round-nozzle impactors. It does not exclude other types of impactor, provided the systems meet the performance criteria specified in ISO 23210:2009 in a validation of the impactor performed by an independent testing laboratory.

There have been trials of measurement methods for PM_{10}, and operators of incinerators and large combustion plant have had improvement conditions applied requiring them to ‘examine the feasibility’ of $PM_{2.5}$ monitoring. As yet there is no established industry consensus regarding the feasibility of $PM_{2.5}$ source monitoring. One issue under consideration is that of the secondary formation of particles in the sampler.

---

[23] The annual mean $PM_{2.5}$ objective for Scotland is 12 µg/m^3 compared with the annual mean limit value of 25 µg/m^3. If the assumed daily mean limit value is 30 µg/m^3, it would be reasonable to assume the daily mean objective would be approximately half of this value, *i.e.* 15 µg/m^3.

7.3 What Ambient $PM_{2.5}$ Monitoring has been Carried Out in the UK?

Ambient $PM_{2.5}$ monitoring using reference or reference-equivalent instruments was very limited in the UK prior to the implementation of the national network of $PM_{2.5}$ monitoring sites during 2009 to meet the requirements of the CAFE Directive (2008/50/EC). PM_{10} monitoring has been carried out over a much longer period of time, although most of the measurements were not made with a reference-equivalent sampler, and so cannot be relied upon in absolute terms. There are also historic data for the measurement of black smoke, which will include a proportion of PM_{10} and $PM_{2.5}$, but will not be directly related to current measurements of PM mass. Black smoke and PM_{10} measurements can, however, be used to provide an indication of trends in PM over a longer time period. Results show a decline in black smoke from the 1960s through to 2000, and a decline in PM_{10} during the 1990s which became less evident in the 2000s (AQEG, 2005). The black smoke network changed over to the use of aethalometers in 2008, which measure black carbon. Black smoke and PM_{10} measurements will not be considered further in this report.

7.3.1 How extensive is the current network?

$PM_{2.5}$ monitoring in the UK, using reference-equivalent FDMS monitors, is now relatively extensive (Figure 31). The majority of the sites form part of the national network (AURN), and as such they are focused on urban background sites, given that exposure-reduction for $PM_{2.5}$ forms a key part of the CAFE Directive (2008/50/EC) requirements.

As of August 2010, there were 67 sites within the AURN, of which 43 were classified as urban background[24], 17 as roadside/kerbside, 4 as industrial and 3 as rural. Figure 31 includes a further 8 sites that operate FDMS monitors; these are all in London, apart from one site in Lisburn in Northern Ireland, and are operated by local authorities. There will also be sites operated by other local authorities, industries, universities, and research institutes, however, information on such monitoring is not readily available. A full list of sites and the networks that they are part of is set out in Appendix 3.

The current AURN includes 45 sites in England (excluding London), 9 sites in London, 6 sites in Scotland, 5 sites in Wales and 2 sites in Northern Ireland[25].

---

[24] There is one site, Harlington, officially classified as 'Airport', however this is around 1 km from Heathrow Airport, and as such, $PM_{2.5}$ concentrations will be affected by the airport to a very limited extent. It is thus better to treat this site as an urban background site for $PM_{2.5}$.

[25] The locations of the sites and site photos are available at: http://aurn.defra.gov.uk

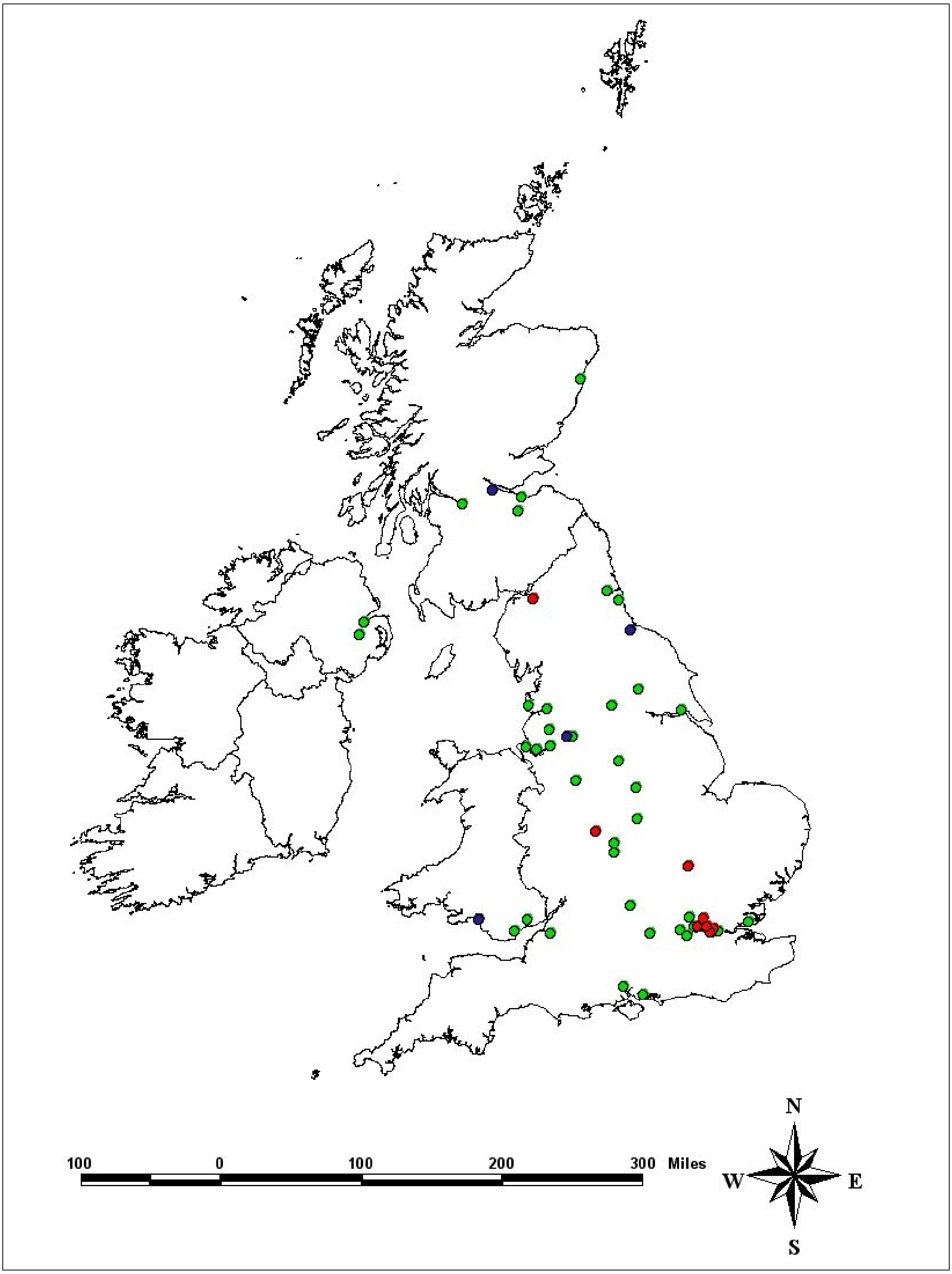

Figure 31 Sites measuring PM$_{2.5}$ in the UK (as of August 2010) using reference equivalent FDMS monitors. Roadside = red, Industrial = blue, urban background = green.

7.4 What $PM_{2.5}$ Concentrations are Measured in the UK?

As has been noted elsewhere in this report, the focus is on annual mean concentrations of $PM_{2.5}$. Valid annual means are those with >90% data capture, although a good indication of the annual mean can be derived with a data capture >75%, *i.e.* results for data capture >75% and <90% can be considered to be indicative. The analyses in the following Sections have been carried out using the OpenAir software package[26] (version 0.2-18, 26 May 2010).

7.4.1 What are current background concentrations?

Annual mean concentrations of $PM_{2.5}$ in 2009 are available for 39 background sites, 10 of which only provide indicative annual means, *i.e.* data capture is between 75-90%. When discussing the results it is useful to separate the sites into rural background and urban background.

Rural background sites

There are 3 rural sites currently operational. During 2009 results were available for just one site. The annual mean for this site, Auchencorth Moss, which is located to the south of Edinburgh, is set out in Table 18. The site recorded the lowest $PM_{2.5}$ concentration of all UK sites in 2009, at 3.4 µg/m^3.

Table 18 Summary of annual mean $PM_{2.5}$ concentrations at rural sites in 2009.

Site	Area of UK	Annual Mean (µg/m^3)
Auchencorth Moss	Scotland	3.4[a]

[a] data capture for this site >90%.

Note: Harwell is a rural site in England, near to Oxford, that started monitoring with a FDMS analyser in August 2009. The mean for the year from 9 August 2009 to 8 August 2010 was 10.4 µg/m^3.

Urban background sites

There are 43 urban background sites currently operational. However annual mean concentrations for 2009 are only available for 37 of these sites; and 10 of these only provide indicative annual means. Annual mean concentrations are summarised in Figure 32 for those sites with >90% data capture, and the results for all sites are set out in Appendix 3.

Figure 32 shows that the majority of the urban background concentrations fall in the 12-16 µg/m^3 concentration range, with no clear pattern across the UK. The highest concentrations in 2009 were at the London Eltham, Lisburn and Stoke-on-Trent Centre sites, at 17.6, 15.5 and 14.8 µg/m^3 respectively. The lowest was at the Edinburgh site, at 8.5 µg/m^3. The Edinburgh site recorded a concentration that was around 5 µg/m^3 higher than the regional background, as measured at the nearby Auchencorth Moss rural site. In London, the London Eltham site was around 7 µg/m^3 higher than the regional background, using the result for the rural site at Harwell as an indication (an annual mean concentration was not available at Harwell for 2009, thus use has been made of the concentration for the 1-year period running from 9 August 2009 to 8 August 2010, of 10.4 µg/m^3), although more typically the urban background sites in London were around 3-6 µg/m^3 above the regional background. The probable reason for the higher concentrations at the Lisburn site is discussed in Section 7.5.1. However, the reason for the higher

---

[26] Available at: http://www.openair-project.org/

annual mean concentration at the Stoke-on-Trent Centre site is currently unclear (both sites are urban background).

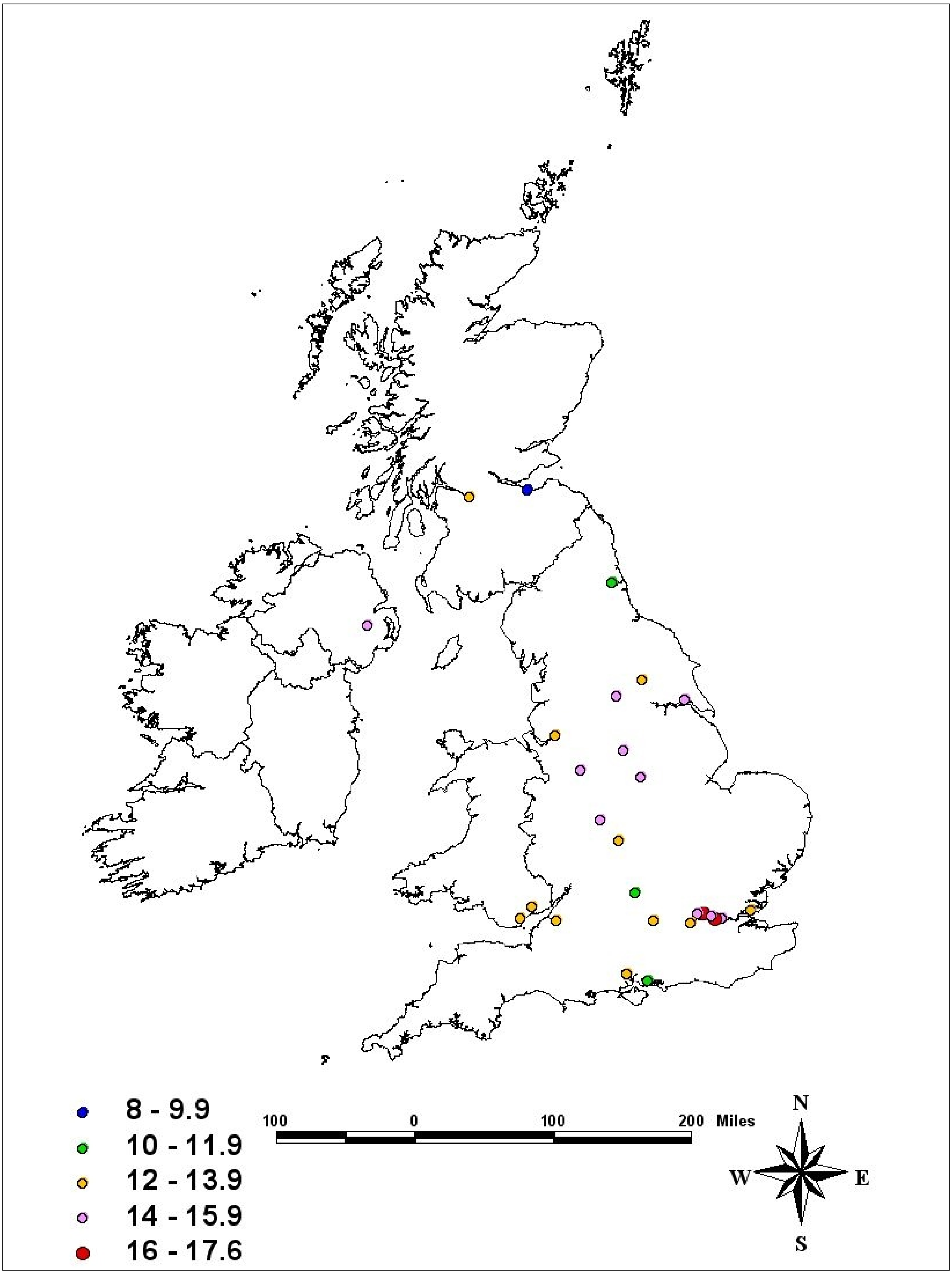

Figure 32 Summary of annual mean $PM_{2.5}$ concentrations (µg/m^3) measured at urban background sites in 2009, data capture >90%.

Comparison with modelled background concentrations

The measured background concentrations in 2009 can be compared with modelled background concentrations (see Section 4.1, Figure 22) in 2009. Figure 33 shows the measured values plotted against the modelled values for the grid square in which the monitor is located. This plot includes results for both the rural and urban background sites. There is reasonable overall agreement, as the values lie about the 1:1 line; however, there is some scatter, which may be due to limitations of the modelling exercise and the representativeness of the monitoring site of the average background concentrations in the locality, as some sites are closer to local sources than others. The clear outlier in the plot is for Lisburn in Northern Ireland, with a modelled concentration of 6.7 µg/m^3 and a measured concentration of 15.5 µg/m^3. This will reflect local sources near to the site that are not reflected in the modelling process (probably domestic combustion). The best-fit line, forced through zero and with the one outlier excluded, is shown, as well as the 1:1 line. The results provide reasonable confidence in the national background model results shown in Section 4.1, Figure 22.

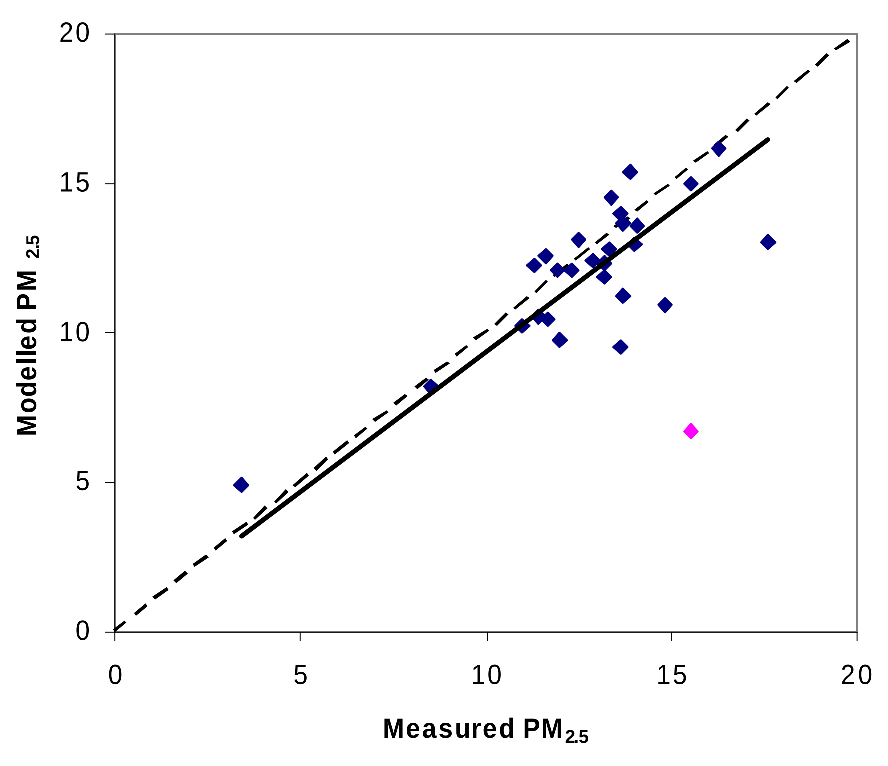

Figure 33 Comparison of measured and modelled urban and rural background $PM_{2.5}$ concentrations (µg/m^3) in 2009, data capture >90%. The best-fit line is forced through zero and excludes the one outlier, the pink diamond, which is Lisburn.

7.4.2 How much higher are roadside/kerbside concentrations?

There are 15 roadside/kerbside FDMS monitoring sites within the AURN, together with 5 additional roadside sites in London. Annual mean results are available for 7 sites in 2009 (Table 19). The box-and-whisker plots[27] in Figure 34 provide a comparison between roadside, urban background and industrial concentrations. This shows that on average

---

[27] The box-and-whisker plots are made up as follows. The box shows the median and the upper and lower quartiles, with the mean shown by the red cross. The lines extending from the box represent the upper and lower bounds of the data for data points that fall within the range of the upper interquartile + 1.5 times the interquartile range and the lower interquartile – 1.5 times the interquartile range. Values outside of these ranges are shown as blue dots.

roadside concentrations are around 2.5 µg/m^3 higher than the urban background (there were no kerbside sites with >90% data capture in 2009). This though should be treated with caution, as the locations are not matched, with the majority of the roadside sites being in London, while the urban background represents urban areas across the UK. The true elevation at roadside sites is thus probably smaller than the value of 2.5 µg/m^3.

Table 19 Summary of annual mean $PM_{2.5}$ concentrations at roadside/kerbside sites in 2009.

Site	Type	Area of UK	Annual Mean (µg/m^3)
Carlisle Roadside	Roadside	Northern England	(11)[a]
Sandy Roadside	Roadside	Southern England	15.1
Marylebone Road	Kerbside	London	(21)
Greenwich Plumstead High Street	Roadside	London	14.3
Greenwich Westhorne Avenue	Roadside	London	15.5
Harringey Town Hall	Roadside	London	14.7
Tower Hamlets Blackwall	Roadside	London	19.0

[a] values in brackets have data capture between 75 and 90%

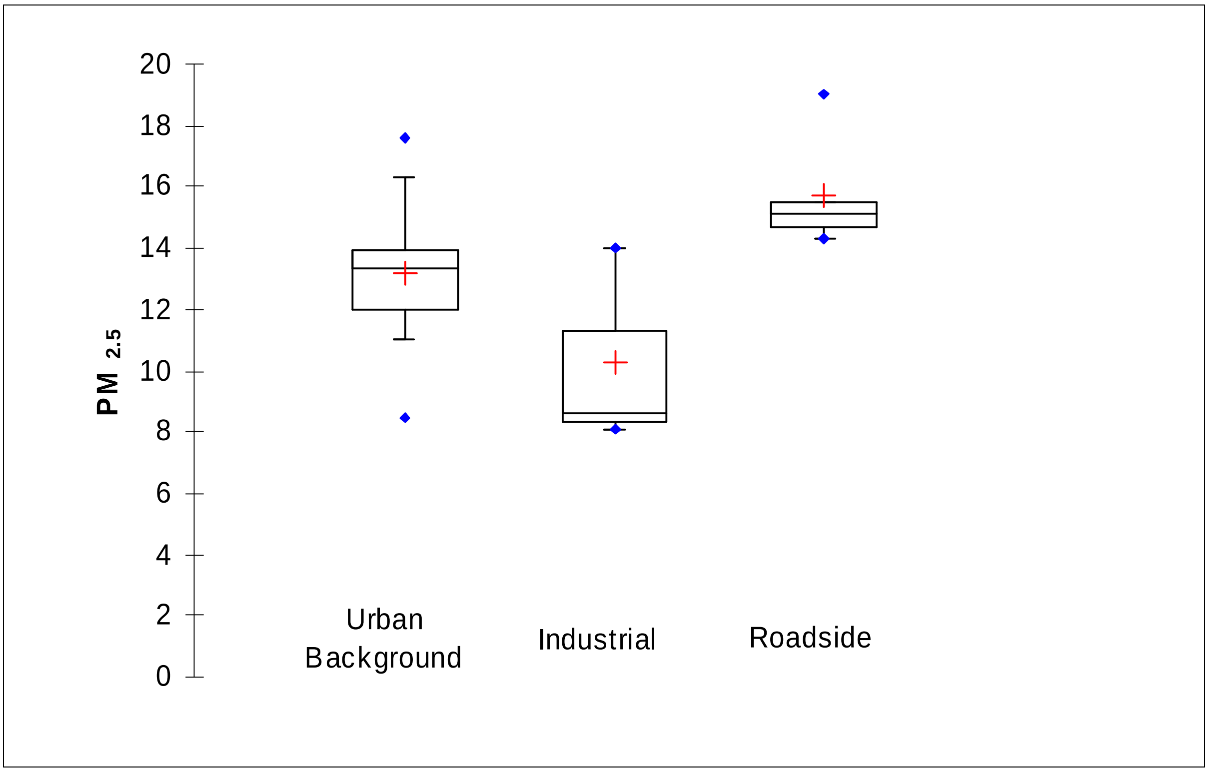

Figure 34 Summary of $PM_{2.5}$ concentrations (µg/m^3) in 2009, at urban background (n=27), industrial (n=3) and roadside (n=5) sites with >90% data capture.

In addition to the general comparison between roadside/kerbside and urban background sites set out in Figure 34, three roadside/kerbside sites can be paired with a nearby background site to allow the roadside increment to be calculated[28]:

- London Marylebone Road (Kerbside) and North Kensington;
- Birmingham Tyburn Roadside and Birmingham Tyburn; and
- Glasgow Kerbside and Glasgow Centre.

The results have been examined for matched periods and the mean roadside enhancements were 8.1 µg/m^3, 1.2 µg/m^3 and 7.2 µg/m^3 respectively. The London Marylebone Road and Glasgow Kerbside sites are close to busy roads in built up areas that are canyon like, with the monitors 1 m from the kerb, while Birmingham Tyburn Roadside site is alongside a wide dual-carriageway road in a mainly residential area, and around 7 m from the kerb. The greater distance of the Birmingham Tyburn Roadside site from the road and the more open nature of the road, together with the lower traffic flow, will account for the lower road contribution to $PM_{2.5}$ at this site.

7.4.3 What $PM_{2.5}$ concentrations have been measured around industrial sites?

There are 4 FDMS monitoring sites termed ‘industrial’ within the AURN. Annual mean results are available for three sites in 2009 (Table 20). The comparison between roadside and urban background concentrations is illustrated in the box-and-whisker plot in Figure 34. The mean concentrations are lower than the urban background values. This will relate in part to the locations of the Grangemouth and Port Talbot sites in parts of the country with lower regional background concentrations and to the fact that these two sites are not in major urban areas. The Salford Eccles site is within the Greater Manchester urban area, and this will account in large part for the higher concentration. The results suggest that industrial sources do not make a major contribution to local annual mean $PM_{2.5}$ concentrations.

Table 20 Summary of annual mean $PM_{2.5}$ concentrations at industrial sites in 2009.

Site	Type	Area of UK	Annual Mean (µg/m^3)
Grangemouth	Industrial	Scotland	8.6
Salford Eccles	Industrial	Northern England	14.0
Port Talbot	Industrial	Wales	8.1

A detailed review has recently been carried out of monitoring around the steelworks complex at Port Talbot in Wales (Hayes and Chatterton, 2009). The focus was on PM_{10}, but some $PM_{2.5}$ monitoring was also reported. Monitoring around the complex by Corus using Topas instruments (indicative optical instruments – see Table 15), showed evidence that some sources could contribute both to PM_{10} and $PM_{2.5}$ concentrations, while others were more important for the coarse component ($PM_{2.5-10}$). PM sources are thus very varied within a major complex such as the steelworks and its ancillary operations. Figure 35 shows the main sources within the Corus complex and the current location of the AURN Port Talbot monitoring site.

---

[28] **During 2009 these sites only had >75% data capture, so there is greater uncertainty associated with the results from these pairings.**

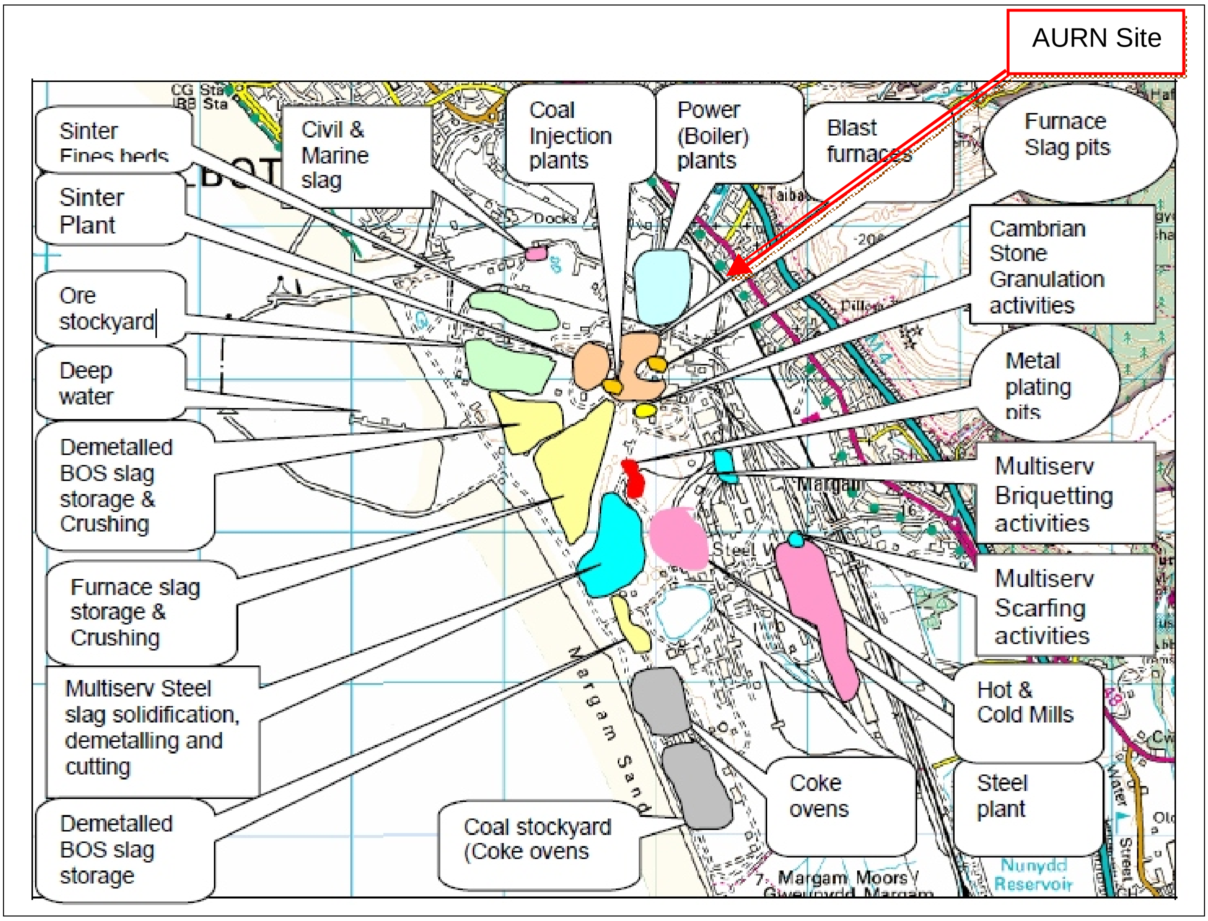

Figure 35 Layout of the Corus site at Port Talbot, showing the location of the current AURN monitoring site. Source: Environment Agency (2009b)

Particular attention was paid by Hayes and Chatterton (2009) to the monitoring at the AURN site within Port Talbot. This was located within the residential area to the east of the steelworks, although the site had to be relocated in 2007. The current 'Fire Station' site is located to the northeast of the steelworks, around 300 m from the nearest works and around 700 m from the blast furnace. The Hayes and Chatterton (2009) report includes polar plots (see Section 7.6 for an introduction to these plots) showing concentrations at the Fire Station site by wind direction and wind speed (wind speed increases away from the centre of the plot). The plots show very different patterns for the three pollutants PM$_{10}$, PM$_{2.5}$ and NO$_x$ (Figure 36). The highest PM$_{10}$ concentrations are associated with stronger winds from the south-southwest, which is consistent with either (or both) process emissions from elevated sources being brought down to ground with stronger winds, or fugitive PM raised with stronger winds. The PM$_{2.5}$ pattern is different, with the highest concentrations associated with stronger winds (>7 m/s) from the east-northeast. The authors of the report ascribe this to a road traffic contribution from the M4 motorway, which lies 370 m to the east. However, this pattern is close to that for sites throughout the UK (see Section 7.6.1), and is thus more likely to be due to long-range transport of secondary PM (see discussion in Section 7.6.1). There is some evidence of elevated PM$_{2.5}$ associated with the PM$_{10}$ source(s) to the south-southwest, but while the PM$_{10}$ concentrations rise to over 80 µg/m^3, the PM$_{2.5}$ concentrations only rise to around 15 µg/m^3 in this sector. The steelworks is thus a more significant source of PM$_{10}$ than PM$_{2.5}$. The pattern for NO$_x$ concentrations is very different to that of PM$_{10}$ and PM$_{2.5}$, showing the highest concentrations with low wind speeds. This is consistent with emission from local ground-level sources, probably road traffic.

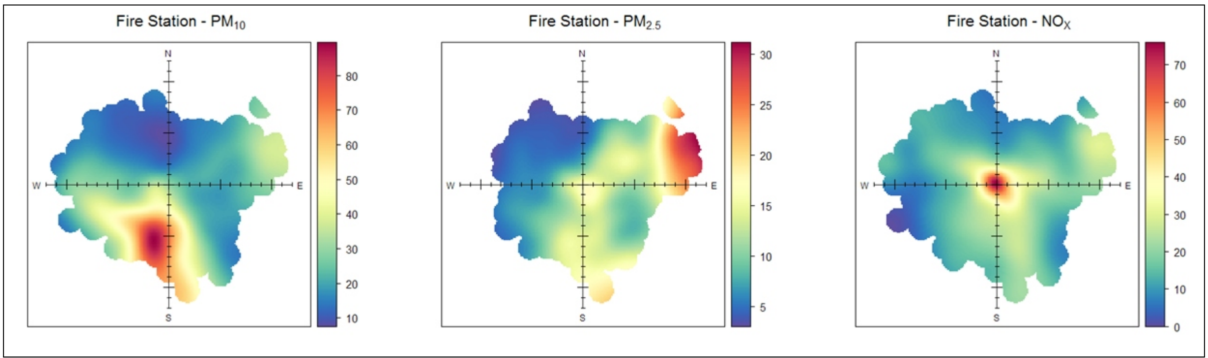

Figure 36 Polar plots of PM_{10}, $PM_{2.5}$ and NO_x concentrations (µg/m^3) at Port Talbot, 2007-2009. Source: Hayes and Chatterton (2009). The wind speeds are shown on the x/y lines at 1 m/s intervals. North is at the top of the diagram.

The FDMS data from the Fire Station AURN site have been examined for the two-year period April 2008 to March 2010 (all ratified data) specifically for this report. The mean $PM_{2.5}$ concentration was 9.1 µg/m^3, while the mean PM_{10} concentration was 25.2 µg/m^3. The $PM_{2.5}$ is slightly elevated above the regional background, which for this part of Wales is 6-8 µg/m^3 (see Figure 22 in Section 4.1), while the PM_{10} is more significantly elevated. The resultant $PM_{2.5}$:PM_{10} ratio is 0.36, which is much lower than the expected background of 0.65 for Wales (see Table 21 in Section 7.4.4). The diurnal pattern of $PM_{2.5}$ concentrations is different to that of PM_{10}, SO_2 and NO_x (Figure 37). The patterns for PM_{10} and SO_2 are similar, and are consistent with process emissions from the steelworks. The NO_x pattern is more consistent with that expected for road traffic. The $PM_{2.5}$ pattern is more complex and may include contributions from the steelworks (following the SO_2/PM_{10} pattern) and from road traffic (following the NO_x pattern), superimposed on the regional pattern, as discussed in Section 7.5.1. This is consistent with the steelworks and road traffic sources making a limited contribution to $PM_{2.5}$ on an annual mean basis.

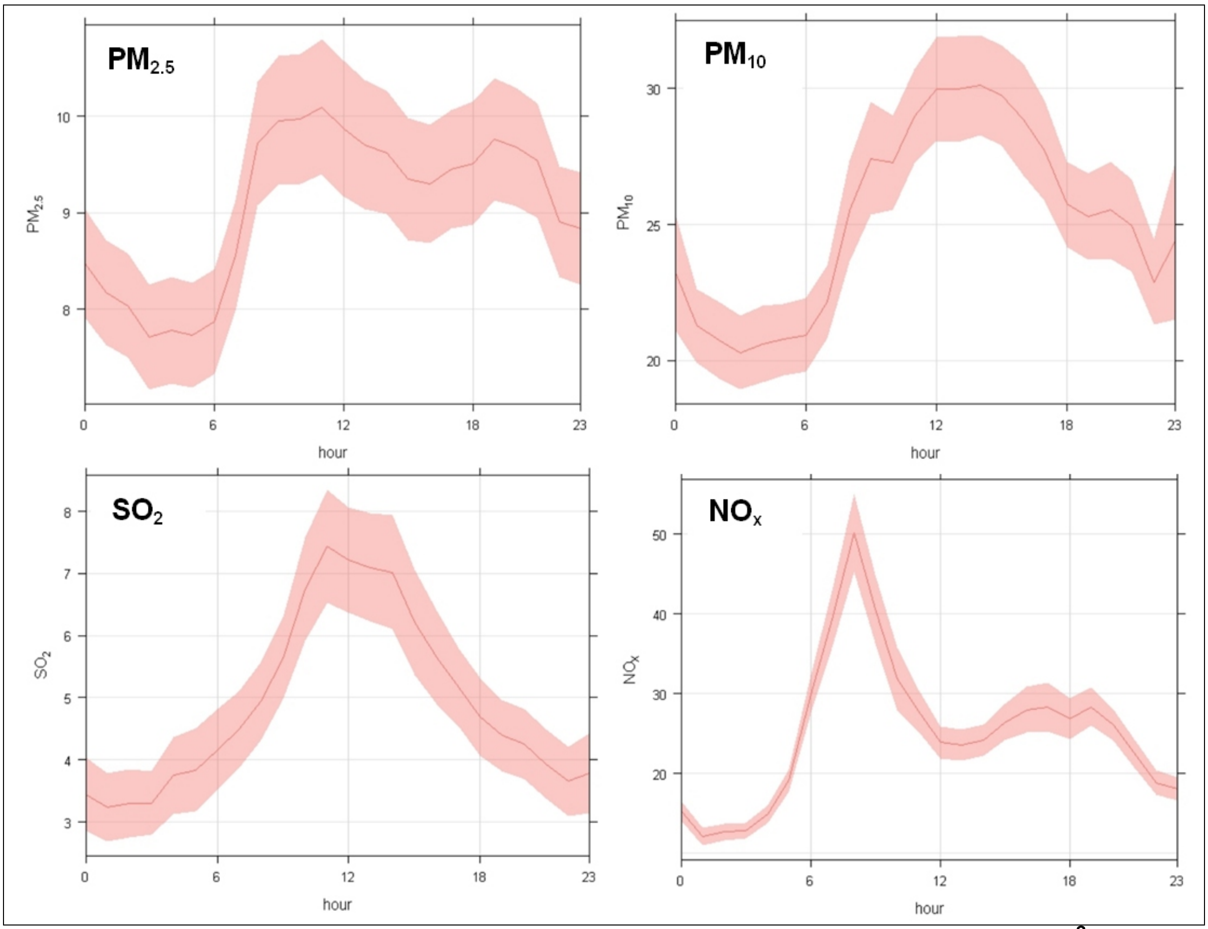

Figure 37 Diurnal patterns of PM$_{2.5}$, PM$_{10}$, SO$_2$ and NO$_x$ concentrations (µg/m^3) at Port Talbot AURN (Fire Station) site, for the 2-year period April 2008 to March 2010. The shading represents 95th percent confidence interval. Note different scales.

7.4.4 What are the ratios between PM$_{2.5}$ and PM$_{10}$?

Reliable ratios can only be derived from measurements made with reference equivalent instruments. This essentially confines the examination of PM$_{2.5}$ to PM$_{10}$ ratios to results from FDMS instruments. During 2009 there were 23 AURN sites where both PM$_{2.5}$ and PM$_{10}$ were monitored and data capture was >75% (13 of the sites had >90% data capture for both pollutants – it was considered helpful to extend the number of sites by relaxing the criterion to >75% data capture). Average PM$_{2.5}$:PM$_{10}$ ratios for each site were derived as the mean of the hourly ratios, to ensure matched periods were used.

The results are summarised as box-and-whisker plots for urban background, roadside and industrial sites in Figure 38, and presented in full in Appendix 5. The mean values are fairly similar, but there is much greater variability in the industrial sites and a suggestion that the ratio is higher for roadside sites. However, as shown below the latter suggestion is not borne out when examining the results in more detail.

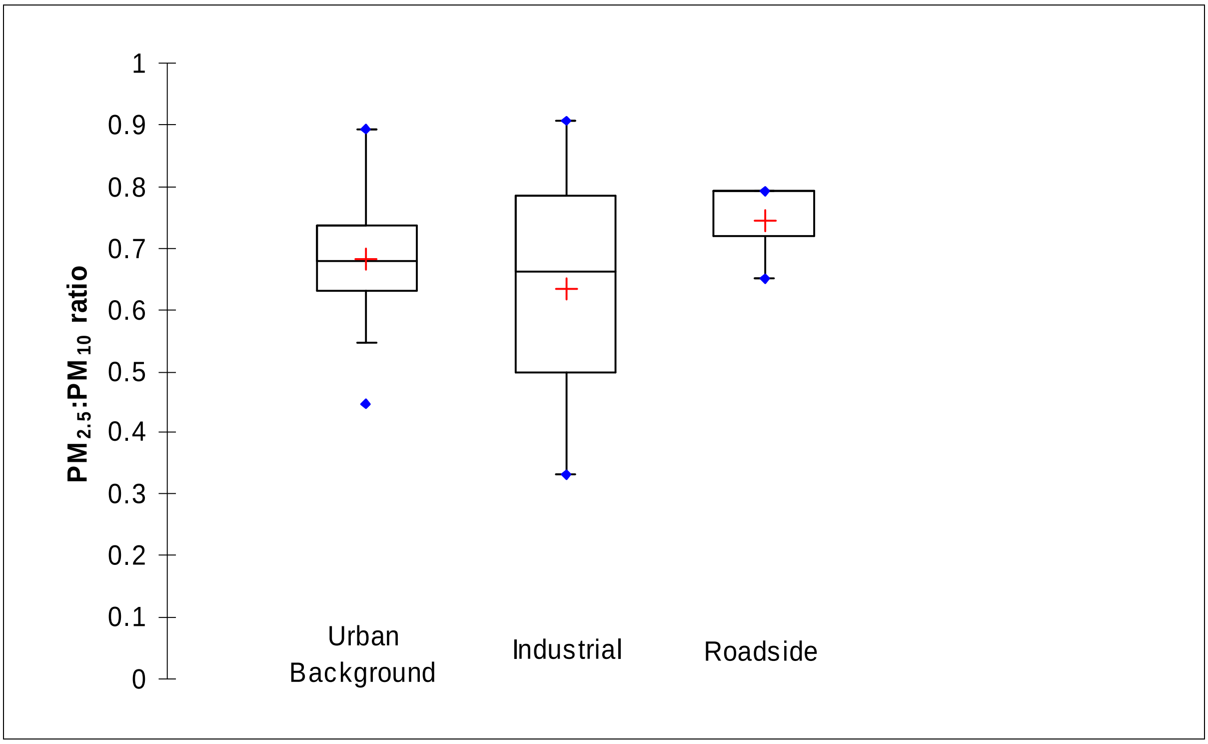

Figure 38 Summary of $PM_{2.5}$:PM_{10} ratios at different site types in 2009, at urban background (n=17), industrial (n=3) and roadside (n=3) sites with >75% data capture.

The $PM_{2.5}$:PM_{10} ratios at urban background AURN sites in 2009 are shown across the UK in Figure 39. There is no immediately clear pattern, although the highest ratio is in central London, while the lowest is in Glasgow Centre. In a further analysis, the results have been plotted as a function of distance from Dover (Figure 40) (Dover was selected as it is known that background concentrations decline from the southeast towards the northwest of the UK (see Figure 22 in Section 4.1)). There is a statistically significant relationship (p=0.15), with ratios declining from around 0.8 in Southeast England to below 0.6 in Scotland. The fine fraction of PM thus becomes more important on moving towards continental Europe, which is consistent with secondary PM being more significant in southeast England (see Figure 23 in Section 4.1.2). The scatter around the line will relate in part to the varying contributions of sea salt and resuspended dust from agricultural and construction activities, as these sources will have a more prominent coarse PM component, tending to reduce the $PM_{2.5}$:PM_{10} ratio.

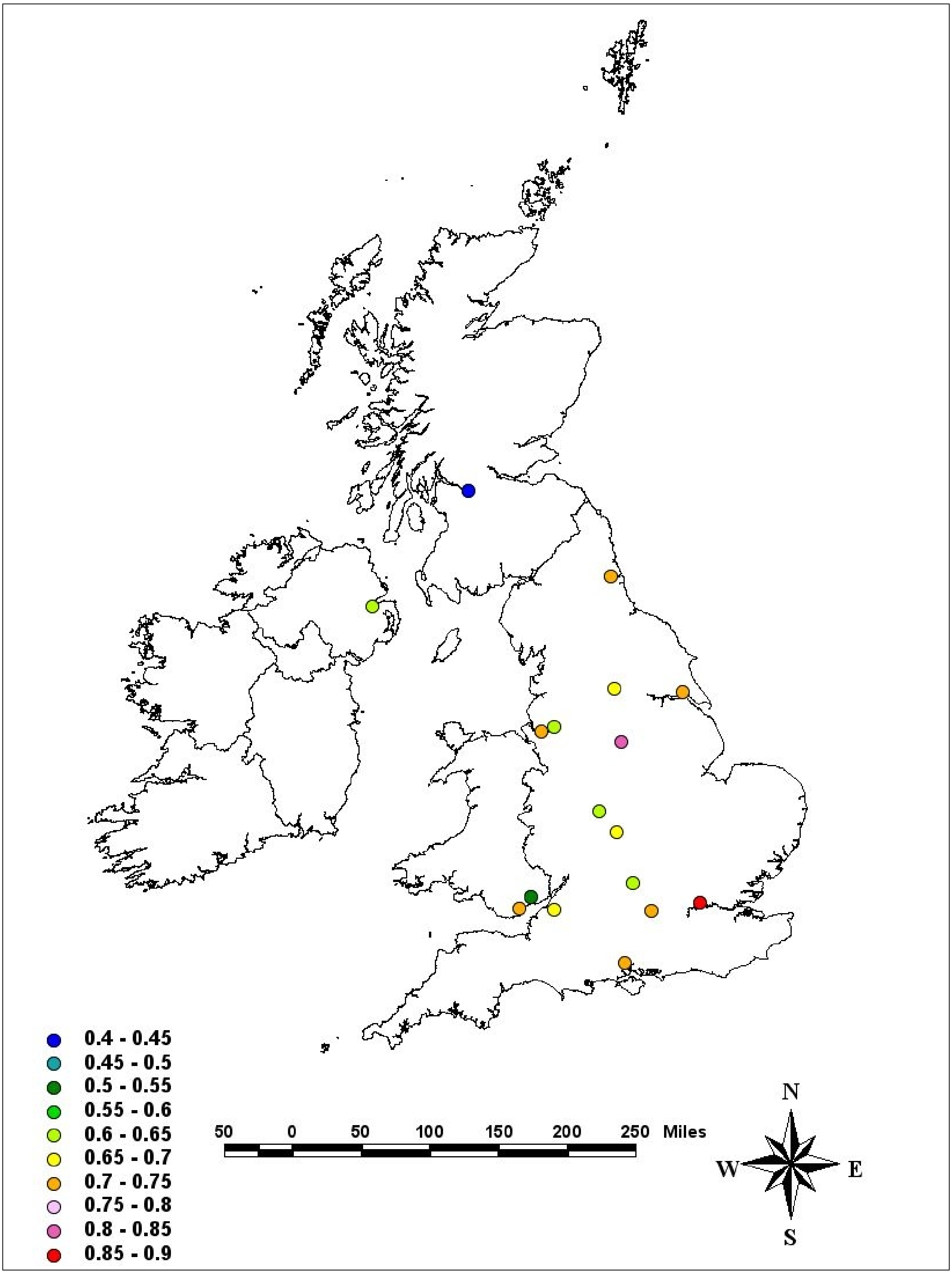

Figure 39 Ratios of $PM_{2.5}$:PM_{10} at urban background sites in 2009 across the UK.

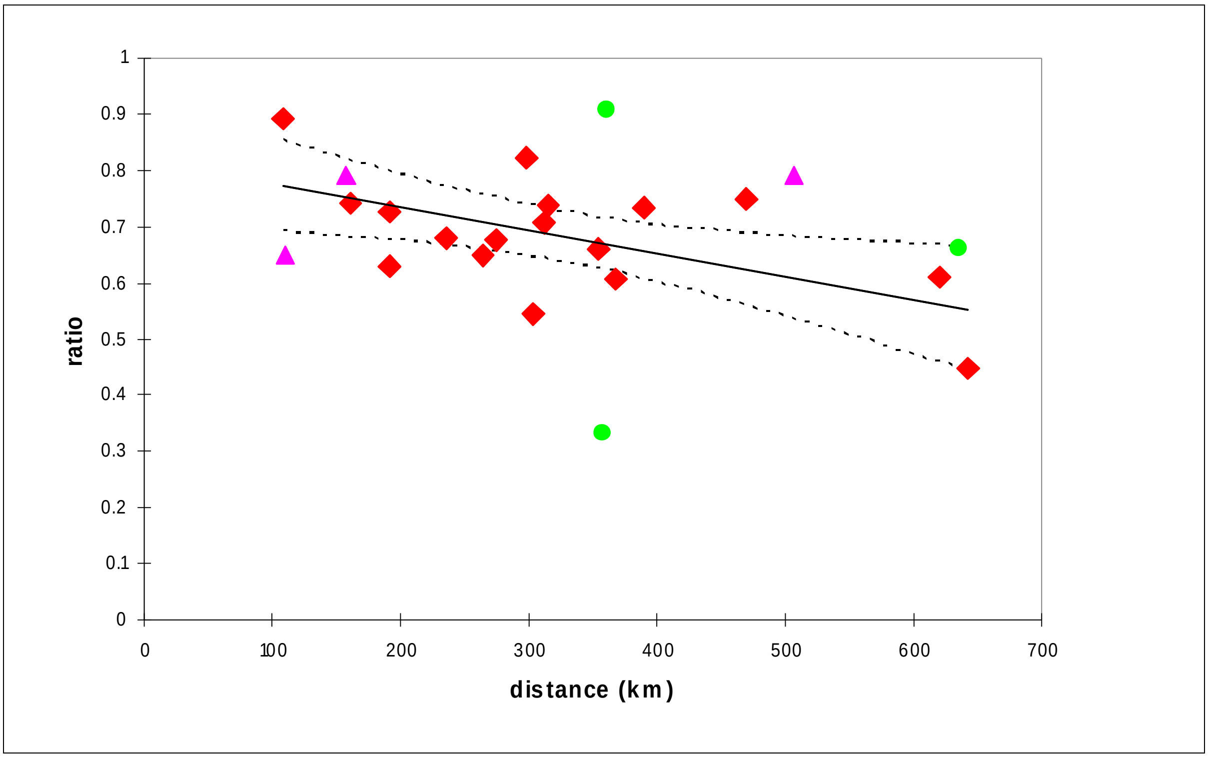

Figure 40 Ratios of $PM_{2.5}$:PM_{10} in 2009 as a function of distance from Dover. Red diamonds = urban background, pink triangles = roadside, green circles = industrial. The line is fitted to the urban background sites, with 95% confidence limits shown as dashed lines.

Also shown in Figure 40 are the ratios at roadside sites, plotted against distance from Dover. There are only three sites, but there is no evidence that the ratio for roadside sites differs from the urban background pattern. This implies there is no particular enhancement of either the coarse or the fine fraction of PM at roadside sites. Industrial sites show much greater variability in the $PM_{2.5}$:PM_{10} ratios, which will relate to the differing proportions of fine and coarse PM being emitted from the industrial sources. The very low ratio in Figure 40 is for the site at Port Talbot, near to the major steelworks complex, which shows that coarse PM is a major component of the emission from these works (see Section 7.4.3).

The analysis set out above would suggest that the $PM_{2.5}$:PM_{10} ratios presented in Table 21 could be applied to PM_{10} data in different parts of the UK to estimate annual mean urban background $PM_{2.5}$ concentrations. These values should be updated as new data become available.

Table 21 Indicative $PM_{2.5}$:PM_{10} ratios that apply in different parts of the UK.

Region of UK	$PM_{2.5}$:PM_{10} ratio	Indicative confidence interval[a]
Scotland	0.55	0.44 - 0.66
Northern Ireland	0.55	0.44 - 0.66
Northern England	0.65	0.60 – 0.70
Wales	0.65	0.60 – 0.70
Southwest and Central England	0.70	0.65 - 0.74
Southeast England and East Anglia	0.75	0.68 – 0.83

[a] The confidence interval is the 95% confidence interval on the line at the values in the ratio column, but no allowance is made for the range that might apply across the geographic areas specified, which will be somewhat greater, hence it is called indicative.

7.4.5 How does $PM_{2.5}$ relate to other pollutants?

There are a number of sites in the national network measuring a range of pollutants in addition to $PM_{2.5}$. The correlations between $PM_{2.5}$ and these pollutants have been derived using hourly data for each site with >75% data capture in 2009. The correlation coefficients of the hourly-mean data are summarised as box-and-whisker plots in Figure 41, separated into background (mostly urban background), roadside and industrial sites.

There is a strong consistency in the correlation coefficients at the different sites, especially in the case of the urban background sites. The highest correlation at all sites is unsurprisingly between $PM_{2.5}$ and PM_{10} (correlation coefficients typically 0.8 to 0.9) (this is in large part because $PM_{2.5}$ accounts for a major proportion of PM_{10}). $V_{2.5}$ concentrations are also highly correlated with $PM_{2.5}$ (correlation coefficient typically 0.7 to 0.8). $V_{2.5}$ is a measure of the volatile $PM_{2.5}$, derived from FDMS monitors as the ‘purge’ concentration (see Section 7.1.2). The correlation with V_{10} is not as strong (correlation coefficient typically 0.4 to 0.7), suggesting that the volatile component of PM_{10} (V_{10}) is somewhat different to that associated with $PM_{2.5}$. The NO_x and NO_2 concentrations are also fairly highly correlated with $PM_{2.5}$ (correlation coefficient 0.5 to 0.6), at all site types. The correlation with CO is weaker at background and industrial sites (correlation coefficient ~0.4), but is similar to that for NO_x and NO_2 at the one roadside site for which CO results are available (Marylebone Road in London). The correlation with SO_2 is lower still at background and industrial sites (correlation coefficient ~0.3), but is much higher at the one roadside site (which is probably due to the SO_2 associated with the residual sulphur in vehicle fuels). The correlation with O_3 is negative (correlation coefficient ~-0.4), thus higher O_3 concentrations are associated with lower $PM_{2.5}$ concentrations. This will be due to increased concentrations of $PM_{2.5}$ being associated with increased NO_x, which leads to lower O_3 concentrations. There may be short periods when $PM_{2.5}$ and O_3 are positively correlated during photochemical episodes, when both $PM_{2.5}$ and O_3 are generated by the photochemical activity, but on an annual basis these are swamped by the negative correlation. These patterns are consistent with those found elsewhere (see Section 2.6).

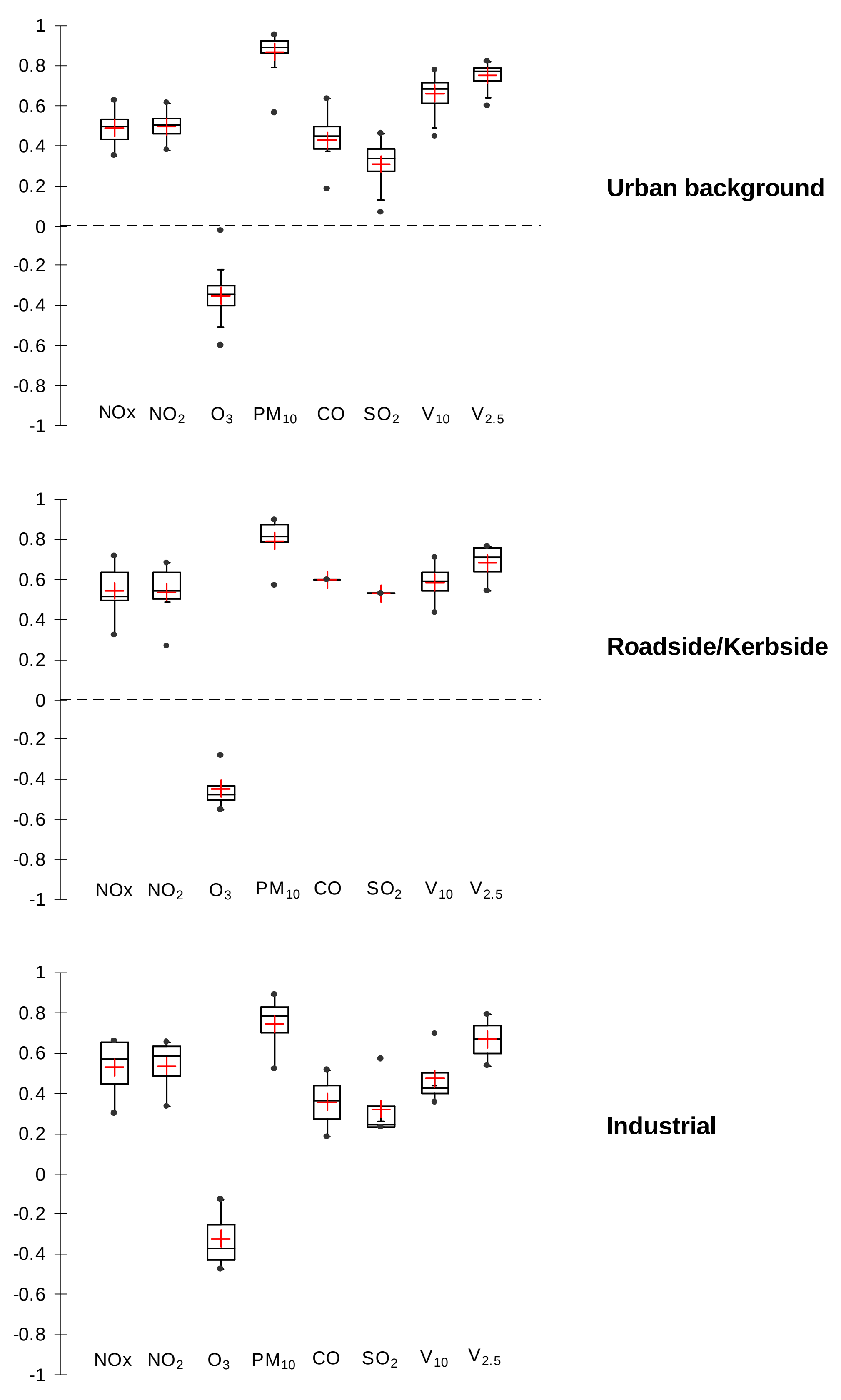

Figure 41 Summary of correlations of $PM_{2.5}$ with other pollutants at urban background (n=34), roadside/kerbside (n=8) and industrial (n=4) sites in 2009 (V_{10} and $V_{2.5}$ are explained in the text). The correlation coefficients are for hourly-mean data for sites with >75% data capture.

7.5 What are the Temporal and Spatial Patterns for $PM_{2.5}$?

Temporal and spatial patterns of $PM_{2.5}$ have been examined using data collected during 2009 using reference equivalent FDMS monitors. Consideration has only been given to sites with >75% data capture.

7.5.1 What are the diurnal patterns for $PM_{2.5}$ and how do they vary across the UK?

Background sites

The diurnal cycle of $PM_{2.5}$ during 2009 is summarised in Figure 42 for four geographic areas of the UK. Concentrations have been averaged by hour-of-the-day using local time, *i.e.* they take account of the change from winter to summer time. The plots show average concentrations for the hour beginning, *i.e.* the value for 23:00 h is the average over the period 23:00 to 24:00 h. Note that the scales change in each of the plots. The delineation of the four areas and the sites within each area are shown in Figure A6.1 in Appendix 6.

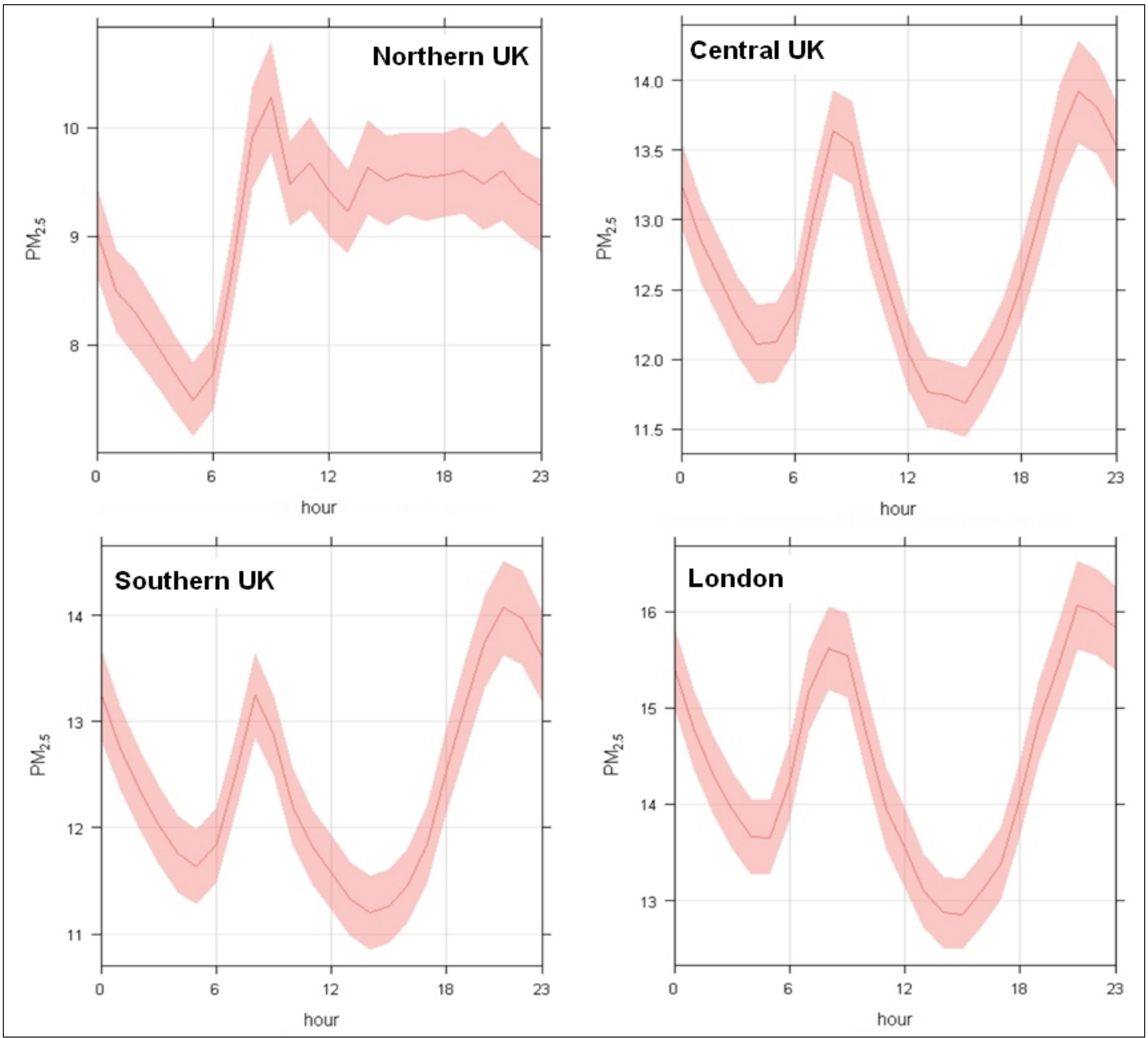

Figure 42 **Variation of $PM_{2.5}$ concentrations (µg/m^3) at urban background sites by hour-of-the-day in 2009, at sites in Northern UK (n=8), Central UK (n=17), Southern UK (n=8) and London (n=8). The shading represents 95th percent confidence interval. Note different scales.**

Afternoon Dip

All sites show a dip in concentrations in the early morning, between 04:00-06:00 h, then a steep rise to a peak between 07:00-10:00 h. At all sites, apart from those in northern UK, there is then a drop to give the lowest concentrations of the day between about 13:00-17:00 h, before rising to the highest peak of the day between about 20:00-23:00 h. The pattern at northern UK sites is distinctly different, as there is no clear drop during the afternoon, and no evidence of a late evening peak. The diurnal range is, however, relatively limited, averaging around 2.75 µg/m^3 at northern UK sites, 2.25 µg/m^3 at central UK sites, 2.75 µg/m^3 at southern UK sites and 3.25 µg/m^3 in London (values quoted to the nearest 0.25 µg/m^3). A separate examination of the diurnal pattern at a site measuring $PM_{2.5}$ with a beta attenuation monitor has confirmed this pattern and shows that it is not an artefact of the FDMS analyser. The diurnal pattern over the whole year is retained in all four areas when separated into winter and summer periods (October-March and April-September respectively). Figure 43 shows the results for the Southern UK region. The pattern is broadly similar to the annual pattern (*cf.* Figure 42, Southern UK), although in the summer the afternoon dip is longer duration, while in winter the evening peak is more prominent. This first observation is consistent with a greater loss of volatile particles from ambient $PM_{2.5}$ during the longer daylight hours in summer months, while the second observation may relate to greater domestic heating emissions during the early part of the night in winter.

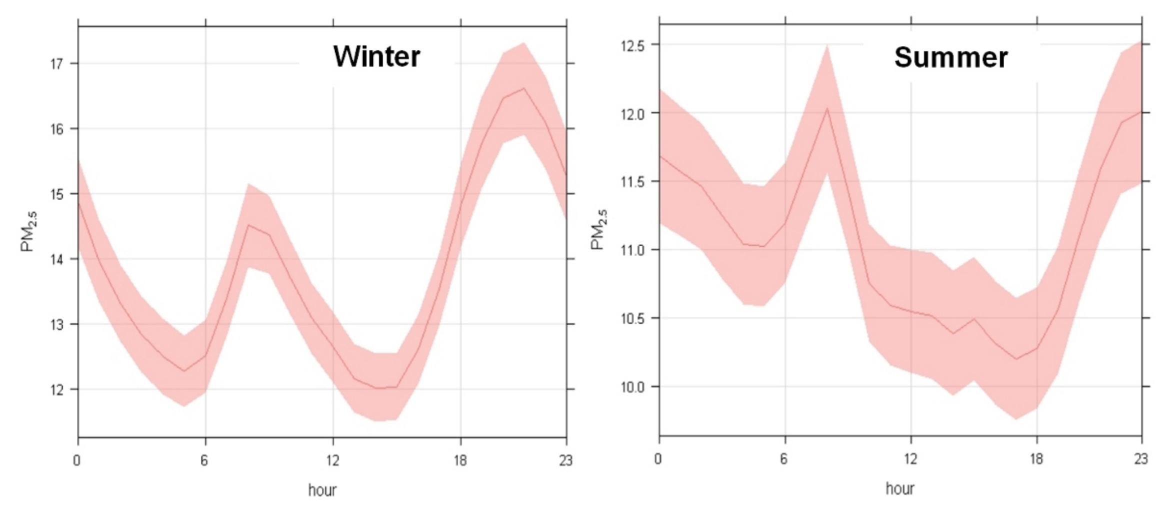

Figure 43 Variation of $PM_{2.5}$ concentrations (µg/m^3) at urban background sites by hour-of-the-day in 2009 at Southern UK sites (n=8) in winter and summer. The shading represents 95th percent confidence interval. Note different scales.

The pattern during the first half of the day is similar to that seen for most pollutants that are related to typical urban emission sources. The lowest concentrations occur in the early hours, when emissions are at their lowest, with a rise to a peak between 07:00-10:00. This peak is usually ascribed to the morning rush-hour traffic, although domestic emissions will also increase at this time. The subsequent decline in the peak is normally related to the greater atmospheric turbulence during the day, as well as to a decline in both traffic flow and domestic heating emissions. Notably for $PM_{2.5}$, this decline continues during much of the daytime to give concentrations in the middle of the afternoon (13:00-17:00 h) that are lower than those in the early morning. This is not expected and is not seen in the results for nitrogen oxides (AQEG, 2004; see also Figure 37). The most plausible explanation for the afternoon dip is the loss of semi-volatile PM (principally nitrate, with some organic compounds) from the ambient PM, as a result of the higher temperatures during this part of the day.

If this dip is due to a loss of semi-volatile PM, then it would be expected that volatile $PM_{2.5}$ ($V_{2.5}$), measured as the purge concentration in the FDMS analysers (see Section 7.1.2 other monitoring methods), would decline during the afternoon, as the ambient particles would have lost their semi-volatile material before they enter the analyser. However, the $V_{2.5}$ results do not show this, as shown in Figure 44, which presents the average diurnal profile for $V_{2.5}$ at southern UK and northern UK sites (the central UK $V_{2.5}$ results are similar to those in southern UK and are not shown). Both plots show high $V_{2.5}$ concentrations in the early hours of the day, 00:00-04:00 h, and a minimum during the time of the morning $PM_{2.5}$ peak (*cf.* Figure 44 and Figure 42). During the middle of the day, when $PM_{2.5}$ is at its lowest, $V_{2.5}$ is higher. This would suggest that the purge measurement from the FDMS analyser is not directly related to volatile $PM_{2.5}$. However, it is worth noting that the $V_{2.5}$ plot for southern UK sites is similar in shape to that for $PM_{2.5}$ (*cf.* Figure 44, Southern UK and Figure 42 Southern UK), but with the $V_{2.5}$ results showing a time shift forwards of some 3-5 hours. This may reflect a delayed loss of volatile PM from the FDMS analyser (which would in turn affect the pattern of total $PM_{2.5}$ concentrations). Green (2007) has reported a 1-2 hour time delay in the purge concentration from an FDMS analyser. It is also of note that the variation in $V_{2.5}$ during the day is less than 1 µg/m^3, which is much smaller than the diurnal fluctuation of $PM_{2.5}$, which is around 3 µg/m^3.

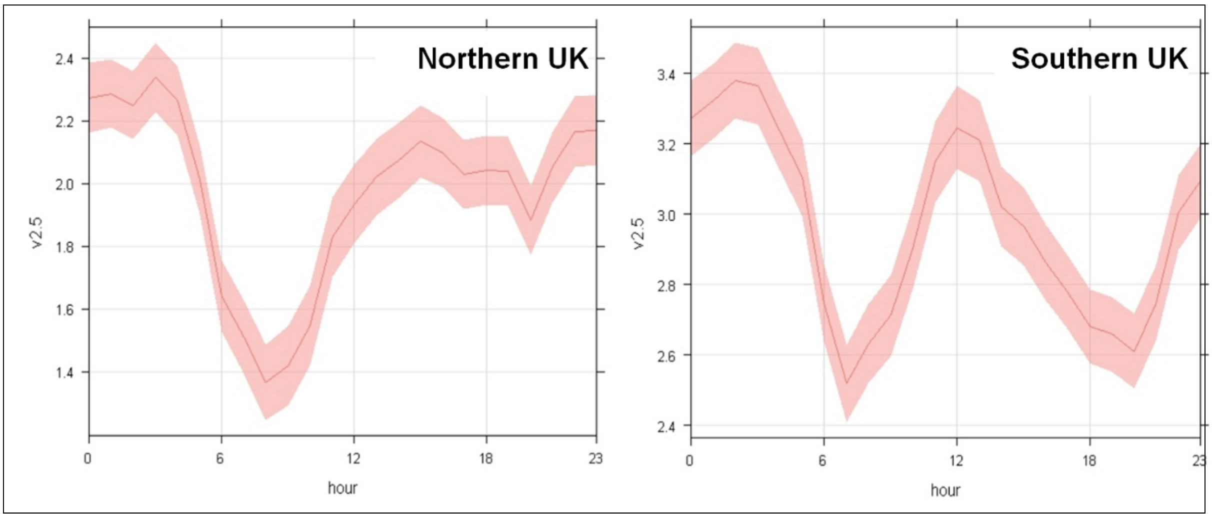

Figure 44 Variation of volatile $PM_{2.5}$ concentrations ($V_{2.5}$ in µg/m^3) at urban background sites by hour-of-the-day in 2009, at sites in Northern UK (n=8) and Southern UK (n=8). The shading represents 95th percent confidence interval. Note different scales.

Further insight into the diurnal pattern can be obtained by examining nitrate concentrations measured using a Rupprecht and Patashnick 8400N Nitrate Analyser with a $PM_{2.5}$ sampling inlet. The hourly-average concentrations for 2009 have been examined and the pattern by hour-of-the-day is shown in Figure 45 for two sites in southern England. There is a dip in nitrate concentrations during the afternoon. This supports the view that the afternoon dip in $PM_{2.5}$ concentrations is related to loss of volatile nitrates from ambient $PM_{2.5}$.

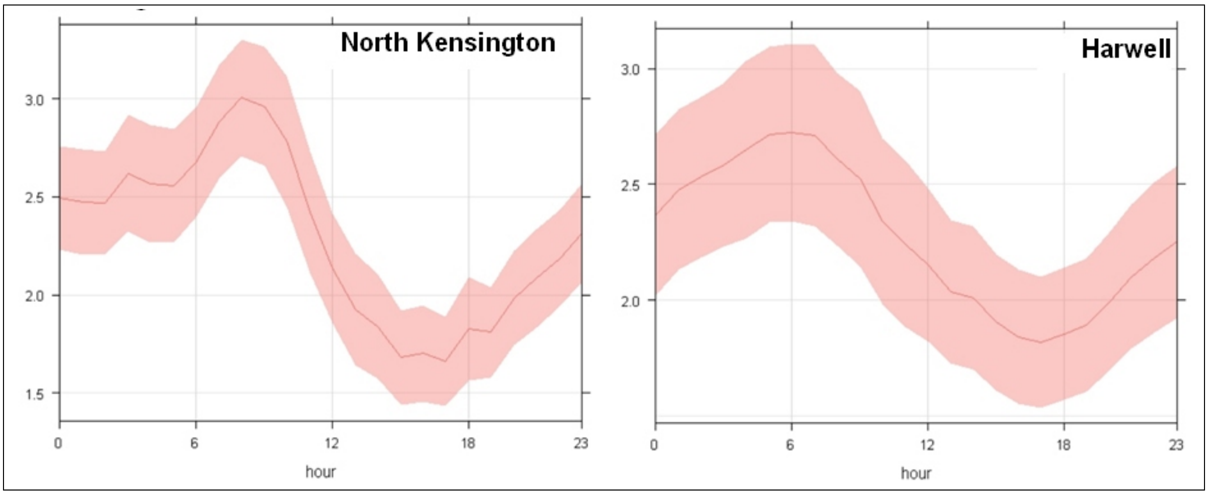

Figure 45 Variation of nitrate concentrations (µg/m^3) by hour-of-the-day in 2009 at North Kensington (urban background) and Harwell (rural). The shading represents 95th percent confidence interval. Note different scales.

To provide further understanding of these observations, the diurnal profiles have been separated into periods: easterly winds (40°-140°) and winds from all other directions. The pattern is similar at sites from Warrington in the north of England to Southampton in the south of England, with an example shown for the site at Leamington Spa in the Midlands (Figure 46). The strong diurnal pattern with low afternoon concentrations is seen in the annual results for all wind directions other than easterly (Figure 46, All other). When the winds are from the east, concentrations are higher overall and the pattern is much more uniform, although with a tendency towards higher concentrations during the evening (seen at all four sites considered – not shown). As is discussed in Section 7.6.1, the high concentrations with easterly winds are likely to be associated with long-range transport of well mixed and spatially homogeneous PM from continental Europe. These observations would suggest that, if the low afternoon concentrations are due to loss of semi-volatile PM, then a) semi-volatile PM is not strongly associated with easterly winds and hence not strongly associated with secondary PM, and b) the semi-volatile PM is associated with winds other than those from the east, which will have limited secondary PM. On the other hand, it may be that the loss of semi-volatile PM is not the explanation for the afternoon dip.

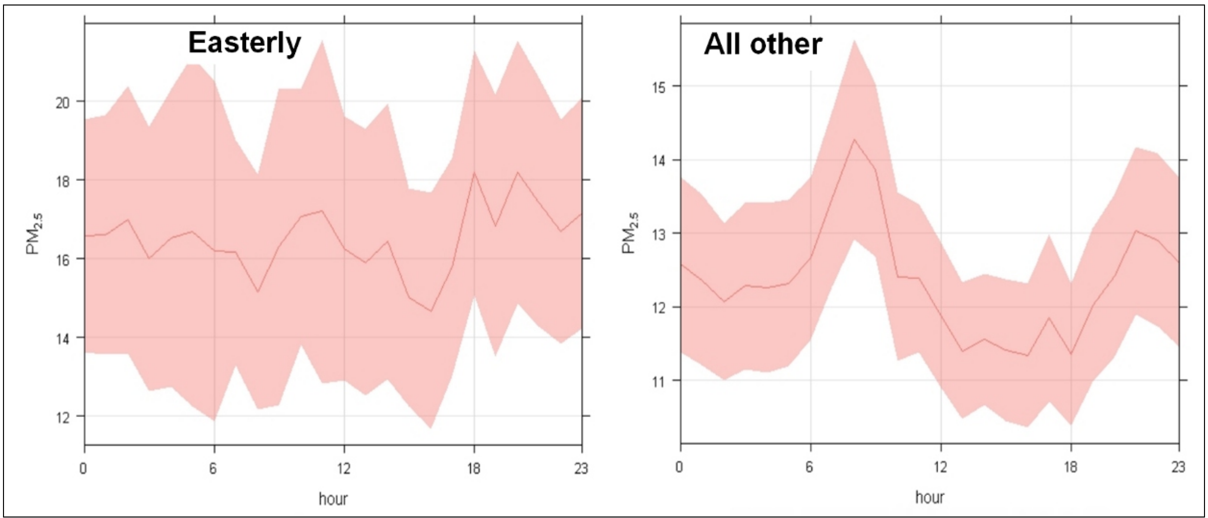

Figure 46 Variation of $PM_{2.5}$ concentrations (µg/m^3) by hour-of-the-day, at the Leamington Spa site in 2009 for easterly winds (40°-140°) and all other wind directions. The shading represents 95th percent confidence interval. Note different scales.

Late evening, early night-time peak

The other unusual feature of the $PM_{2.5}$ results is the late evening, early night-time (20:00-23:00 h) peak. In part this will be due to reduced dispersion arising from the more stable atmospheric conditions during the night. It may also reflect a contribution from domestic sources, both from heating and cooking (see Section 4.1.2 for a discussion of cooking sources). It is also possible that there is a contribution of volatile PM condensing on ambient particles, with the lower night-time temperatures (nitrate concentrations increase overnight - see Figure 45).

Roadside and industrial sites

The diurnal patterns for roadside and industrial sites during 2009 are shown in Figure 47. The diurnal range is only slightly larger at roadside sites, averaging around 3.75 µg/m^3 (ranges cited to nearest 0.25 µg/m^3) but is substantially less at industrial sites, averaging around 1.5 µg/m^3. The afternoon dip is also not as prominent at roadside sites as it is at background sites, nor is the late evening peak (*cf.* Figure 47, Roadside and Figure 42). There is no clear peak during the evening ‘rush hour’ 16:00-18:00, although concentrations during this period are somewhat higher than seen in the pattern at urban background sites, suggesting an influence of the higher traffic flows at this time.

The absence of a clear late evening / early night-time peak is more apparent than real, as the late evening concentrations are elevated above the early morning minimum to a similar extent at roadside and urban background sites; being around 2.25 µg/m^3 higher, compared with values of around 1.75, 1.75, 2.25 and 2.5 µg/m^3 for the four background regions (*cf.* Figure 47, Roadside and Figure 42). This would suggest that the late evening peak is not strongly related to road traffic, it is though being disguised at roadside sites by higher early evening concentrations which are likely to be related to road traffic, *i.e.* the traffic peak is superimposed on the rising limb of the late evening peak.

The range of diurnal concentrations is much lower at industrial sites. The morning and evening peaks are still evident, suggesting that the background pattern is playing a role, but the emissions from the industrial sources, which are likely to be more constant throughout the day, and will occur to some extent at night time, will dampen the background pattern.

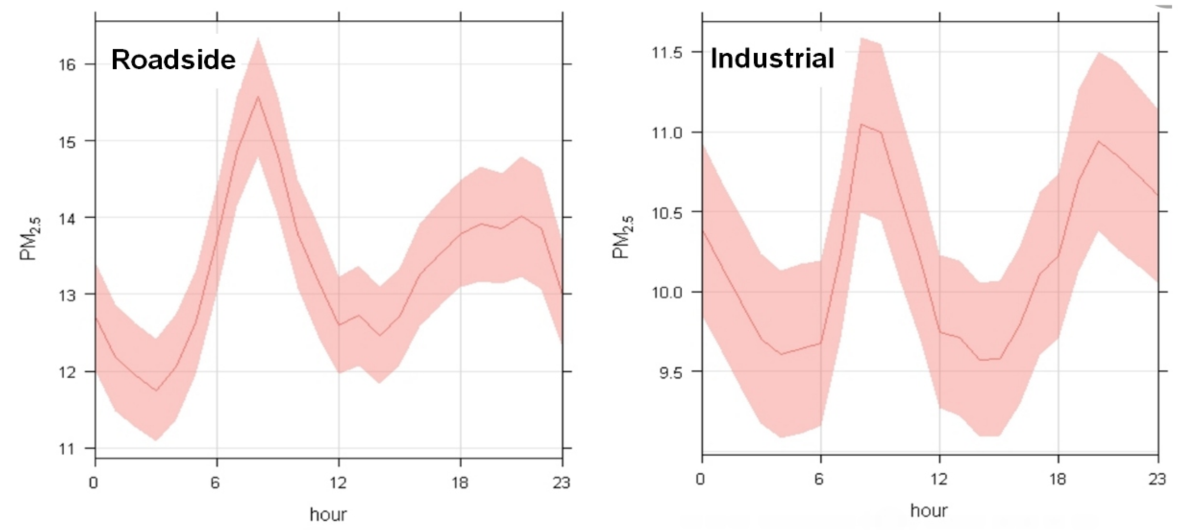

Figure 47 Variation of $PM_{2.5}$ concentrations (µg/m^3) by hour-of-the-day, at roadside (n=10) and industrial sites (n=4) in 2009. The shading represents 95th percent confidence interval. Note different scales.

Lisburn, Northern Ireland

It was noted in Section 7.4.1 that the Lisburn site in Northern Ireland was anomalous, with much higher measured $PM_{2.5}$ concentration in 2009 than suggested by the modelled background concentration for this location. The diurnal variation at the Lisburn site is shown in Figure 48, revealing a very different pattern, with the range over the day being three times higher than at other urban background sites, at just over 9 µg/m^3. This is primarily due to high over-night concentrations at this site, which peak at a similar time to the peaks seen at other background sites (*cf.* Figure 42). It is suspected that these high concentrations are due to domestic heating, which is still supplied in part by solid fuel. This is supported by the observation that the evening peak is much larger during the winter than the summer (not shown).

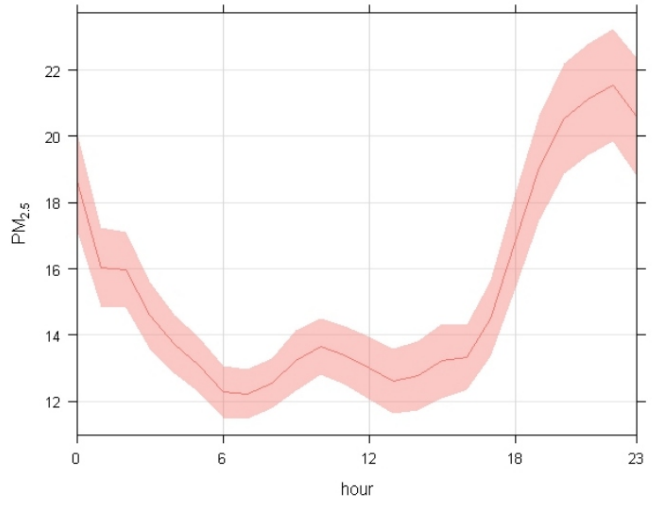

Figure 48 Variation of $PM_{2.5}$ concentrations (µg/m^3) by hour-of-the-day at the Lisburn site in Northern Ireland in 2009. The shading represents 95th percent confidence interval.

7.5.2 How does $PM_{2.5}$ vary by day-of-the-week in different parts of the UK?

The pattern of $PM_{2.5}$ concentrations at urban background sites by day-of-the week is summarised in Figure 49 for four geographic areas of the UK. It was broadly similar at all sites in 2009 (including roadside and industrial sites – not shown), with a steady increase during the week to give the highest concentrations on Thursday/Friday, before dropping to a low on Sunday. The increase in concentrations from Sunday to Friday is of the order of 4 µg/m^3 at all site types (including roadside and industrial – not shown). This pattern is also seen at a few sites for which $PM_{2.5}$ data were available in previous years, so it is not peculiar to 2009 (results not shown).

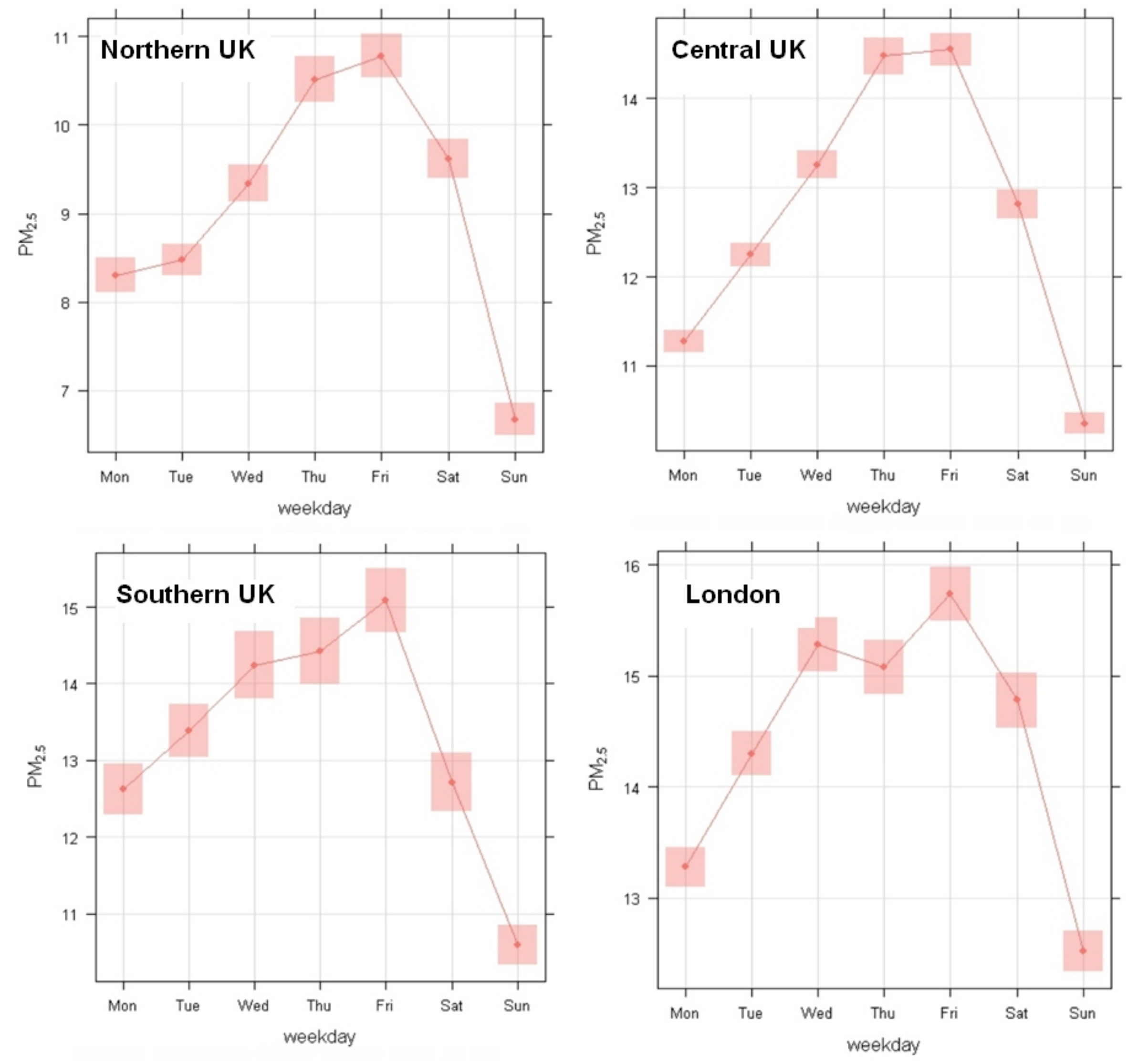

Figure 49 Variation of PM$_{2.5}$ concentrations (µg/m^3) at urban background sites by day-of-the-week in 2009 at sites in Northern UK (n=8), Central UK (n=17), Southern UK (n=8 and London (n=8). The shading represents 95th percent confidence interval. Note different scales.

The analysis has been extended to an examination of the pattern during periods with winds from the east (40°-140°), when long-range transport of secondary PM is important, compared with the pattern for all other wind sectors. An example is shown in Figure 50 for the site at Leamington Spa in the Midlands. The pattern of increasing concentrations during the week and lower weekend values is seen with both easterly winds and those from all other directions, although it is less clear with easterly winds.

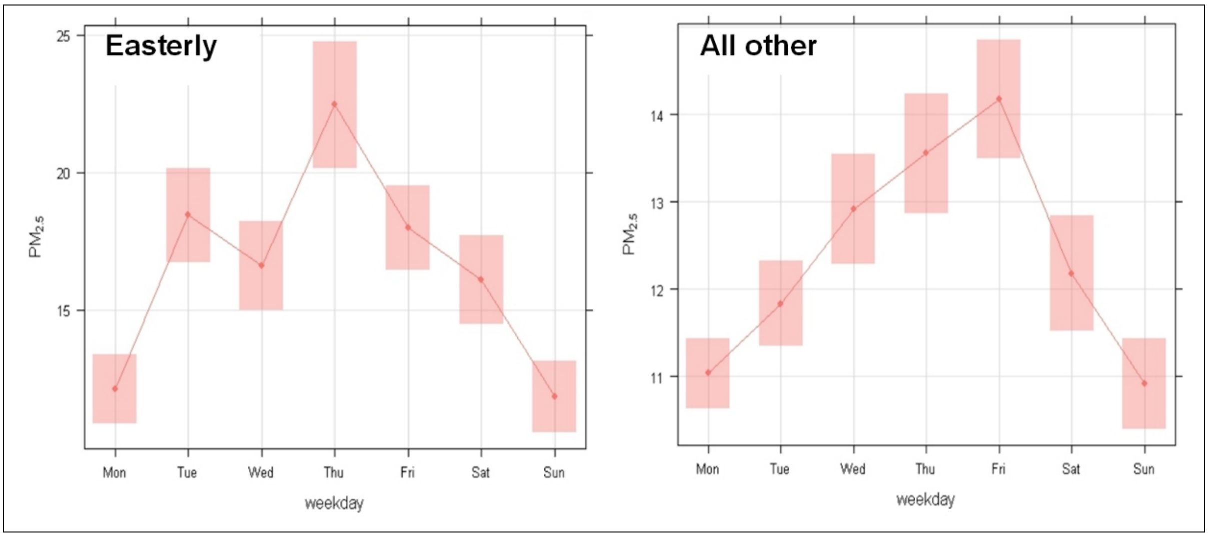

Figure 50 Variation of PM$_{2.5}$ concentrations (µg/m^3) by day-of-the-week at the Leamington Spa site in 2009 for easterly winds (40°-140°) and all other wind directions. The shading represents 95th percent confidence interval. Note different scales.

There is no clear explanation for this pattern of PM$_{2.5}$ concentrations through the week. The build up during the week would suggest a relatively slow response of the atmosphere, allowing concentrations to build up. However, the decline from Friday to Sunday would suggest a rapid response of the atmosphere to a reduction in emissions from traffic and industrial sources at the weekend. A slow response can be seen for pollutants such as ozone during photochemical episodes, when air follows a long trajectory over sources in Europe and ozone builds up from one day to another as precursor pollutants are added to the air mass. It is possible that secondary PM could also behave in this way, but this would only be during periods with trajectories passing for several days over source regions in Europe, which is not a frequent occurrence. Also such a pattern would be expected to persist over the weekend, and thus if it was the dominant cause of the steady rise during the week, then a sharp weekend decline would not be expected, as PM formed earlier in the week would still be transported into the UK over the weekend.

Jenkin *et al.* (2000) have examined temporal patterns in emissions from the UK, finding evidence of a slight increase in emissions of NOx, CO and VOCs from Monday through to Friday. This was related to the changing pattern of traffic emissions during the week, rather than emissions from other sectors. It may therefore be that the pattern observed for PM$_{2.5}$ is related to the changing emissions on different days of the week.

7.5.3 How does PM$_{2.5}$ vary by season in different parts of the UK?

The pattern of PM$_{2.5}$ concentrations by month-of-the-year is summarised in Figure 51 for urban background sites in four geographic areas of the UK. The pattern during 2009 was broadly similar at all sites (and at roadside and industrial sites – not shown), with the highest concentrations during the first four months of the year, the lowest concentrations during late summer (July-September), and somewhat higher concentrations during the autumn, early winter (October-December). This is a pattern that is similar to that found for other pollutants, such as NO$_x$. It will relate to greater emissions of both primary PM and secondary PM precursors during the winter, due to the higher heating load, as well as to reduced dispersion of local sources during the winter period. It will also relate, in part, to the loss of semi-volatile PM during summer months, which will be less prevalent during winter months. The range of monthly means is substantial, from 5 to 14 µg/m^3 in northern UK, 5 to 21 µg/m^3 in central UK, 8 to 20 µg/m^3 in southern UK and 10 to 22 µg/m^3 in London.

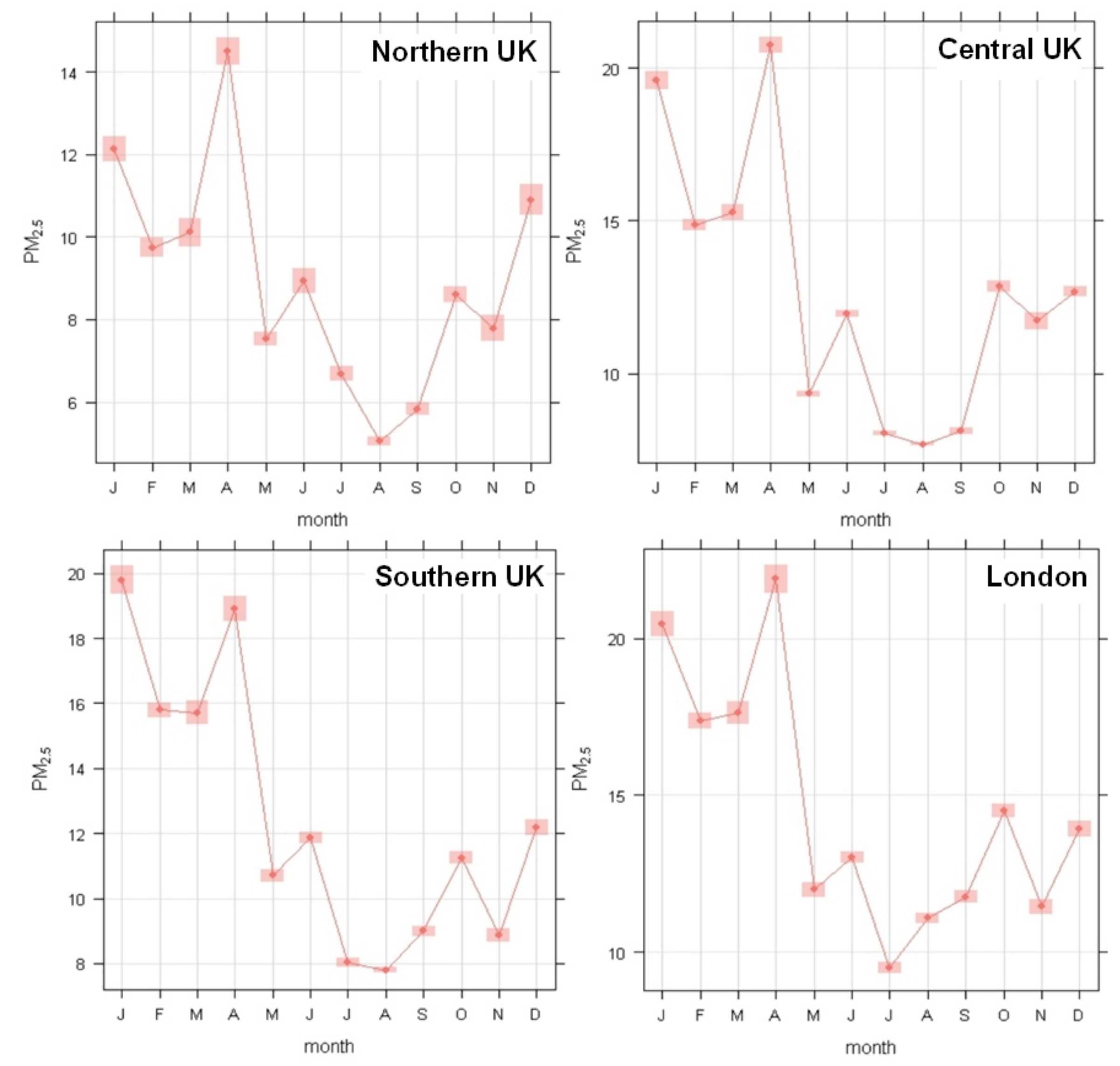

Figure 51 Variation of $PM_{2.5}$ concentrations (µg/m^3) at urban background sites by month of the year in 2009 at site in Northern UK (n=8), Central UK (n=17), Southern UK (n=8) and London (n=8). The shading represents 95th percent confidence interval. Note different scales.

7.5.4 How spatially correlated are $PM_{2.5}$ background concentrations?

An analysis has been carried out of the spatial homogeneity of $PM_{2.5}$ background concentrations. Hourly data for 11 sites within the Thames Valley, from Oxford through to Southend, but mostly within Greater London, have been correlated site by site. The correlation coefficients of all the pairs of sites have been plotted as a function of distances between the sites in Figure 52. This shows that hourly-mean data for sites within 20 km of each other are highly correlated (correlation coefficient >0.8). The correlation then declines with distance, but is still high at 100 km separation (correlation coefficient ~0.7). This is consistent with $PM_{2.5}$ being dominated by regional sources, including secondary PM, with local sources being less important.

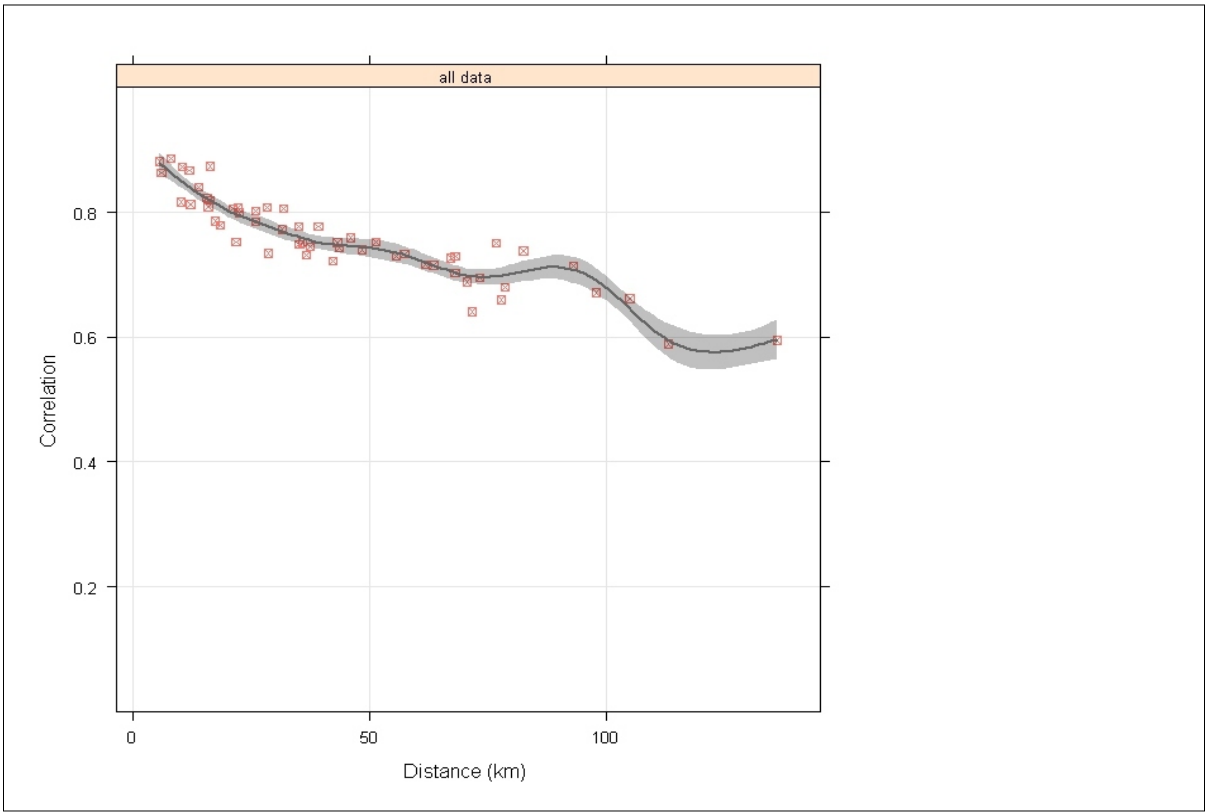

Figure 52 **Correlation coefficients of hourly-mean PM$_{2.5}$ concentrations (µg/m^3) at 11 urban background sites in the Thames Valley area of southern England as a function of distance between the sites. The shading represents 95th percent confidence interval.**

7.6 How do PM$_{2.5}$ Concentrations Vary by Wind Direction?

The relationship between PM$_{2.5}$ and wind direction can provide valuable insight into the sources of the measured concentrations. Two different types of plot relating concentrations to wind direction have been prepared using OpenAir. Polar plots relate concentrations to both wind direction and wind speed. Polar annulus plots show concentrations as a function of wind direction and time-of-day.

7.6.1 What is the relationship between PM$_{2.5}$ and both wind direction and wind speed?

The 2009 PM$_{2.5}$ monitoring data have been analysed in relation to wind direction at a number of sites in different areas of the UK. Eight meteorological stations have been used to provide data for the analysis:

- Belfast
- Birmingham
- Bristol
- Glasgow
- Heathrow
- Manchester
- Solent
- Southend

Polar plots have been produced for monitoring sites relatively near to these meteorological stations. A polar plot shows the measured concentrations by colour shading as a function of wind direction and wind speed. The wind speed increases from zero at the centre of the plot, to typically around 15 m/s at the edge. Two examples are shown in Figure 53, one for Manchester Piccadilly in the north of England, the other for

Reading in the south of England. Both plots show the highest concentrations associated with winds from the east, and in particular with higher wind speeds.

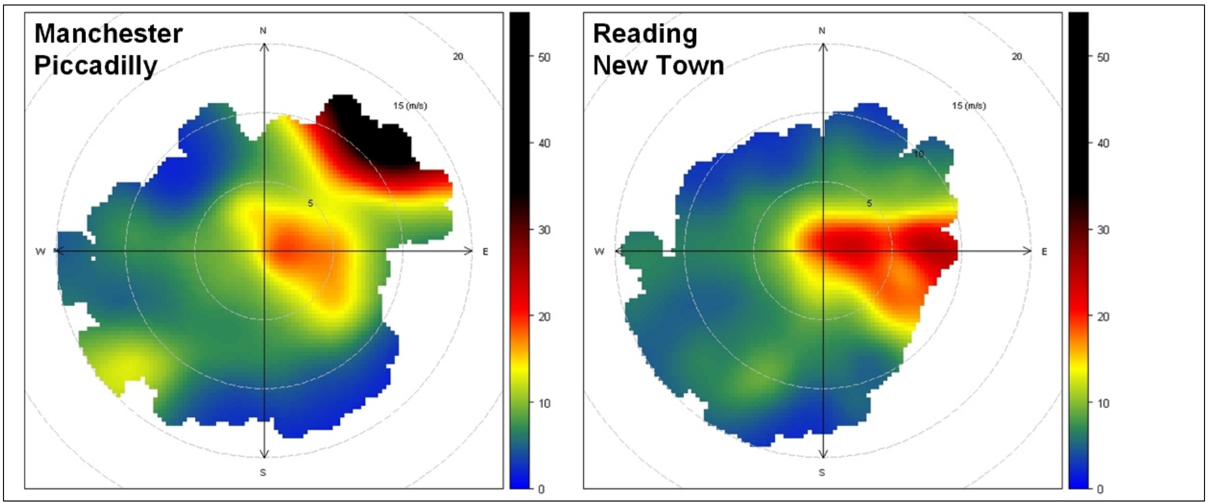

Figure 53 PM$_{2.5}$ concentrations (µg/m^3) at two sites in 2009 as a function of wind direction and wind speed. The circles are at 5 m/s intervals, with the outer limit at around 15 m/s.

To provide a clearer national picture, results for sites in northern UK are shown in Figure 54 and for southern UK in Figure 55. All plots are to the same scale. Annual mean concentrations fall in the green to yellow colour range. There is a remarkable consistency in the patterns across the UK. Concentrations are generally lower than the annual mean when winds are from the south-southeast clockwise through to north, while they are generally above the annual mean with winds from the northeast through to southeast. There is, however, a subtle difference between sites in the south and those in the north, with southern sites, from Cardiff to Southend and Birmingham to Portsmouth, having the highest concentrations associated with winds from the east through to southeast, while the northern sites, from Manchester to Edinburgh to Belfast, have a more significant component associated with winds from the northeast through to east.

The highest concentrations with easterly winds are mostly associated with the strongest winds (>10 m/s), although concentrations are elevated in this wind sector for all wind speeds. These results indicate that a significant source of PM$_{2.5}$ at all background sites will be emissions (mainly of precursors to secondary PM) within continental Europe. Easterly winds in the southern parts of the UK are frequently associated with a blocking high pressure over the Nordic countries that gives rise to an easterly or southeasterly air flow that will transport emissions from eastern Europe, northern Germany, the Netherlands and Belgium to the southern parts of the UK. In northern parts of the UK the air arriving from the east to southeast sector will not have passed over the same emission sources, hence the lower concentrations associated with these winds in northern UK. The high concentrations associated with more northeasterly winds in the northern parts of the UK are likely to arise when a low pressure runs up the English Channel, drawing air northward across European source areas, out into the North Sea, then round the top of the low pressure to reach the northern parts of the UK from a northeasterly direction. The association of the highest concentrations with higher wind speeds probably relates to the balance between greater dilution at higher wind speeds and the shorter transport times at these higher wind speeds, which allow less time for dispersion.

These observations reinforce the view that urban background PM$_{2.5}$ concentrations are dominated by regional sources, rather than local sources, and that PM derived from sources in Continental Europe, probably as secondary PM, plays a significant role in affecting concentrations in the UK.

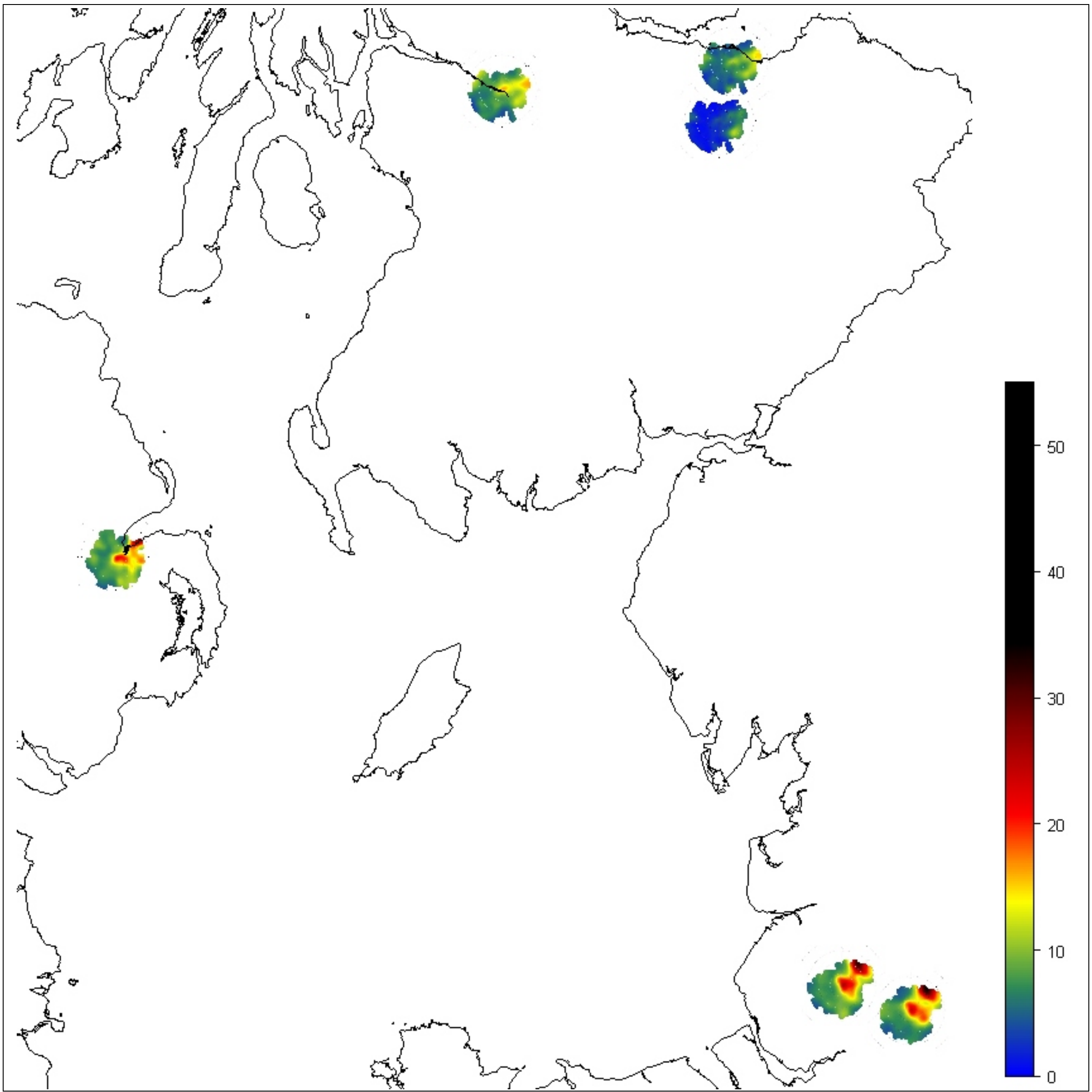

Figure 54 $PM_{2.5}$ concentrations (µg/m^3) at background northern UK sites as a function of wind direction and wind speed in 2009.

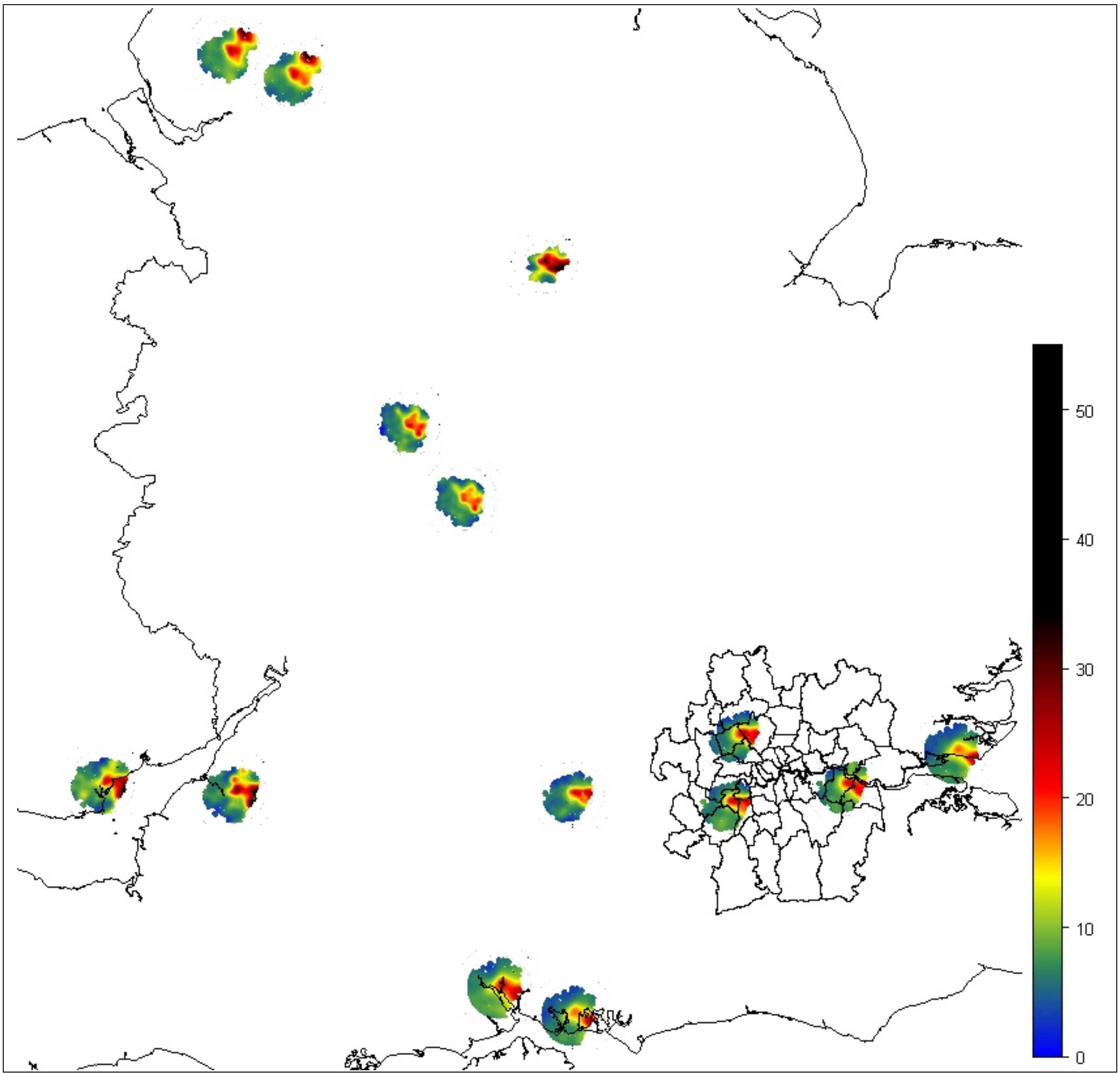

Figure 55 $PM_{2.5}$ concentrations ($\mu g/m^3$) at background southern UK sites, as a function of wind direction and wind speed in 2009. Note two sites in Manchester and Wigan repeated from Figure 54.

7.6.2 What is the relationship between $PM_{2.5}$ and both wind direction and time-of-day?

The 2009 $PM_{2.5}$ monitoring data have been analysed by wind direction and time-of-day at urban background sites in central London (London Bloomsbury) and in Southampton. The polar annulus plots are set out in Figure 56 and Figure 57, where concentrations are shown by both wind direction and time-of-day. The time-of-day runs from 00:00-01:00 h on the inside of the circle through the day to 23:00-24:00 h on the outside of the circle. The patterns for $PM_{2.5}$ and PM_{10} are broadly similar at both sites. NO_x concentrations, on the other hand, show different patterns at the two sites. The NO_x patterns are likely to relate to local traffic sources within a kilometre or so of the site. The peak NO_x concentrations are during the morning rush hour, and arise from the north and northwest at London Bloomsbury (possibly Marylebone Road / Euston Road), and from the south and southwest at Southampton (probably the A3024 Northam Road, located about 20 m to the south).

The $PM_{2.5}$ pattern is very different to that of NO_x, with the highest $PM_{2.5}$ concentrations being associated with winds from the east, a substantial proportion of which occur during the night-time hours, from around 20:00 to 01:00 h. There are also higher concentrations between around 04:00 to 10:00, with lower concentrations during the afternoon. This diurnal pattern of concentrations with easterly winds broadly matches the average diurnal patterns for all wind direction discussed in Section 7.5.1. The PM_{10} patterns are broadly similar to those of $PM_{2.5}$ at both sites, suggesting an important contribution of regional sources to background PM_{10} as well as $PM_{2.5}$.

It is also notable that there is only a limited increase of $PM_{2.5}$ concentrations associated with the higher NO_x concentrations at both sites, suggesting a minimal contribution of road traffic to $PM_{2.5}$ at urban background sites.

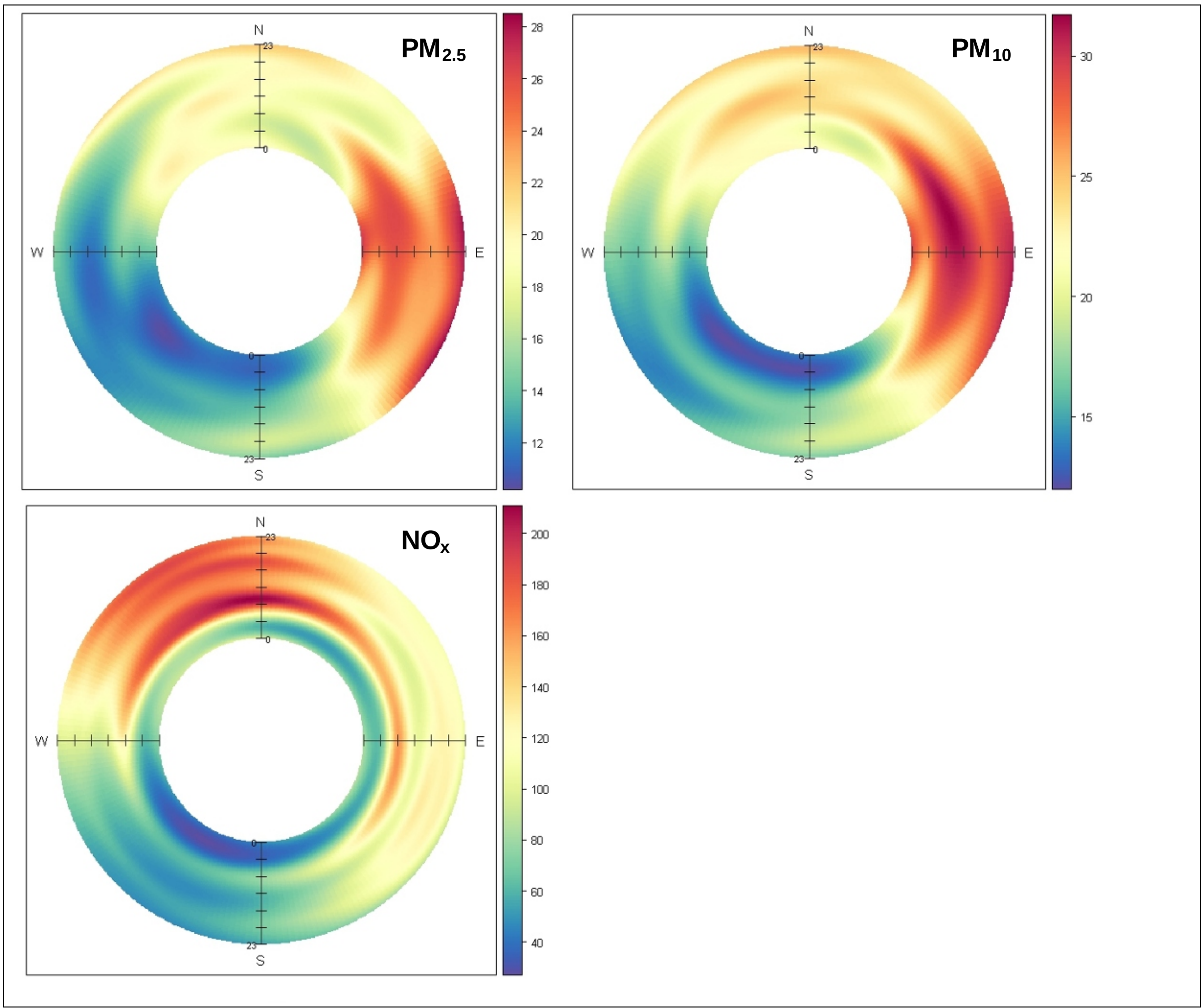

Figure 56 $PM_{2.5}$, PM_{10} and NO_x concentrations (µg/m^3) at London Bloomsbury urban background site as a function of wind direction and time of day. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00.

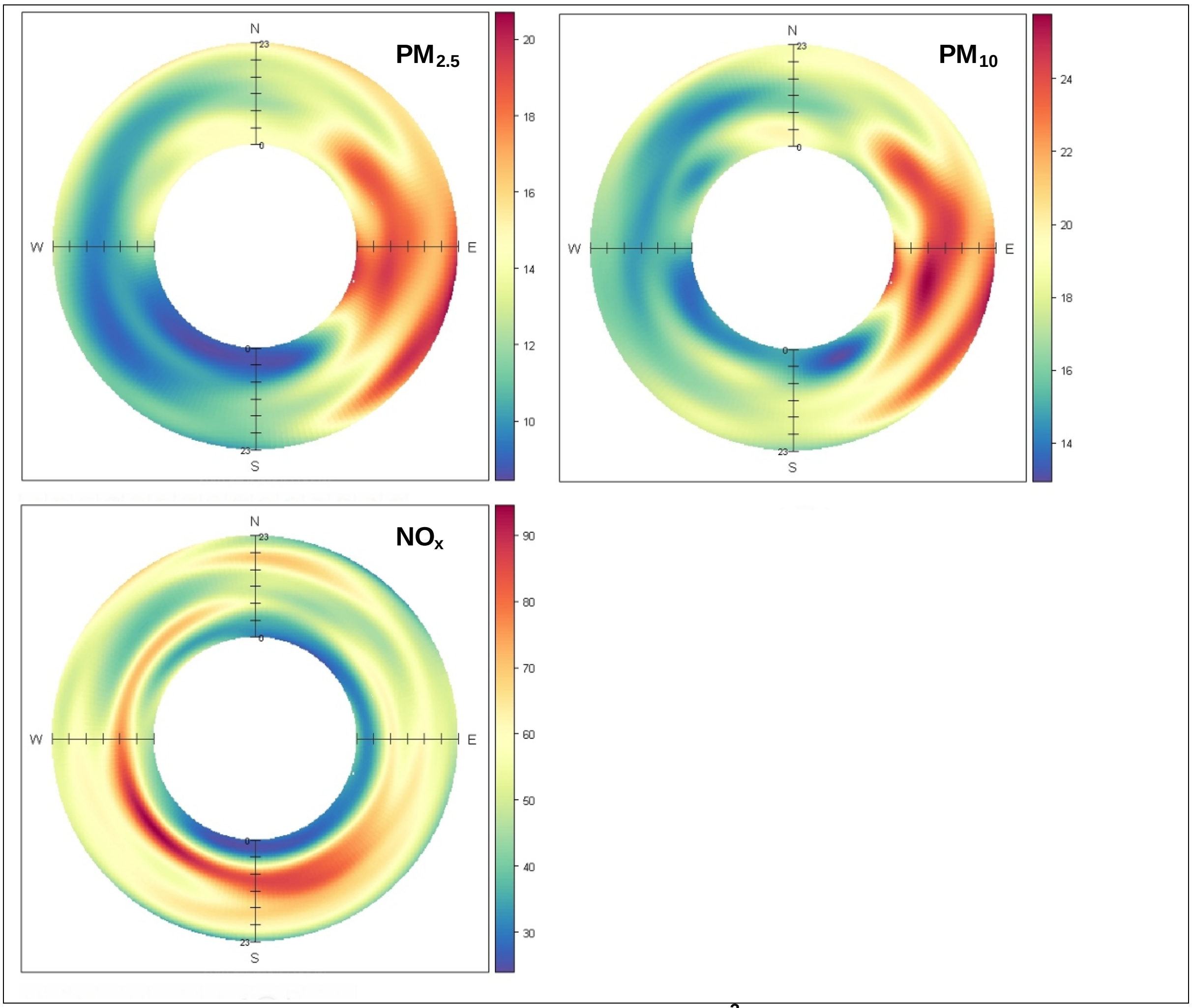

Figure 57 $PM_{2.5}$, PM_{10} and NO_x concentrations (µg/m^3) at Southampton urban background site as a function of wind direction and time of day. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00 h.

A clearer picture of the contribution road traffic to $PM_{2.5}$ at locations near to busy roads can be seen in the results for the roadside site Greenwich Burrage Grove, which is located around 15 m south of the A205 Plumstead Road in east London (Figure 58). The same predominance of sources to the east is seen, but there are also associations of $PM_{2.5}$ with road traffic contributions as shown by the matching of the $PM_{2.5}$ and NO_x patterns, when winds are from the northwest and north during the morning rush hour. The morning peak in NO_x is not as clearly defined in the $PM_{2.5}$ concentrations, which are more spread out through the day. This may be due to a proportion of $PM_{2.5}$ being related to resuspension, as during the rush hour, traffic speeds will be lower, giving rise to less resuspension, with the converse occurring later in the day. The concentrations related to the road are though much smaller than for those associated with the easterly winds.

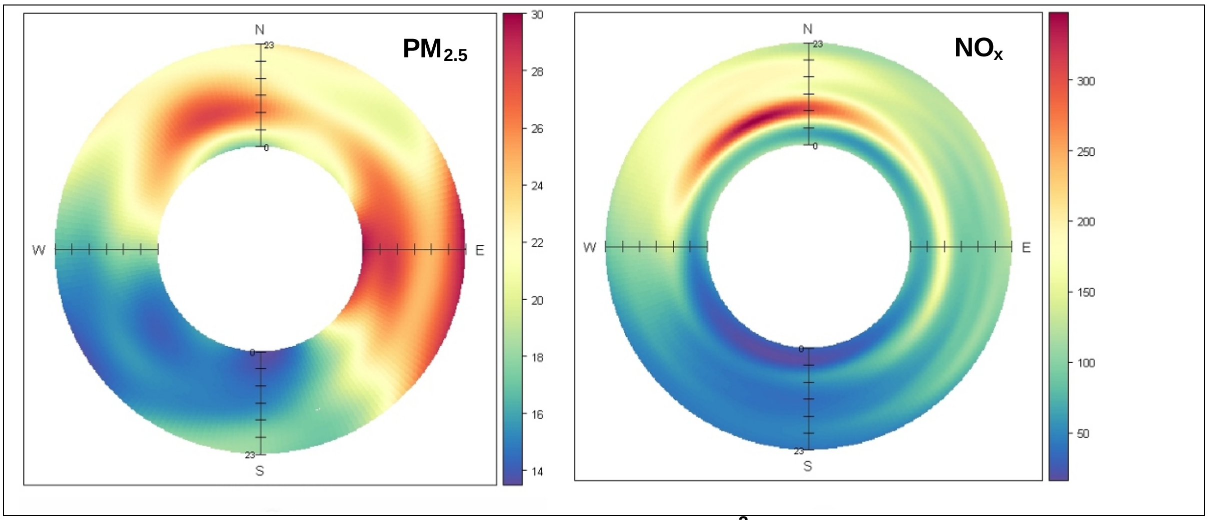

Figure 58 $PM_{2.5}$, PM_{10} and NO_x concentrations (µg/m^3) at Greenwich Burrage Grove roadside site, London as a function of wind direction and time of day. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00 h.

7.7 How have $PM_{2.5}$ Concentrations Changed Over the Last Decade?

No monitoring sites with long data runs (>5 years) using reference equivalent instruments for $PM_{2.5}$ have been identified. There are therefore no reliable trend data available for $PM_{2.5}$.

An indication of trends is, however, provided by the $PM_{2.5}$ monitoring that has been carried out at a number of sites in southern England using TEOM instruments over the last decade (Figure 59, Figure 60 and Figure 61). Data have been obtained for these sites from the national Air Quality Archive and from the London Air Quality Network, using the data retrieval tool within the OpenAir software package. These data are unadjusted TEOM values, as there has never been an empirical basis to provide a rough adjustment to a gravimetric equivalent (such as the default 1.3 factor applied to TEOM measurements of PM_{10} over many years). This means that they will not include any losses associated with the semi-volatile component, nor any trends associated with this component. The lines are fitted using the smooth-trend function in OpenAir, which fits a trend line to the monthly data. This option was fitted using the de-seasonalise function to improve the evidence of the overall trend. The band about the line shows the 95%ile confidence limits[29]. The following discussion of the trends is qualitative not quantitative, given the limitations of the data.

Results for two rural background sites, Harwell (west of London) and Rochester Stoke (east of London) are shown in Figure 59. They show no clear evidence of a downward trend, although the volatile component that is not captured by the TEOM instrument may have declined over this period. Results for the 3.5 years of monitoring at the Auchencorth Moss rural site in Scotland using a FDMS monitor confirm this picture for rural sites, as they show no change over the period December 2006 to August 2010 (Figure 62, Auchencorth Moss).

The results for two urban background sites in London show slight evidence of a small downward trend, of perhaps 2 µg/m^3 over the 10 years (Figure 60). There is clearer evidence of a downward trend at roadside sites, although the pattern at Marylebone road is not simple. It must be emphasised that these data can only be treated as indicative. Results are available from one AURN roadside site, Swansea Roadside, using the reference equivalent FDMS monitor for a period of almost 4 years, October 2006 to

---

[29] Further details available at http://www.openair-project.org/downloads/openair2010-05-26.pdf

August 2010. They also indicate a downward trend (Figure 62, Swansea Roadside), thus it is likely that there has been a real decline in $PM_{2.5}$ concentrations at roadside sites over the last decade. If road traffic emissions have declined, as this implies, then this would only be expected to be reflected to a small extent in urban background concentrations, since road traffic is a relatively minor contributor to urban background concentrations (see Section 4.1.2), although it is likely to have been larger in the past when exhaust emissions were higher.

These results provide evidence of a decline in motor vehicle emissions of $PM_{2.5}$ over the last decade, but no clear change in secondary $PM_{2.5}$.

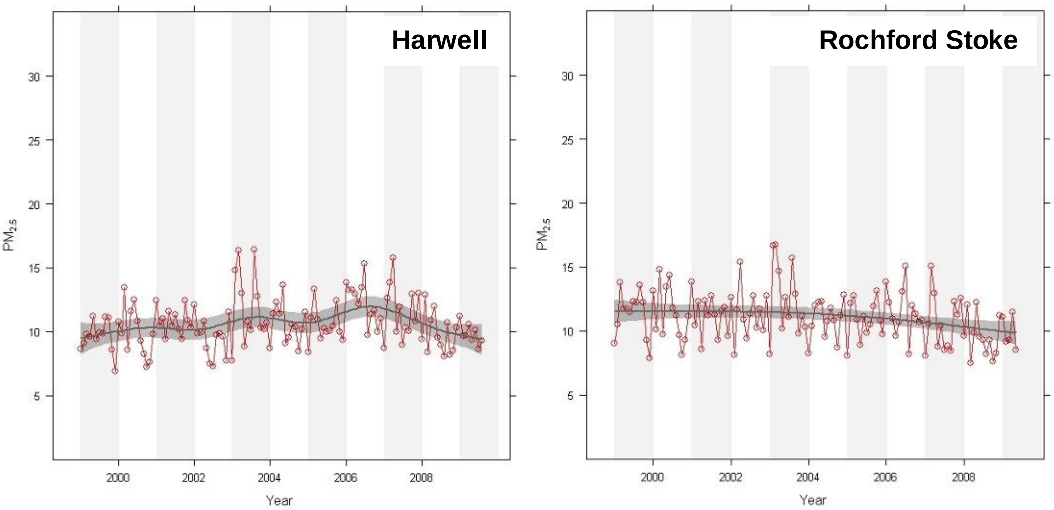

Figure 59 $PM_{2.5}$ concentrations (raw TEOM µg/m^3), 1999-2009, at rural background monitoring sites to the west and east of London. The shading represents 95th percent confidence interval.

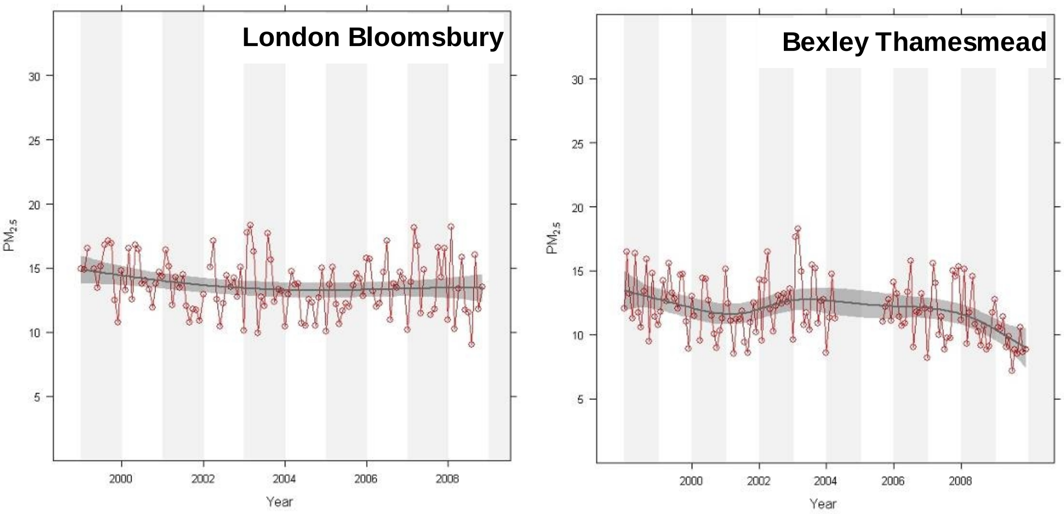

Figure 60 $PM_{2.5}$ concentrations (raw TEOM µg/m^3), 1998-2009, at urban background monitoring sites in London. The shading represents 95th percent confidence interval.

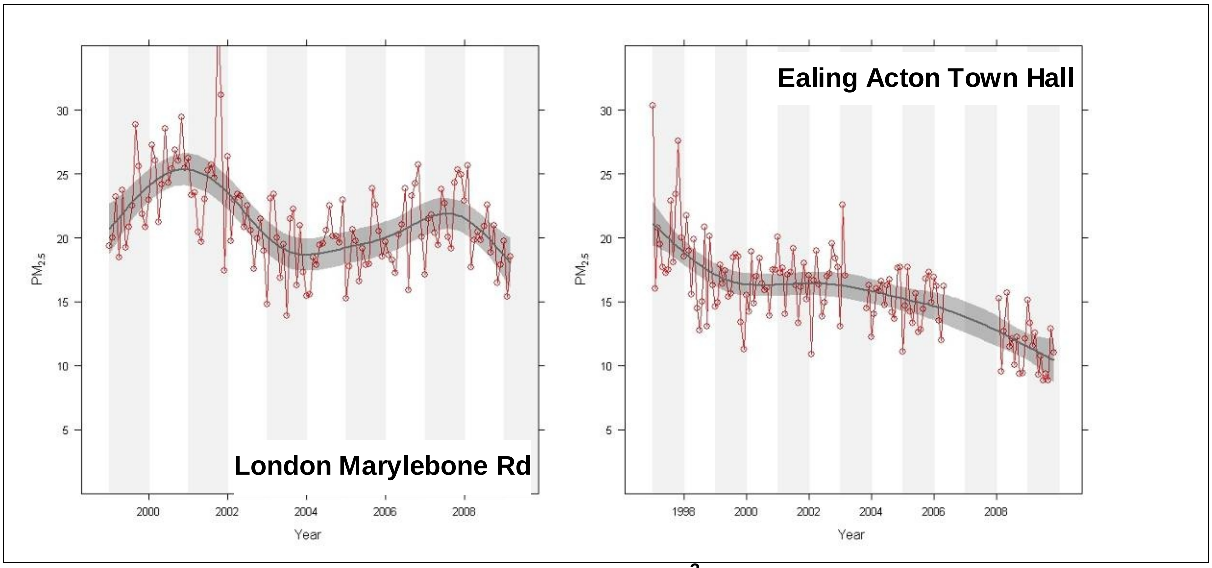

Figure 61 **PM$_{2.5}$ concentrations (raw TEOM µg/m^3), 1999-2009, at roadside monitoring sites in London. The shading represents 95th percent confidence interval.**

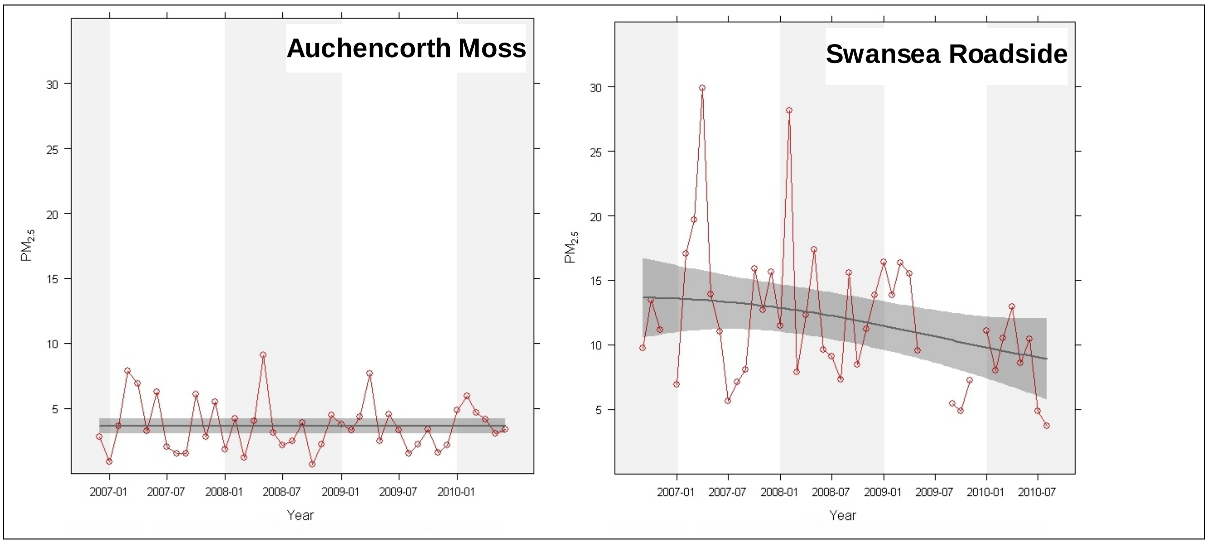

Figure 62 **PM$_{2.5}$ concentrations (FDMS, µg/m^3), 2006-2010, at Auchencorth Moss rural site in Scotland and Swansea Roadside site in Wales. The shading represents 95th percent confidence interval.**

7.8 What Overall Picture of PM$_{2.5}$ Arises from the Monitoring Data?

Monitoring data for PM$_{2.5}$ in the UK using reference equivalent instruments are still fairly limited, although this will change soon, as there has been a substantial increase in the number of monitoring stations across the UK.

Urban background concentrations of PM$_{2.5}$ across the UK fall within a relatively narrow range, generally from 12-16 µg/m^3. Rural concentrations are lower, but there are currently few rural monitoring stations and insufficient results are available to generate a robust national picture. There are, though, indications that rural concentrations range from 3-10 µg/m^3. Kerbside concentrations within 1 m of the kerb of busy urban centre roads are up to 7-8 µg/m^3 above the urban background. These roadside increments, however, decline rapidly on moving away from the edge of the road, and in most situations roadside concentrations will only be 1-2 µg/m^3 above the urban background. There is no evidence that individual industrial operations give rise to annual mean increments of $PM_{2.5}$ of more than a few µg/m^3.

Monthly mean concentrations at urban background sites show a substantial range, from a low of 5 µg/m^3 to a high of 14 µg/m^3 at northern UK sites, 5 to 21 µg/m^3 at central UK sites, 8 to 20 µg/m^3 at southern UK sites and 10 to 22 µg/m^3 at London sites, the lower concentrations occurring during the summer and the higher values during the winter. This contrasts with a much smaller diurnal range, which is typically 3 µg/m^3. Concentrations rise on average during the week to give the highest levels on a Friday, before dropping by around 4 µg/m^3 to their lowest levels on a Sunday. Currently there is no explanation for the rise during the week.

Analysis of the diurnal patterns of $PM_{2.5}$ concentrations and the patterns related to wind direction has provided strong evidence of a continental European source, probably secondary PM, with a limited role of local traffic sources. These findings are consistent with the source apportionment evidence set out in Section 4.1.2.

7.9 How do Monitoring Data Relate to Air Quality Objectives and Limit Values?

The EU limit values and targets for $PM_{2.5}$ set out in the CAFÉ Directive (2008/50/EC) are all based on annual mean concentrations (Table 1). The limit value is 25 µg/m^3 as an annual mean and it is to be met by 2015. This limit value is seen as a long-stop and is not designed to drive policy. All the evidence indicates that this concentration will not be exceeded in the UK. The highest roadside concentration in 2009 was 21 µg/m^3 at the Marylebone Road kerbside site in London (84% data capture). There is though a Stage 2 indicative limit value of 20 µg/m^3 to be achieved by 2020, which is currently being exceeded.

The EU Directive exposure-reduction target and exposure concentration obligation are based on the AEI. The AEI is an exposure index calculated as the average across UK urban background sites. The sites identified by Defra for calculation of this index, for which annual mean concentrations were available in 2009, based on >90% data capture, are shown in Table 3.1 in Appendix 3. The average for these sites was 13.2 µg/m^3. This is very close to the boundary between a requirement for a 10% or 15% exposure-reduction target. It is thus unclear at this stage what the EU target reduction for the UK will be, as the AEI is based on the 3-year mean for a larger number of sites than used in this analysis. A 10% target would take a 13 µg/m^3 AEI to 11.7 µg/m^3 while a 15% reduction would take the concentration to 11.0 µg/m^3.

Finally, there is the exposure concentration obligation within the Directive, which sets a ceiling of 20 µg/m^3 for the AEI, to be achieved by 2015, as a 3-year mean for the same set of urban background sites used to determine the exposure-reduction target.

The UK Government has set an annual mean objective for $PM_{2.5}$ of 25 µg/m^3, which applies at all relevant exposure locations in England, Wales and Northern Ireland from 2020. Given the discussion above, this is likely to be achieved throughout the UK. The Air Quality Strategy has also set an exposure-reduction objective, which in this case is a 15% reduction between 2010 and 2020. This will either be the same as, or possibly more stringent than the EU target, which will be either 10 or 15%.

In Scotland the annual mean objective for $PM_{2.5}$ has been set at 12 µg/m^3 to be achieved by 2020 at all relevant exposure locations. Given that the measured urban background concentration in Glasgow was 12 µg/m^3 in 2009, it is highly likely that the objective is currently being exceeded at some roadside sites in major urban areas in Scotland (perhaps by ~1-2 µg/m^3 at the building façades that represent relevant exposure). There is thus a risk that the Scottish objective for $PM_{2.5}$ may still be exceeded in 2020.

Given this analysis, it is expected that the exposure-reduction target will be the key driver for UK policy on exposure to PM$_{2.5}$, while the PM$_{10}$ objectives and limit values will supplement this control by driving policy on short-term (daily average) concentrations at hot-spots. There may also be a role for the annual mean PM$_{2.5}$ objective in Scotland driving policy in Scotland. However, the PM$_{10}$ objective is likely to be more stringent for these locations (see Section 7.9.1) and is thus more likely to drive policy.

7.9.1 How do the PM$_{10}$ limit values and objectives relate to PM$_{2.5}$ concentrations?

The 24-hour limit value and objective of no more than 35 days >50 µg/m^3 is taken to be equivalent to an annual mean PM$_{10}$ concentration of 31.5 µg/m^3. Given the ratios of PM$_{2.5}$:PM$_{10}$ identified for different parts of the UK in Table 21, it is possible to equate this limit value / objective to annual mean PM$_{2.5}$ concentrations (Table 22). The results in Table 22 show that the PM$_{10}$ 24-hour objective and limit value is more stringent than the annual mean PM$_{2.5}$ limit value of 25 µg/m^3, especially in northern UK areas.

Table 22 Indicative annual mean PM$_{2.5}$ concentrations that equate to the 24-hour PM$_{10}$ limit value and objective in different parts of the UK.

Region of UK	PM$_{2.5}$ (µg/m^3)
Scotland	17.3[a]
Northern Ireland	17.3
Northern England	20.5
Wales	20.5
Southwest and Central England	22.1
Southeast England and East Anglia	23.6

[a] In Scotland, this analysis only applies to the limit value, as a different objective applies.

In Scotland the PM$_{10}$ annual mean objective of 18 µg/m^3 is more stringent than the 24-hour mean PM$_{10}$ objective of no more than 7 days >50 µg/m^3. Given the average PM$_{2.5}$:PM$_{10}$ ratio of 0.55 identified for urban background sites in Scotland (Table 21), the annual mean PM$_{10}$ objective is equivalent to a PM$_{2.5}$ concentration of 9.9 µg/m^3. The PM$_{10}$ objective will therefore be more stringent than the PM$_{2.5}$ objective of 12 µg/m^3.

In summary:

- PM$_{2.5}$ limit values and the objectives for England, Wales and Northern Ireland should be met if the PM$_{10}$ 24-hour limit value and objective are met, with the one proviso that the PM$_{10}$ objective is not being met by measures that only focus on the coarse PM fraction (PM$_{2.5\text{-}10}$), as such measures would not necessarily reduce PM$_{2.5}$ concentrations. In Scotland the PM$_{2.5}$ objective should be met if the annual mean PM$_{10}$ objective is met, with the same proviso.
- The greatest policy driver for reducing PM$_{2.5}$ concentrations across the UK will be the EU and UK exposure-reduction targets for the AEI, a 10-15% reduction over the decade 2010 to 2020.

8. HOW AND WHY IS $PM_{2.5}$ MODELLED?

8.1 What is Meant by Modelling and Why is it Useful?

Modelling is taken here to refer to making predictions about conditions in the 'real world', based on intrinsic or empirically determined mathematical relationships. While in theory these calculations might be very simple, more typically they are complex, and are performed using specially-developed computer software. Modelling extends knowledge of the behaviour of $PM_{2.5}$ and its precursors and provides an important addition to the information obtained from measurement and monitoring programmes. In particular models have the following advantages:

- the ability to predict concentrations at a large number of receptors, providing more detailed spatial coverage than is generally possible with a monitoring network;
- the ability to model concentrations at locations where siting a monitor would be impractical;
- the ability to trace $PM_{2.5}$ concentrations, and $PM_{2.5}$ precursor concentrations/reactions, forwards or backwards in time in order to demonstrate the fate of emitted pollutants and the contributions made by different sources to ambient concentrations; and
- assessment of past and future environmental conditions, including simulation of the impact of potential policy implementations and other changes (for example, climate change).

8.2 Can Models Offer a Complete or Accurate Representation of Real-world Processes?

Different models have different levels of reliance on empirical data, and different levels of reliance on first principles. Where first principles are considered, it is important to recognise that atmospheric processes always contain more complexity than can be represented in a usable model. Simplifications are thus essential. Simplifications are sometimes also required because of a lack of understanding of the systems being characterised. The complexities that are omitted are often ignored or not recognised by users of models; but this is not of great concern if previous work has shown that they have a negligible effect on the system.

Models are always incomplete, and efforts to make them more complete can cause problems. Increasing the complexity of models can introduce more parameters with uncertain values, decreasing transparency and increasing overall uncertainty. It is sometimes preferable to omit capabilities that do not improve model performance substantially. Some complex models are characterised by substantial uncertainties because they contain more parameters than can be reliably estimated with the available observations. On the other hand, adding complexity can mean replacing arbitrary parameters with those that can be more closely tied to measurable processes (Derwent *et al.*, 2010).

Poor input data are often blamed for inaccurate outputs, but if the model being used is, itself, based upon poor input data, or flawed assumptions, then no amount of extra effort defining site-specific input parameters will significantly improve the model outputs.

Model outputs are thus inherently uncertain, but many modelling studies fail to quantify, or even acknowledge, this uncertainty. The reasons for this omission are typically: a) that an accurate quantification of the uncertainty is either beyond the technical scope of the study or impossible; and b) that stressing the uncertainty will detract from the clarity of any message being presented. This does not invalidate model results, but the strengths and weaknesses of model results should be recognised. It should also be appreciated that even where the overall accuracy and precision of a model is in question, this does not necessarily invalidate all of the results from that model; the model might still provide useful source-apportionment information, or be able to predict accurately the relative change in concentrations between two emissions scenarios.

8.3 How is Model Performance Evaluated?

The language used to describe the testing of model veracity is confusing and the same terms are often used to describe very different processes. In particular, the terms 'validation' and 'verification' are often taken to mean different things.

In a broader sense, 'validated' might be taken to mean that a model is fit for purpose. However, Derwent *et al.* (2010) cite a view that validation of models of natural environmental systems is impossible, since such models are frequently applied in new, untested, situations. Thus, a 'validated model' might be better described as 'conditionally-validated'.

Defra (2009), in its guidance to local authorities, describes model 'validation' as the general comparison of modelled results against monitoring data carried out by the model developers; while 'verification' is the process by which the model results are compared with local measurements in order to take into account local conditions. In this context, 'verification' implies a mistrust of the process of 'validation'. If a model was thoroughly and accurately validated, then verification would be unnecessary. Models typically used by local authorities (*e.g.* to predict local road traffic impacts) have consistently shown a need for local verification and adjustment; it is not uncommon to find that the models under-predict the road contribution by factors of four or more.

In these cases, Defra advises local authorities to adjust the road component of the model results to compensate for this under-prediction. Thus, the adjusted models work well at those monitoring sites used for verification. The adjusted model is then applied to other locations, under other conditions, with the hope that it performs equally well. Ideally there would be a further step to verification, where the adjusted model is compared against fresh monitoring data, but this step is very seldom practical and is not required by Defra. Further, since $PM_{2.5}$ monitoring data are seldom available within local study areas, the approach that is often taken is to adjust the $PM_{2.5}$ outputs using an adjustment factor derived for PM_{10}, or even for NO_x.

As an alternative to the terms 'verification' and 'validation', Derwent *et al.* (2010) propose the use of the term 'evaluation', which is defined as the assessment of the adequacy and correctness of the science represented in the model through comparison against empirical data from laboratory and *in-situ* tests and the analysis of natural analogues. Using Defra's language, this thus encompasses the 'validation' carried out by the model developers and the 'verification' carried out by model users.

Derwent *et al.* (2010) explain that there are four general questions to be answered when evaluating models:

a) does the model code contain errors such that it does not faithfully represent the model specification?
b) is the scientific formulation of the model broadly accepted and does it use state-of-the-art process descriptions?
c) does the model replicate observations adequately?; and
d) does the model reflect the needs and responsibilities of the model user and is it suitable for answering policy questions and fulfilling its designated tasks?

In terms of question (a), users of proprietary models can find that they give different results when nominally applied in the same way, for example there can be differences between receptor concentrations and contour concentration for the same location. Often, this can be shown to relate to imperfections in the way in which the model is coded, or packaged. While this is the responsibility of the model developers, model users can feel that basic quality assurance steps are left to them.

Model users often ignore question (b). If a model has been used and accepted before in a similar situation, then it is usually considered to be suitable.

The attention given to question (c) differs across different studies. As noted above, models of local road traffic impacts tend not to rely solely on model validation, but also include local verification against an independent monitoring dataset. Models of local industrial impacts, on the other hand, tend not to include this verification step, relying solely on model validation. This is appropriate as it is difficult to verify point source models, given the spatial uncertainty of the peak concentrations and the ability to locate the monitor at the point of the peak concentrations. Models of larger areas tend to be bespoke and thus comparison with measurements in the setting to which the model is applied is an integral part of developing the model.

Question (d) is sometimes taken out of the hands of model users, who are provided with a detailed specification and often a prescribed model. Derwent *et al.* (2010) provide a detailed and thorough review of how each of questions a-d are addressed in UK modelling.

Defra is currently undertaking an air quality modelling review. The aim is to evaluate which models are best suited to meet policy needs and which should be investigated and developed. This includes the application of the model evaluation guidance developed for Defra by Derwent *et al.* (2010).

8.4 Over What Spatial Scales are $PM_{2.5}$ Modelled?

Models which cover larger geographical areas tend to fulfil different purposes than those which cover smaller areas. There is no fixed delineation between one spatial category and another, but the following broad categories can be defined.

- **Regional** – This category encompasses continental to country-wide studies.
- **Urban** – This involves modelling most, if not all, emissions within a city or conurbation, with the regional background being added either empirically or from a regional-scale model.
- **Local** – This may range from an assessment of the impact of a single source on a single receptor a few metres away, up to complex road networks covering a few square kilometres. The results are added to the local background either from local monitoring or from urban- and regional-scale models.

These different scales are particularly important for $PM_{2.5}$, as it is both a primary and secondary pollutant. While the primary sources will usually require local-scale models, secondary sources will require regional scale models that cover the long-range transport of the precursor emissions, allowing the necessary time for the reactions that form secondary $PM_{2.5}$.

Generally, regional models tend to have less finely resolved outputs, predicting average concentrations across grids of a kilometre or more. For example, the EMEP4UK model currently simulates over a 5 km x 5 km grid. Near to emission sources, concentrations can vary considerably within just a few metres, so regional models are generally not appropriate to predict concentrations at specific locations near to emission sources. Regional models do, however, usually take into account more diverse sources and transformation mechanisms (*e.g.* chemical reaction schemes) than is the case with urban or local models. Some regional models also explicitly simulate synoptic-scale meteorological and atmospheric transport processes. The results from regional models are often useful when applying urban or local models (for example results from the UK

Pollution Climate Mapping (PCM) model form the local background in many local-scale modelling studies).

At the other end of the spectrum, local models will often be run without taking any account of chemical transformations or depletion from deposition, and often without explicit inclusion of any emission sources other than those within a very small domain. An advantage of local models is that they consider more detailed local dispersion, thus allowing concentrations at individual receptor locations to be predicted. This is necessary when assessing against the limit values and thus these models tend to be used to assess the impacts of individual permits or planning applications.

Urban-scale models fill the middle ground between these two extremes. They often use the same dispersion algorithms as local models, but take account of more diverse emission sources and sometimes include functions for chemical transformations and depletion by deposition. Emissions from roads and small point sources are usually treated as area sources.

Until recently, there was no single model that could be applied on all three scales. Some models (*e.g.* CMAQ) now claim to allow this ‘one atmosphere’ approach, but their application in this way is still at an early stage and not well established in the UK. Typically, finer spatial resolution is achieved by ‘nesting’ one model within another, so that, for example, regional processes are counted within one ‘module’ and local processes are addressed in another. The Environment Agency is currently evaluating the use of CMAQ. The application of CMAQ to power station emissions is discussed in 8.12.3.

There is a further category of sub-models or sub-routines, which have no spatial delineation. These include modules such as the Plume Rise Model Enhancements (PRIME) model that is used in some US Environmental Protection Agency (EPA) dispersion models, or stand-alone models such as Defra’s Emission Factor Toolkit (EFT), which calculates traffic emissions for input to dispersion models.

8.5 What are the Main Purposes for which $PM_{2.5}$ is Modelled at Each Spatial Scale?

The reason *why* $PM_{2.5}$ is modelled should be central to *how* $PM_{2.5}$ is modelled, and the reason why $PM_{2.5}$ is modelled varies between the three defined spatial scales.

Regional-scale modelling is carried out for a variety of reasons. The UK PCM provides UK-wide predictions of annual mean concentrations for EU reporting purposes. It is also used to support local-scale modelling exercises by providing background concentrations. Other regional-scale modelling is not carried out specifically to characterise $PM_{2.5}$ concentrations but may nevertheless be relevant. For example, the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model was developed to focus on the transport and deposition of ammonia. While an understanding of ammonia concentrations is relevant to aspects of $PM_{2.5}$ concentrations, FRAME has been extended to cover PM explicitly.

Urban-scale modelling characterises concentrations across city-wide areas. This may be used to assess against exposure-reduction targets or to assess the impacts of large-scale schemes (for example the London Low Emission Zone).

The purpose of **local-scale** $PM_{2.5}$ modelling is usually to determine the impact of emissions from one or more local sources on nearby receptors. This might be for permitting, planning application, or other purposes, such as in response to specific individual concerns. Assessment is likely to take account of the EU limit values, and will often focus on the relative magnitude of the process or local source contribution in relation to these limit values.

8.6 Over What Temporal Averaging Periods is $PM_{2.5}$ Modelled?

Modelling may be carried out using meteorological data based on anything from one hour (*e.g.* ADMS) to annual (*e.g.* FRAME) averaging periods. For regional models the key advantage of using a one-hour timescale is the ability to track pollutants more accurately and to allow atmospheric processes to be incorporated, for example the influence of the diurnal photochemistry cycle. This can then be used to test the ability of the model to replicate measured spatial and temporal patterns, such as those identified in Section 7 for $PM_{2.5}$, which will give confidence in the ability of the model to replicate real-world conditions (or highlight weaknesses of the model). On the other hand, the advantage of modelling longer time intervals is the reduction in model run times.

Modelling for $PM_{2.5}$ will frequently be driven by the requirement to predict concentrations that can be related to the air quality standards. For $PM_{2.5}$, the relevant standards all relate to either a full calendar year or a 3-year average. Thus, while predicting short-term (24 h or less) concentrations can often be of interest, predictions for calendar years are those of most policy relevance.

8.7 How are Emissions from Outside a Model Study Area Handled?

At **regional scales**, most emission sources are explicitly included (*i.e.* input directly into the model), although they are sometimes aggregated before being used as input. Emissions from outside regional study areas are accounted for either through existing monitoring data (as in the case of the UK PCM model) or from other models (*e.g.* FRAME UK incorporates the contribution from FRAME Europe).

At **urban scales**, the majority of $PM_{2.5}$ emission sources within the model domain are likely to be included explicitly, either as discrete individual sources, or as aggregated area sources. There will still, however, be a contribution from sources outside the study area. These sources are often taken into account using data from rural monitoring sites (*i.e.* the ‘rural background’). Figure 63 shows a schematic of a typical urban-scale model.

At **local scales**, only nearby sources tend to be included explicitly. The contribution from sources not included explicitly in the model is termed the ‘local background’. The total predicted concentration is the sum of the explicitly modelled local contribution and the local background concentration (Figure 64).

The local background will be made up of all primary and secondary $PM_{2.5}$ from sources not included explicitly, which may range from domestic sources to the contribution from long-range transport. The background is relatively spatially homogenous over at least a 1 km x 1 km area. It can thus be taken from local monitoring at background sites, or from the results of regional models. For existing sources, unless modellers employ some mechanism of removing the modelled local contribution from the local background, there will often be a degree of double-counting, since the modelled sources will also be included in the local background. Generally, this is not a significant issue, since the contribution of sources modelled at this scale to local background concentrations tends to be very small.

Adding the background to the model outputs is usually done as a post-processing step. Some models allow the background contribution to be added within the model interface, but these models do not apply $PM_{2.5}$ background concentrations in a complicated manner and the routine remains: total concentration = local contribution + local background.

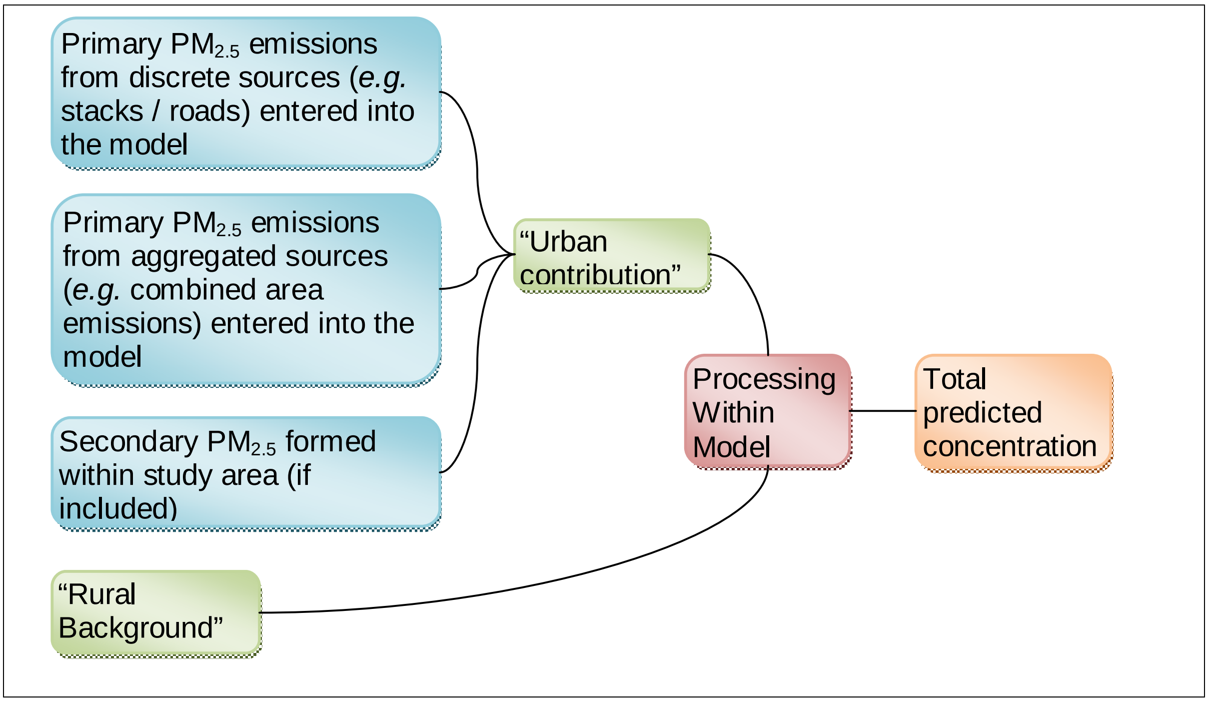

Figure 63 **Role of rural background concentrations in urban-scale modelling.**

Table 23 summarises some typical results from several recent local-scale road-modelling exercises carried out by Air Quality Consultants Ltd. It shows that local sources tend to contribute less than half of the total predicted $PM_{2.5}$ concentration, and often less than 10%, even at roadside receptors.

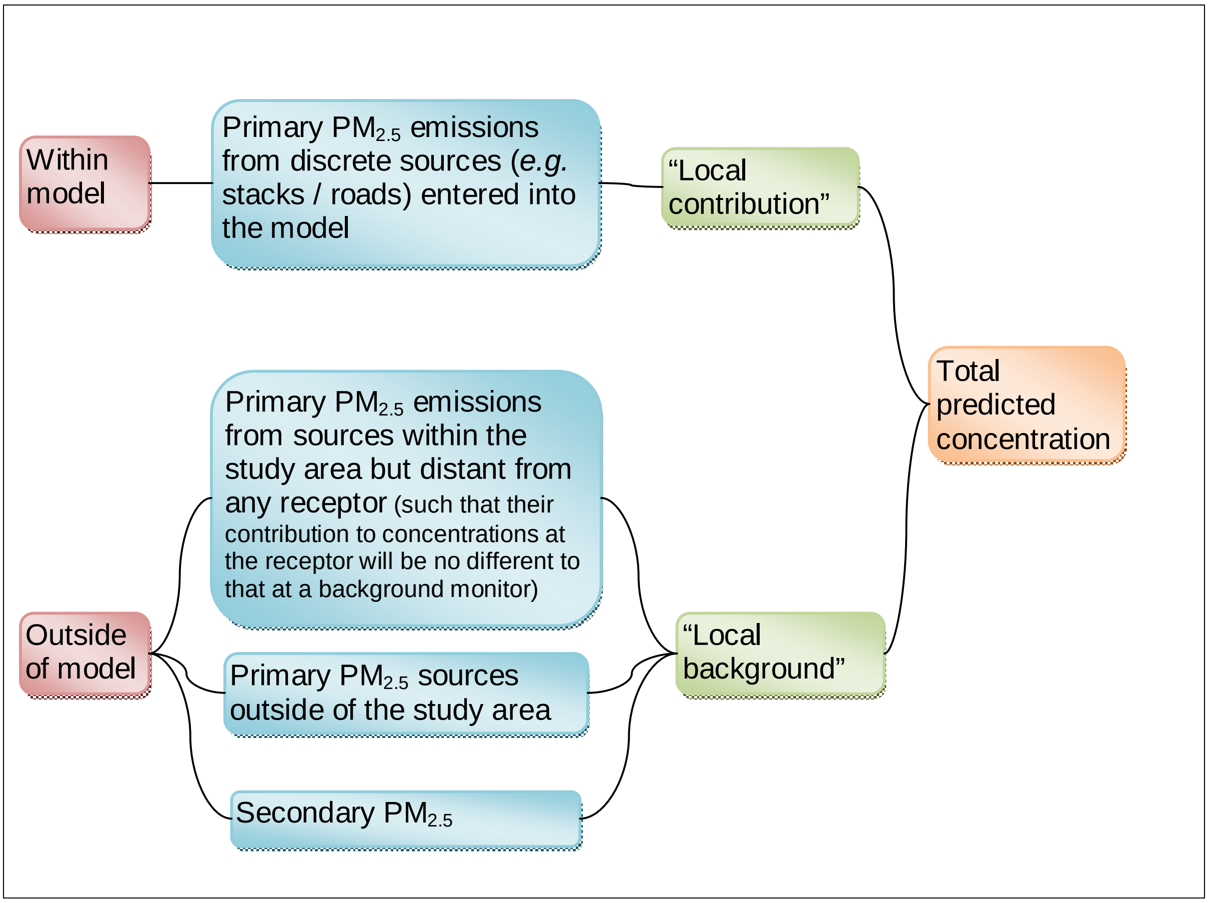

Figure 64 **Role of local background concentrations in local-scale modelling.**

Table 23 Typical local $PM_{2.5}$ model results at roadside receptors and the relative contributions from local and local background sources.

Location	Maximum Predicted Annual Mean $PM_{2.5}$ Concentration (mg/m^3)	% Local Contribution	% from Local Background
Northampton	11.3	5.3	94.7
Surrey	12.0	7.6	92.4
Suffolk	14.0	3.0	97.0
Bridgend	14.2	22.3	77.7
Greenwich	15.4	9.7	90.3
Cambridge	15.8	24.7	75.3
Lincoln	24.7	49.4	50.6

8.8 Which Models are Typically Used at Each Spatial Scale?

A very large number of different models can be, and are, used internationally to model ambient pollution concentrations on the different spatial scales considered here. Not all of them are of particular relevance to this review. Appendix 7 gives a brief summary of some common models, with some of the key features and limitations of each model set out in Tables A7.1 and A7.2.

At **regional scales**, atmospheric transport models can be broadly grouped into two types: Eulerian and Lagrangian. In Eulerian models, the calculation of physical and chemical variables is undertaken simultaneously for all the grid points in the model domain. With a Lagrangian approach, calculations are made along a defined trajectory that a parcel of air is assumed to follow. Large numbers of trajectories (typically tens of thousands) are required to generate statistically significant results. A major difference between the Eulerian and the Lagrangian approach is that, whilst calculations in Lagrangian trajectories are independent, the calculations at the grid locations of an Eulerian model are inter-dependent.

ROTAP (2009) uses a further distinction between regional scale models: ‘simple’ and ‘complex’. Examples of simple models include FRAME and HARM. These models use relatively simple chemical schemes, assume straight line trajectories and are driven by annually-averaged statistical meteorology. Their main advantage is a fast simulation time, which allows them to be applied to source-receptor calculations (involving hundreds of model simulations) and uncertainty analyses (involving thousands of model simulations), as well as the ability to be used with finer grid resolutions covering extensive domains. Examples of complex Eulerian models include EMEP4UK and CMAQ. These models include more complex chemical schemes and require detailed meteorological information. Such models are able to simulate short-term pollution events and the detailed interaction between pollutants and meteorology (ROTAP, 2009).

Models used at **urban-scales** may be packaged products, sold for application to many different situations (*e.g.* ADMS Urban and AIRVIRO), or they may be bespoke models built specifically to characterise air quality across a particular urban area (*e.g.* the London Air Quality Model). Typically, the dispersion algorithms are the same as those applied at local scales, but include a greater variety of emission sources. Urban-scale models will often include emissions near to a modelled receptor with a high degree of spatial resolution, while away from receptors emissions may be aggregated and entered as area sources.

Local-scale models often rely on Gaussian (*i.e.* assumptions of normal 'bell-curve' distributions) or modified Gaussian equations to describe the dispersion away from the source. Emissions may be treated as steady-state or as a series of instantaneous 'puff' releases. Emissions are typically entered as line (*e.g.* for a road), point (*e.g.* for a chimney), or area/volume sources (*e.g.* from a car park), although other permutations are possible. Emissions may be assigned an initial velocity, or temperature (which relates to plume buoyancy).

Even though local-scale models are often applied to satisfy the requirements of regulatory agencies, there is no definitive guidance on which models should be used. It is thus left to modellers to argue the validity of the model that they have chosen. The most common dispersion models in use in the UK are ADMS and AERMOD. AIRVIRO is also often used at local scales, even though the spatial resolution of its outputs makes it not ideal for this purpose.

8.9 How do Models Treat Future-Year Projections?

The physics and chemistry characterised in models does not change when modelling future conditions. The emissions environment is, however, assumed to change. Possible changes in the meteorological environment may also be taken into account. In local and urban models, concentrations will typically show a linear relationship with emissions. Regional models which account for chemical reaction schemes in $PM_{2.5}$ formation are unlikely to show a linear response between $PM_{2.5}$ concentrations and emission of any one precursor species.

Future projections of emissions may come from extrapolating measured trends, but more often from the anticipated influence of known or predicted policies or patterns. One example of this is the UK road vehicle EFT, which assumes year-on-year changes in vehicle fleet composition (associated with the uptake of new vehicles), and combines this information with statistics on emissions from vehicles meeting different European type approval emission standards.

8.10 What Sort of Information on Source-Attribution can Modelling Provide?

In terms of local and urban models, which do not consider secondary $PM_{2.5}$ formation, the level of source-attribution detail is only limited by the resolution of the model input data. It is thus, for example, possible to isolate the impact of any individual source for which emissions can be individually characterised.

Results from the UK PCM model have been discussed in Section 4, where typical background concentrations and the contribution of various sources to these totals are described. The results presented in Table 23 all take their local background concentration from the UK PCM model, together with the additional contribution from local sources.

In regional models where $PM_{2.5}$ formation can be studied, it is possible to analyse the impact of individual gaseous precursors on ambient $PM_{2.5}$ concentrations. Simulations conducted with CMAQ have considered the influence of abatement of gaseous emissions on particulate formulation. The biggest effect of the chemical interactions was found to be where a 30% reduction in NH_3 emissions implied a more than 30% reduction in fine particulate NO_3^- concentrations, a greater change than when NO_x emissions were reduced by 30%. This suggests that in some regions of the UK, NH_3 may have a limiting effect on formation of NH_4NO_3 aerosol from the chemical equilibrium between gaseous NH_3 and HNO_3 (ROTAP, 2009).

8.11 What Steps are Followed in a Typical Local Modelling Study and What Data are Needed?

Figure 65 outlines the typical steps that are followed for a simple point source modelling exercise, while Figure 66 does the same for a road traffic assessment. One key difference between the two approaches is the requirement for emissions data. In the case of a point-source assessment, modellers would typically attempt to obtain emission rates from the site operator (or in the case of a planning or permitting application, from the applicant). In such cases, the emission rates might come from on-site monitoring, or from the design specifications of the plant. If the operator or applicant was unable to provide these data, emission rates might be estimated from, for example, the EMEP/EEA Air Pollution Emissions Inventory Guidebook (EEA, 2009), or the study might be based on the emission limits, thus demonstrating the maximum impact that a facility might have (assuming the emission limits were not breached). For a road traffic assessment, emissions are typically calculated (either within or outside of the modelling software package) from traffic flows using the UK road vehicle EFT.

Suitable maps are key requirements of modelling studies. Near to an emission source, the contribution of the source to ambient concentrations can vary considerably within a metre, and this defines the requirements for map resolution (*i.e.* the position of a receptor will often need to be determined with sub-1m precision). Ordnance Survey ‘mastermap’ (or ‘landline’) data show this level of detail but are expensive (often more than consultants charge for carrying out a modelling study). Because of this cost, modellers often make do with maps that are less than ideal, inferring crucial distances from other information sources. Recent moves by Ordnance Survey to provide free open access to some of its data are unlikely to change this position, as the maps being made available are of insufficient resolution.

Terrain data are typically only included if the gradients of the terrain are more than 10%, since it is usually accepted that terrain has a minimal impact on modelled concentrations below this value (Defra, 2009). Other geographical information will typically be taken from architects’ drawings, site inspections, and aerial photography.

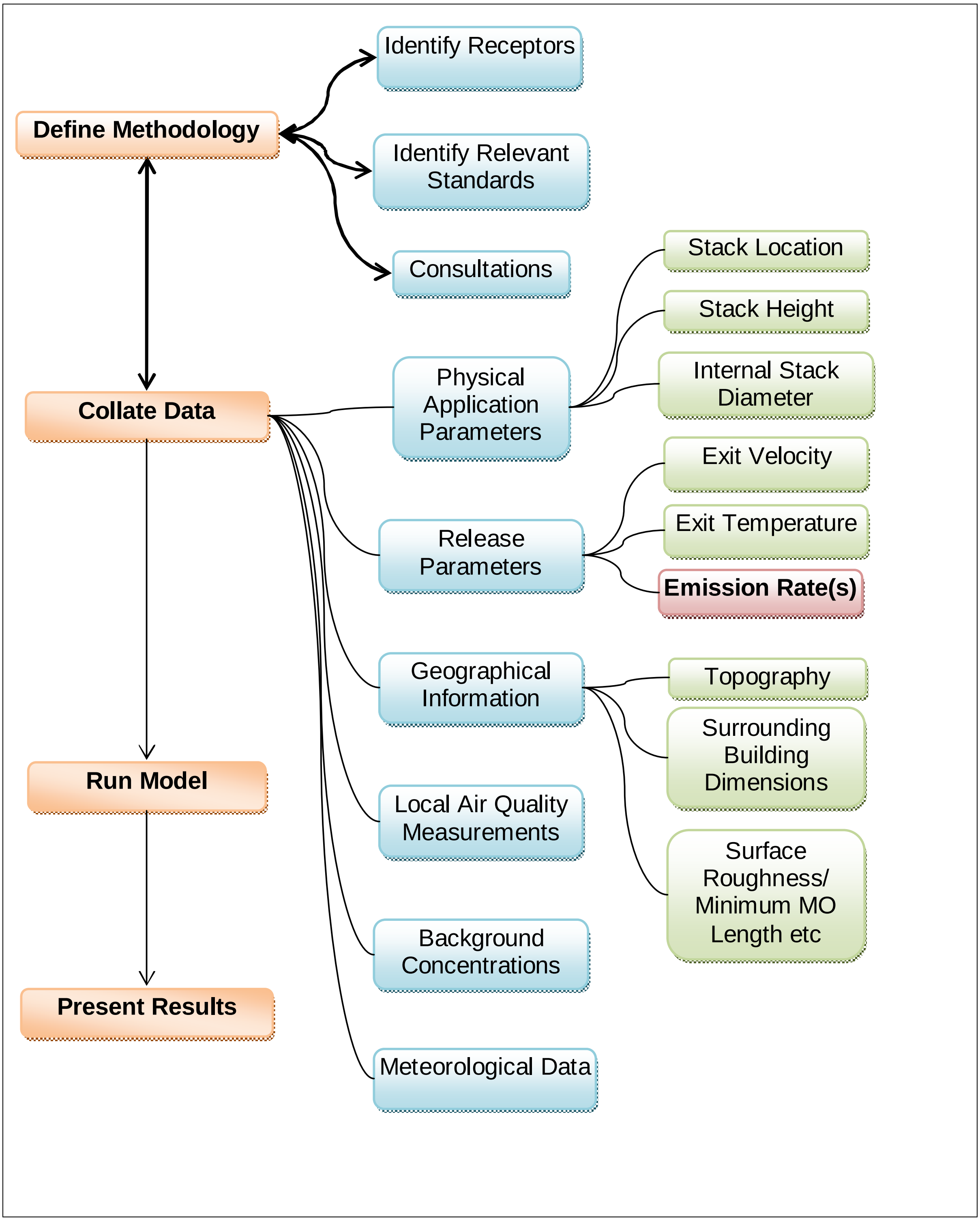

Figure 65 Steps taken in a simple industrial point-source modelling exercise

Meteorological data are usually purchased on a study-by-study basis and are obtained as hour-by-hour data for a full calendar year. For industrial applications, data for three or five years are typically used, but as the data are expensive it is common practice for road assessments to rely on data for just one year. When using more than one year's worth of data, modellers will typically present only the worst-case impacts, but this is often easier said than done. One problem is defining a worst-case impact (*i.e.* is it the highest predicted concentration, the highest absolute increase, or the highest percentage increase). Another problem is that different meteorological files will give different impacts at different receptors over different averaging periods (*i.e.* the worst-case years are usually different for annual means and 1-hour maxima). Modellers writing reports for public consumption will typically try to summarise this plethora of information to make it easier for non-specialists to digest, and this can be a source of tension between modellers and regulators, as the latter are looking for all the details to be provided.

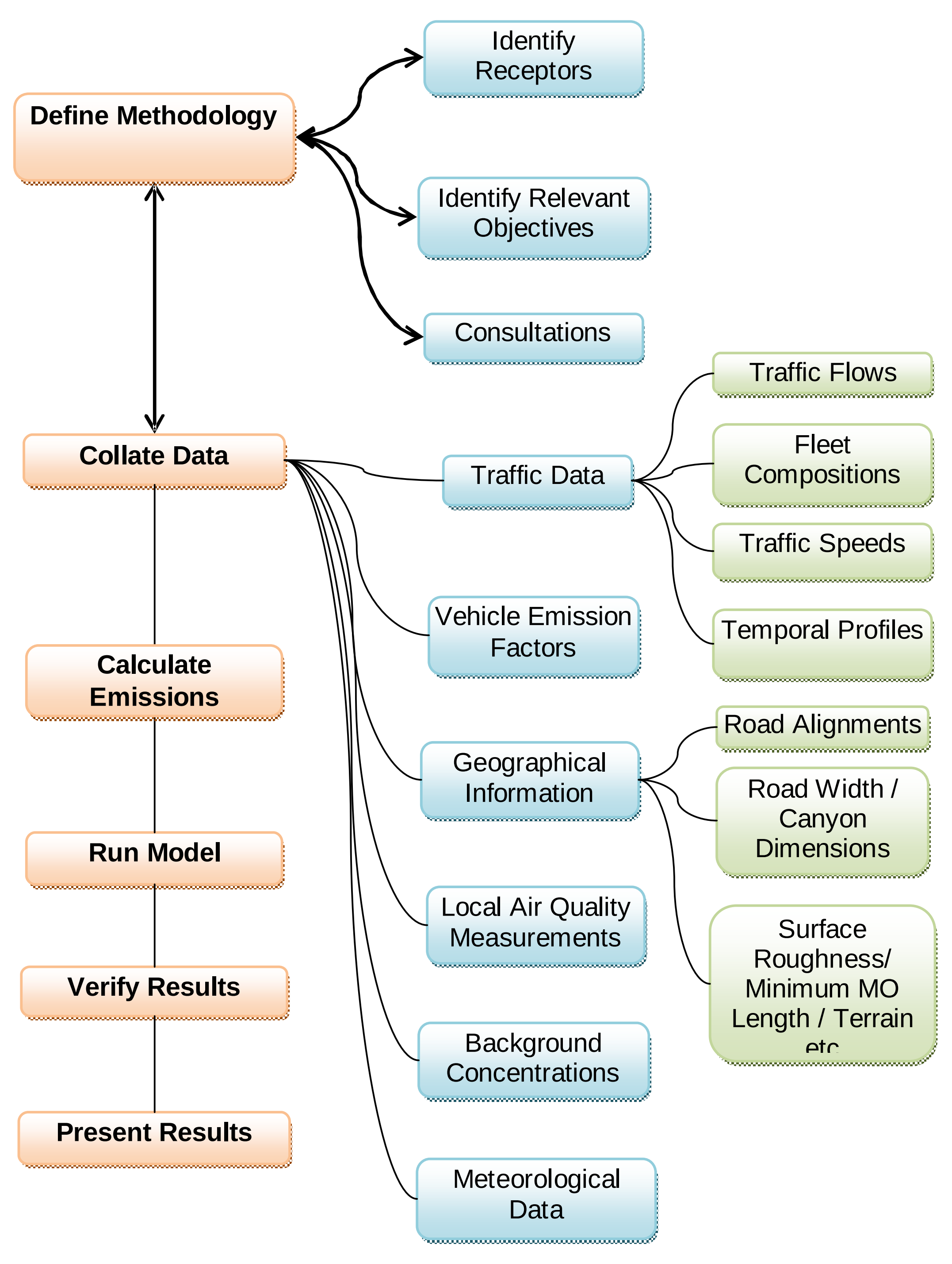

Figure 66 **Steps taken in a road-source modelling exercise**

Background concentrations are taken either from local monitoring, or from the UK PCM model published by Defra.

As mentioned in Section 8.3, another key difference between industrial stack modelling and roads modelling is model verification. It is usually found that roads models under-predict concentrations. As a result, road models are usually run to predict concentrations at a nearby air quality monitoring site, or better still several sites, and adjusted accordingly. The adjustment is almost always to increase predicted concentrations. Where $PM_{2.5}$ data are unavailable, adjustment factors will often be derived for PM_{10} or NO_x and applied to $PM_{2.5}$. This local 'verification' step is not usually feasible for industrial point source modelling and is thus omitted, *i.e.* it is assumed that the models are presenting results without bias.

8.12 What Examples are There of the Application of Modelling to $PM_{2.5}$?

8.12.1 Example study of road traffic emissions

Modelling of annual mean concentrations of $PM_{2.5}$, PM_{10} and NO_2 was carried out across Weymouth. The aim of the assessment was to test proposed traffic interventions, although only the baseline results are discussed here. Local road traffic sources were modelled using ADMS-Roads. All of the roads shown in Figure 67 were included in the model. The Simulation and Assignment of Traffic to Urban Road Networks (SATURN) traffic model was run for morning peak, evening peak, and inter-peak periods, with the results output as individual turning movements for each junction shown in Figure 67. The modelled flows were combined into annual averages using factors provided by transport engineers. Diurnal flow and speed profiles were calculated by combining the local predictions with national average hour-by-hour profiles published by the Department for Transport (DfT). It is interesting to note that the Paramics micro-simulation model had also been run for sections of the network, but only for a few discrete hourly periods. Because of this limitation, it was concluded that the micro-simulation traffic model results could not be used for annual mean pollution modelling.

Emissions were calculated using the UK road vehicle EFT (v4.1). Emissions were calculated on a link-by-link and hour-by-hour basis, by entering each link-hour into the EFT separately. Near to junctions, the road network was split into a large number of short sections. Predicted peak-hour speeds on these sections were often less than 5 km/h – the minimum speed at which the DfT's vehicle emission factors can be applied. For these links, the additional queue time was calculated and an emission from idling vehicles was derived by extrapolating the DfT emission factors (in g/s) to zero km/h. The speed-related link emission was then augmented by the time-based queuing emission.

Background concentrations were taken from the 1 x 1 km maps of UK background concentrations generated by the PCM model. Since local roads had been included explicitly, local roads were removed as a component within the background maps. The model was run using one complete year (2009) of hour-by-hour meteorological data from the meteorological office station at Bournemouth Airport. Surface roughness and minimum Monin-Obukhov length (a height scale related to atmospheric stability) were set at 0.5 m and 30 m respectively. A separate diurnal profile was assigned to each link, which was calculated as the variation in emissions rather than flows, taking account of link-specific flow and speed profiles.

Neither $PM_{2.5}$ nor PM_{10} were measured within the model domain and so the model was not verified for these pollutants. The model was, however, run to predict the road contribution to annual mean NO_x concentrations at fourteen passive diffusion tube locations where NO_2 was measured. Defra's NO_x to NO_2 calculator (Defra, 2010c) was used to calculate the local road increment to measured NO_x at each diffusion tube. Modelled road NO_x was then compared with measurement-derived road NO_x. The model showed an under-prediction and, as a result, the local road component of NO_x was multiplied by 1.6. Following this adjustment, the model predicted the NO_2 measurements without bias, and at thirteen of the fourteen diffusion tube locations the model results were within 25% of the measurements. Since no verification was possible for $PM_{2.5}$, the modelled road component of $PM_{2.5}$ was also multiplied by 1.6.

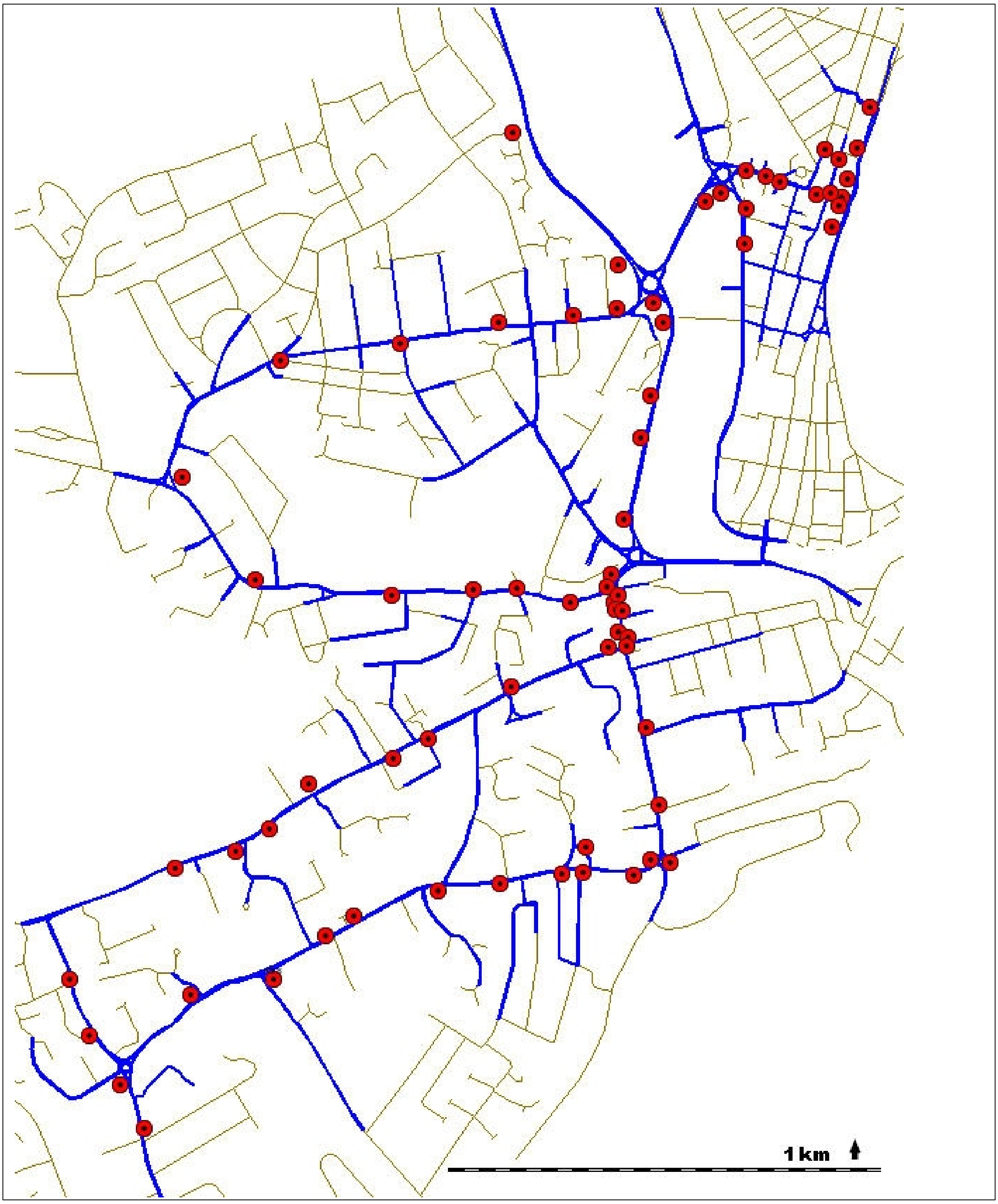

Figure 67 Road links and receptors included in Weymouth modelling exercise.

Concentrations were modelled at 70 discrete receptor locations (Figure 67) representing worst-case residential exposure. Modelling was carried out for the year 2009. Background annual mean $PM_{2.5}$ concentrations ranged from 8 $\mu g/m^3$ to 9 $\mu g/m^3$ in 2009, while total modelled $PM_{2.5}$ concentrations ranged from 9 $\mu g/m^3$ to 13 $\mu g/m^3$. Assessment against the 25 $\mu g/m^3$ limit value showed that $PM_{2.5}$ concentrations were not of concern. For comparison, modelled annual mean PM_{10} concentrations ranged from 14 $\mu g/m^3$ to 18 $\mu g/m^3$, while modelled annual mean NO_2 concentrations ranged from 10 $\mu g/m^3$ to 45 $\mu g/m^3$. The latter were compared against the UK objective of 40 $\mu g/m^3$. Thus, modelled NO_2 concentrations exceeded the objective, while $PM_{2.5}$ concentrations were just over half the limit value.

Contours of annual mean $PM_{2.5}$ concentrations for the study area are shown in Figure 68.

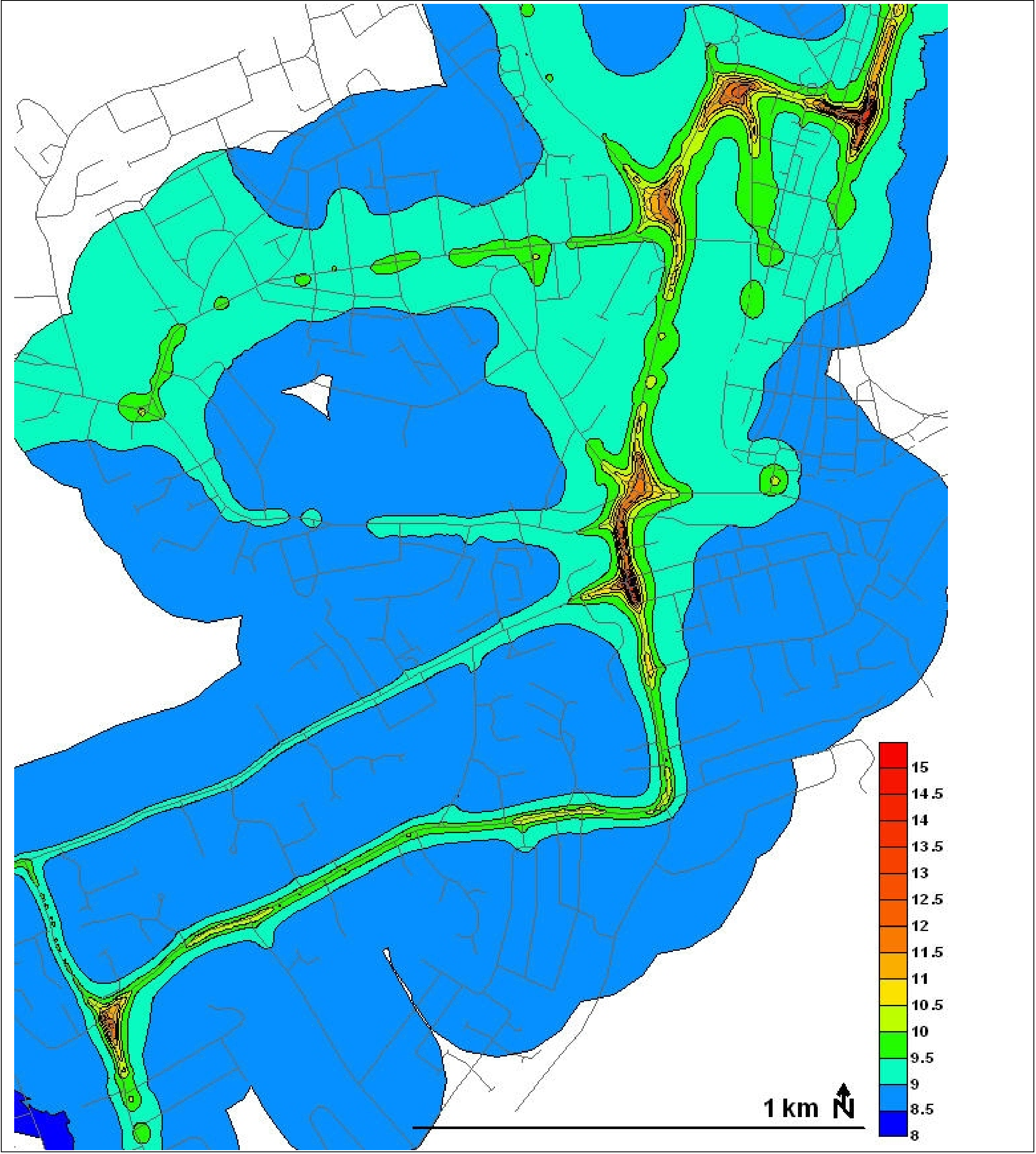

Figure 68 Annual mean $PM_{2.5}$ contours (mg/m^3) alongside road network.

8.12.2 Example study of industrial stack emissions

The proposed facility was near to protected habitats (Special Areas of Conservation (SAC) and Sites of Special Scientific Interest (SSSI)) in Scotland, as well as near to a town. There are no relevant standards for $PM_{2.5}$ concentrations over ecological sites and thus the town was of principal concern for this pollutant. In the immediate vicinity were a school and a large number of residential properties. Consultations were held with the Scottish Environment Protection Agency and the local authority. The principal concern was not $PM_{2.5}$, but the effects of NO_x emissions on ecosystems and human receptors. However, $PM_{2.5}$ was included in the assessment, for completeness.

The model used was AERMOD PRIME. The assessment required detailed plans of the proposals, showing the locations and heights of each building near to the stack. A range of stack heights was considered in order to determine the optimal configuration. The developer provided all required physical input parameters specific to the site, but site-specific release parameters were not available. Instead advice on stack exit temperature and exit velocity was sought from the equipment manufacturers. Since specific emission rates were unavailable, it was assumed that the plant would operate constantly at the maximum permissible emission limits. In the absence of size-speciated emissions data, the PM emission limits were assumed to represent $PM_{2.5}$. The parameters used for the modelling are summarised in Table 24.

Table 24 Model input parameters used in example industrial stack assessment.

Parameter	Value
Stack diameter (m)	1.7
Stack height (m)	45
Exit velocity (m/s)	12.3
Exit temperature (°C)	146
Pollutant	**Emissions Limits (mg/Nm^3)**
NO_x (as NO_2)	400
PM	50
SO_2	200
Pollutant	**Emission Rates (g/sec)**
NO_x (as NO_2)	9.92
PM	1.24
SO_2	4.96

The site was near to some steep terrain and so a terrain file was purchased, along with suitable base maps. The surrounding buildings were entered into the model in order to account for building downwash, and an appropriate value of surface roughness was entered into the model, taking account of the surroundings.

$PM_{2.5}$ concentrations were not measured locally, so annual mean background concentrations were taken from the UK PCM model. The model was run using five years of hour-by-hour meteorological data, purchased for a nearby site. The results were inspected, and those for the year of meteorological data which (on balance) gave the largest impacts were selected for inclusion in the report. The model was not verified against local data.

The model was run to predict concentrations at 11 discrete receptor locations, as well as across a polar grid of 14,000 receptors covering 5^o sectors in 15 m increments out to 3 km from the source. The maximum predicted annual mean $PM_{2.5}$ concentration at a location with relevant exposure was 8.1 $\mu g/m^3$, of which 8 $\mu g/m^3$ came from the local background and 0.1 $\mu g/m^3$ came from the proposed plant. The contribution of the plant across the modelling domain is shown in Figure 69. The maximum impact was just over 0.7 µg/m^3 to the northeast of the proposed plant, but this did not represent a location with relevant exposure. Similar elevated concentrations were also predicted further from the point of emission, on the elevated ground forming the valley edge.

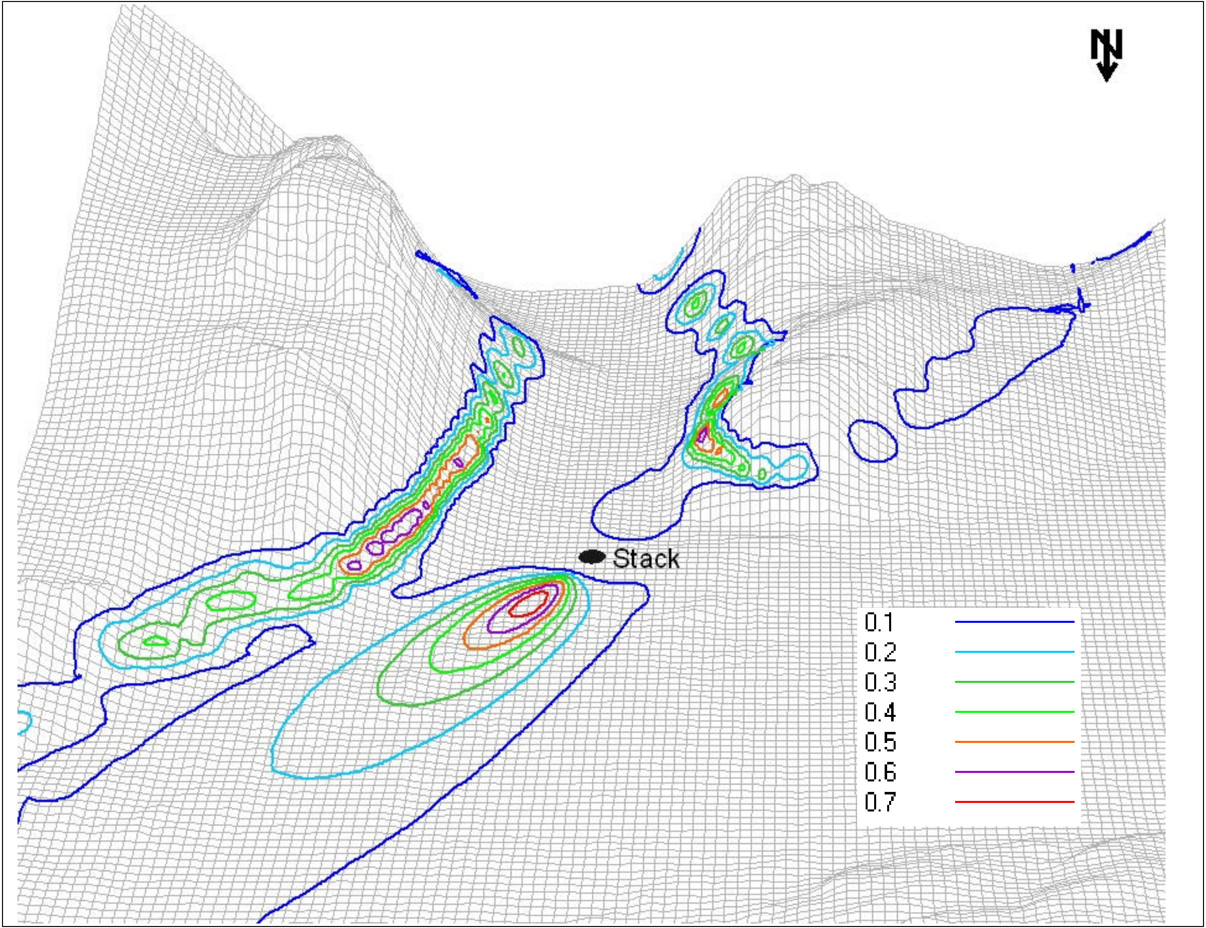

Figure 69 Contours of plant contribution to annual mean PM$_{2.5}$ concentrations (mg/m^3) on topographic map of the area around the stack. The view is looking south, with a 100 m horizontal grid.

8.12.3 Example study of power station emissions

Chemel *et al.* (2010) have applied three models, CMAQ, TRACK-ADMS and FRAME, to an evaluation of regional air pollution arising from a power station. The CMAQ model was applied at a resolution of 5 km. The TRACK-ADMS modelling used the TRACK model for the long-range impacts, at a resolution of 20 km, and ADMS for the short-range, at a resolution of 1 km. The FRAME model had a 5 km resolution. However, this model was not applied to PM predictions. The CMAQ model (both versions 4.6 and 4.7) was used in a mode that mixed emissions instantaneously in the entire 5 km grid cell. CMAQ has a Lagrangian plume-in-grid option to provide better spatial resolution, but this was not used, as it is no longer supported in version 4.7. The models were run at an hourly time resolution for a number of species, although consideration is only given here to the PM results. The modelling covered 2003 emissions from a coal-fired power station located in the South-East of England.

The PM$_{10}$ results were compared with measurements made at AURN background sites. It was found that both CMAQ and TRACK-ADMS under-predicted annual mean concentrations of all pollutants, including PM$_{10}$. The calculated maximum percentage contributions of the power station to regional annual mean PM$_{10}$ concentrations ranged from 3.0% for CMAQ v4.7 to 10% for TRACK-ADMS. It was found that the impacts predicted by CMAQ are more localised than for TRACK-ADMS. None of the models was found to provide the overall best performance. The authors concluded that simple modelling systems, such as TRACK-ADMS are attractive for source receptor calculations involving a large number of model calculations, as run times are faster than those of advanced systems such as CMAQ.

As noted, the models predicted a maximum power station contribution of some 3-10% of background PM_{10} averaged over a 5 x 5 km grid square. This equates to an annual mean contribution from the power station of around 0.5-1.5 $\mu g/m^3$, assuming a background of around 15 $\mu g/m^3$. Chemel (2010) also provides information from model runs specific to $PM_{2.5}$. The power station contributed around 2-8% of the background $PM_{2.5}$ concentration averaged over the 5 x 5 km grid square with the maximum impact. This equates to an annual mean $PM_{2.5}$ contribution averaged over the 5 x 5 km grid square of around 0.2-0.6 $\mu g/m^3$.

8.12.4 Example application of the Photochemical Trajectory Model

Derwent *et al.* (2009) describe the application of the Photochemical Trajectory Model (PTM) to a study of non-linearities in the system of secondary PM response to changes in precursor emissions. They used the NAME model to generate 96-h air trajectories arriving at Harwell, in Oxfordshire, i.e. the path followed by an air parcel over the 96 hours before it reached Harwell. Use was made of 30 randomly generated trajectories for each day of 2006. Emissions along the route of each trajectory were generated from the gridded emissions within the European Monitoring and Evaluation Programme (EMEP) inventory, together with isoprene from natural biogenic sources, while UK emissions were input from the 10 x 10 km emission grid produced as part of the NAEI. The gas-phase chemistry was based on the Carbon Bond Mechanism, while the formation of PM was based on a highly simplistic chemical kinetic approach, applied to the inorganic NH_4NO_3 – HNO_3 – NH_3 system, and to the oxidation of biogenic compounds. The authors note that the approach to the formation of secondary organic aerosol represented a preliminary first attempt.

The model outputs were compared with measurements made at Harwell. The modelled annual mean $PM_{2.5}$ of 10.6 $\mu g/m^3$ compared closely with the measured value of 10.5 $\mu g/m^3$. However, the ability of the model to predict daily concentrations was not as good, especially during the summer months. Ten out of 12 months had monthly means within a factor of 2 of the observations.

The study then examined the modelled concentrations resulting from a 30% reduction in emissions of SO_2, NO_x, NH_3, VOC and CO. It was concluded that $PM_{2.5}$ concentrations in rural southern UK are likely to be influenced strongly by reductions in SO_2, NO_x and NH_3 emissions in a complex and interlinked manner. The largest reduction in $PM_{2.5}$ was derived from a reduction in NH_3. There were also weak influences from VOC and CO emission changes. In all cases, reductions were less than proportional to the changes in SO_2, NO_x and NH_3 emissions.

8.12.5 Example applications of the EMEP and EMEP4UK regional models

The EMEP regional chemistry-transport model framework is a collection of model pre-processors and post-processors (Figure 70) which work together to produce a detailed representation of the physical and chemical state of the atmosphere over Europe at 50x50 km resolution (Simpson *et al.,* 2003). The EMEP4UK version of the model (Vieno *et al.*, 2009, 2010a) is of 5 x 5 km resolution over the British Isles, nested within the main EMEP Unified Model (Figure 71).

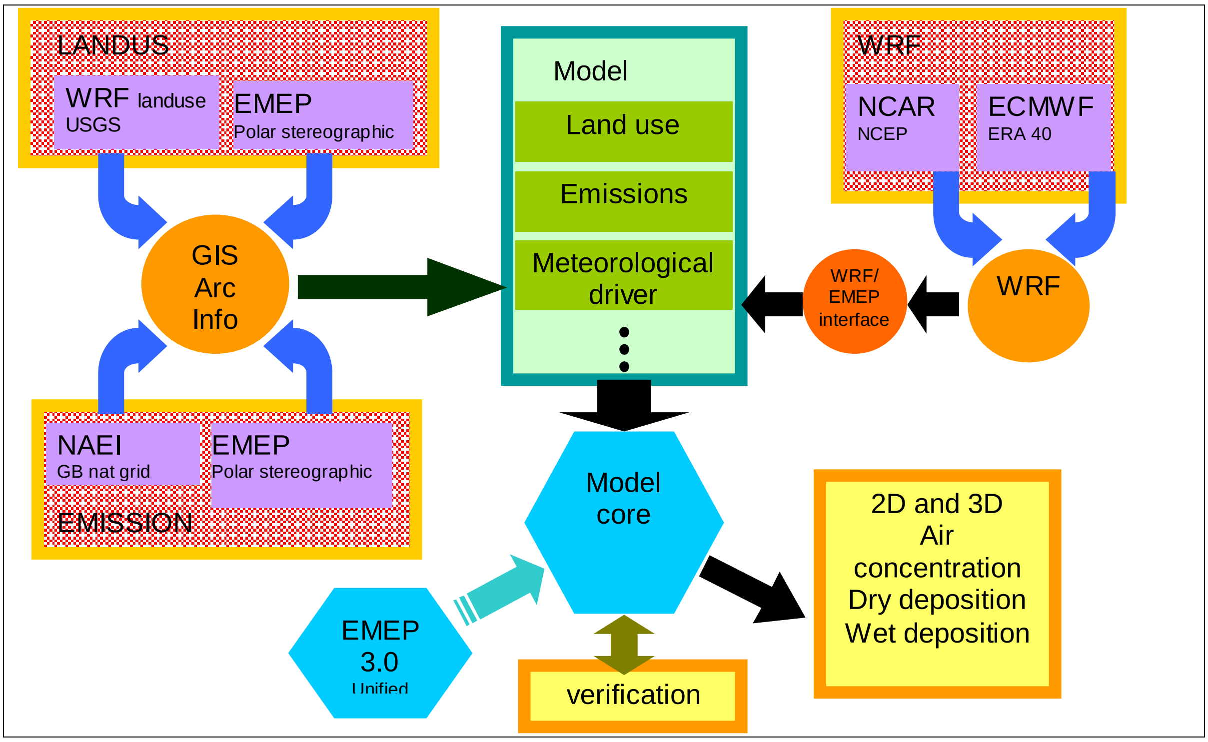

Figure 70 Schematic of the EMEP and EMEP4UK modelling components. Abbreviations as defined in the text. Source: Vieno (2010b).

In the present implementation, the US Weather Research Forecast (WRF) model (http://www.wrf-model.org/) is run specifically to provide the 5 x 5 km meteorological inputs necessary to drive the EMEP4UK model and the initial and boundary conditions (IC & BC) for the whole regional domain. WRF is run in back-cast mode, including data assimilation (Newtonian nudging) of the United States National Center for Environmental Prediction (NCEP) / National Center for Atmospheric Research (NCAR) Global Forecast System (GFS) numerical weather prediction model reanalysis of meteorological observations. The European Centre for Medium-Range Weather Forecasting (EWMWF) re-analysis data can also be used as meteorological input. Both the WRF and EMEP4UK models use 20 vertical layers, with terrain-following coordinates, and resolution increasing towards the surface. The vertical column extends from the surface (centre of the surface layer ~45 m) up to 100 hPa (~16 km).

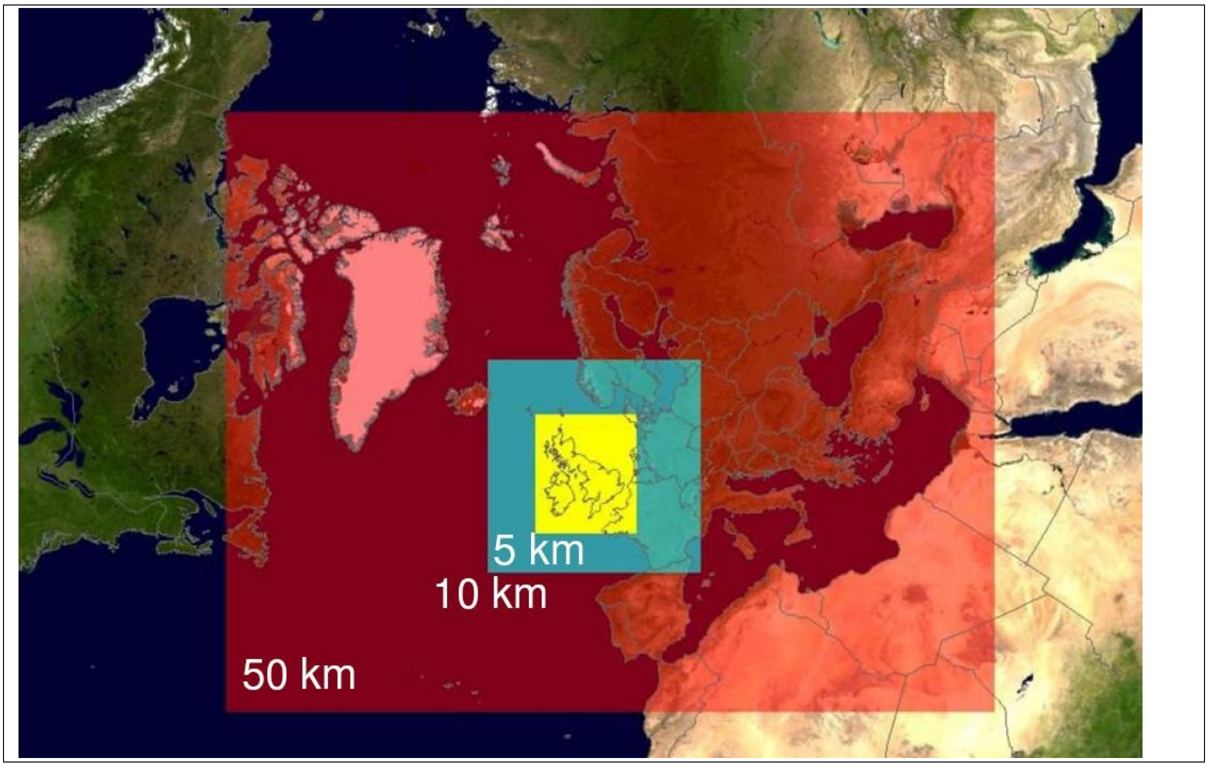

Figure 71 Geographical coverage of the EMEP4UK inner model domain (5 km resolution in yellow) within the main EMEP outer model domain (50 km resolution in red), and of the intermediate (10 km) domain used in the nesting between the two (in blue).

The chemical scheme is identical to the EMEP Unified Model and includes the output variables NH_3, NH_4NO_3, $(NH_4)_2SO_4$, NO, NO_2, NO_3-, HNO_3, peroxyacetyl nitrate (PAN), O_3, SO_2, H_2SO_4, SO_4^{2-}; a full description can be found in Simpson *et al.* (2003). The chemical scheme includes ammonium chemistry, gas and aqueous phase oxidation of SO_2 to sulphate, providing a comprehensive chemistry for both photo-oxidant and acidification studies. Full details of the emissions, chemistry and deposition are given in Vieno *et al.* (2010a).

Examples of the spatial resolution that can be achieved over a wide geographical area with the EMEP4UK model are provided in Figure 72 and Figure 73, which show annual mean nitrate and ammonium concentrations respectively ($PM_{2.5}$ fraction) in 2007 simulated at a 5 x 5 km resolution. These annual averages are derived from hourly simulated values for the whole year. The annual pattern is broadly consistent with that for secondary $PM_{2.5}$ shown in Figure 23.

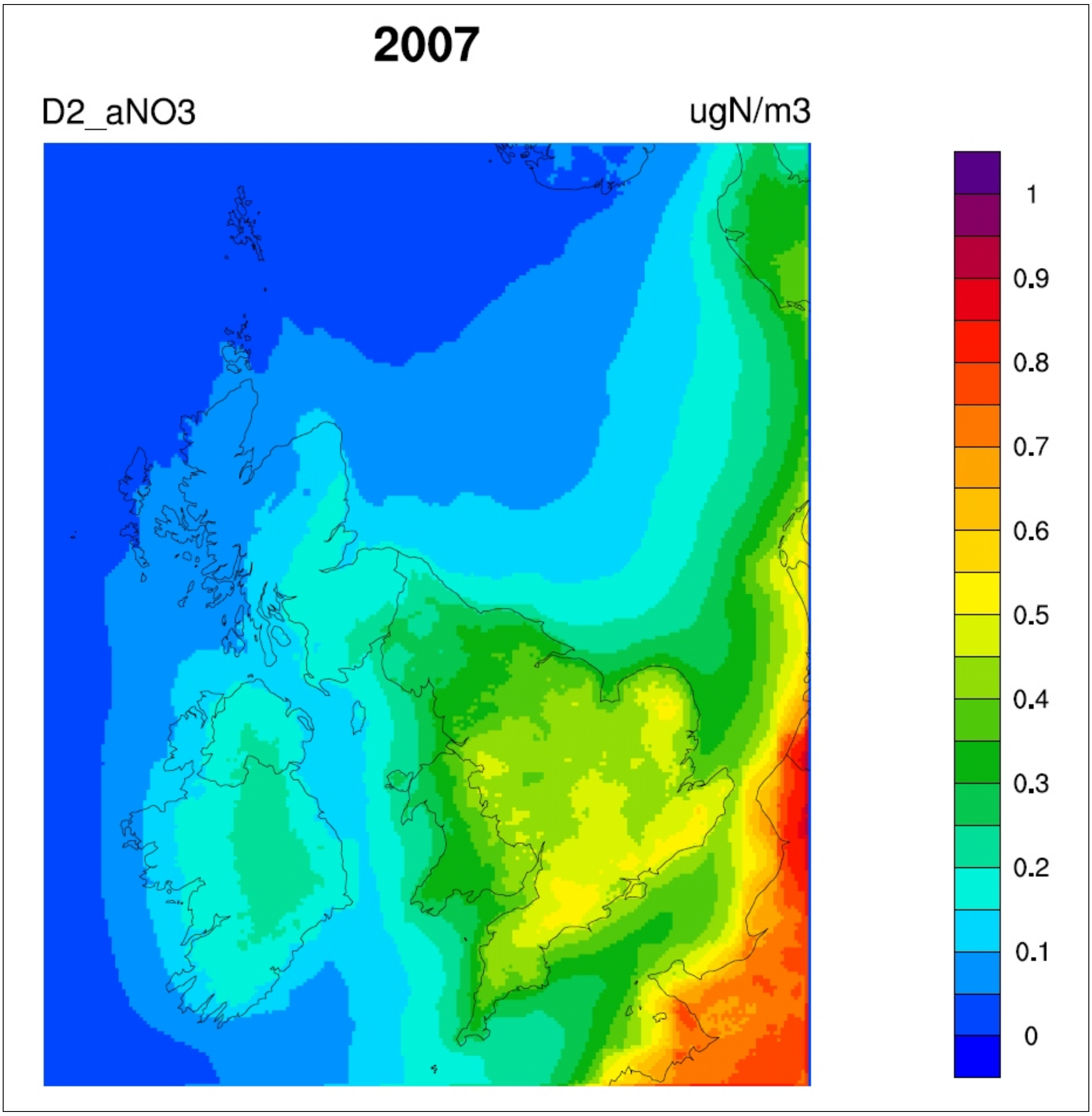

Figure 72 **EMEP4UK simulated surface annual mean nitrate concentrations ($PM_{2.5}$ fraction) in 2007. Units μg/m^3 as N; concentrations as NO_3^- will be 4.43 times higher. Source: Vieno (2010b).**

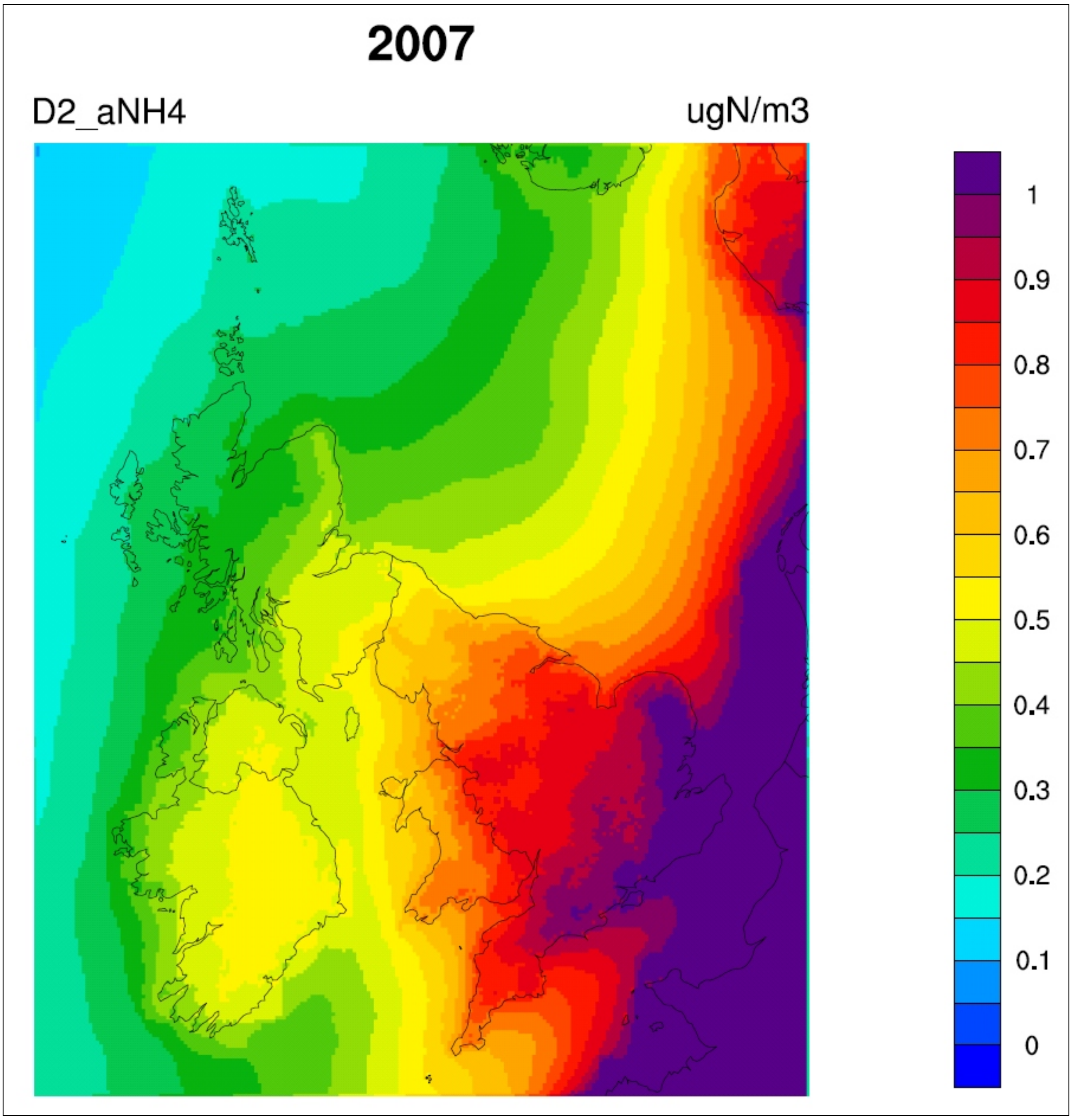

Figure 73 EMEP4UK simulated surface annual mean ammonium concentrations ($PM_{2.5}$ fraction) in 2007. Units mg/m^3 as N; concentrations as NH_4^+ will be 1.29 times higher. Source: Vieno (2010b).

Examples of time series of simulations of the sulphate and ammonium components within fine PM, in comparison with instrumental measurements, are illustrated in Figure 74 and Figure 75, respectively. These illustrate the ability of the model to simulate the temporal patterns of these secondary inorganic PM components at this receptor (and simultaneously for all other 5 × 5 km grid boxes across the UK). A high-resolution process-based model such as this can then be used to investigate the nature of the source-receptor relationships between precursor emissions reductions in the UK and elsewhere, and UK concentrations (and deposition fluxes) of $PM_{2.5}$ and other pollutants.

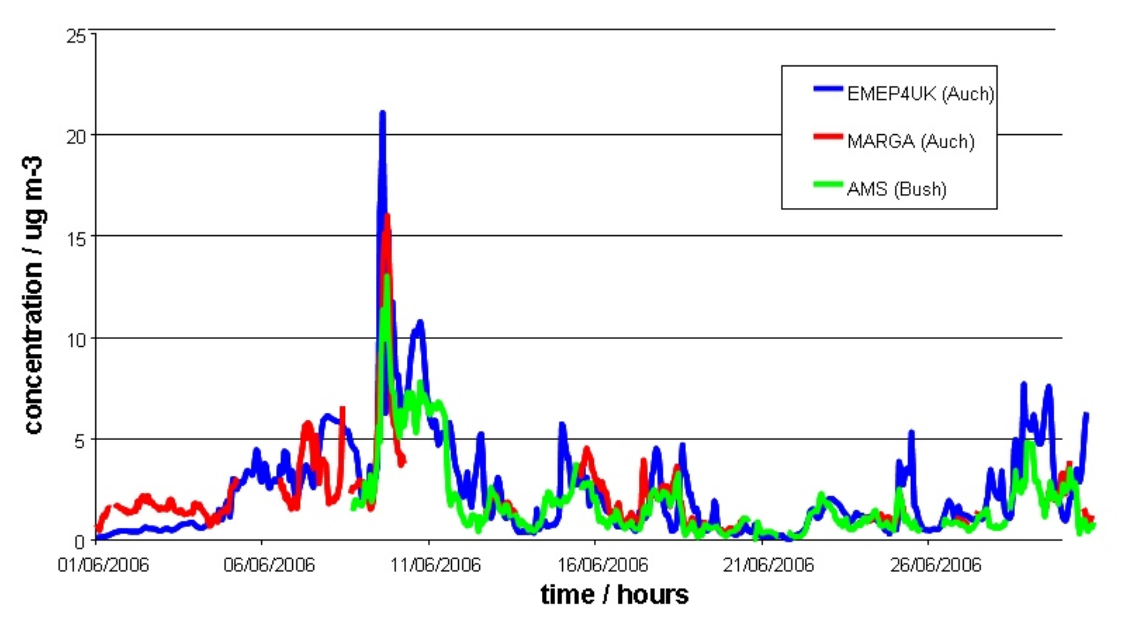

Figure 74 Time series of hourly simulated sulphate aerosol at Auchencorth rural site (blue trace) compared with measurements made at the same site by a Measurement of AeRosols and Gases (MARGA) instrument (red trace) and by an aerosol mass spectrometer (AMS) (green trace). Source: Vieno (2010b).

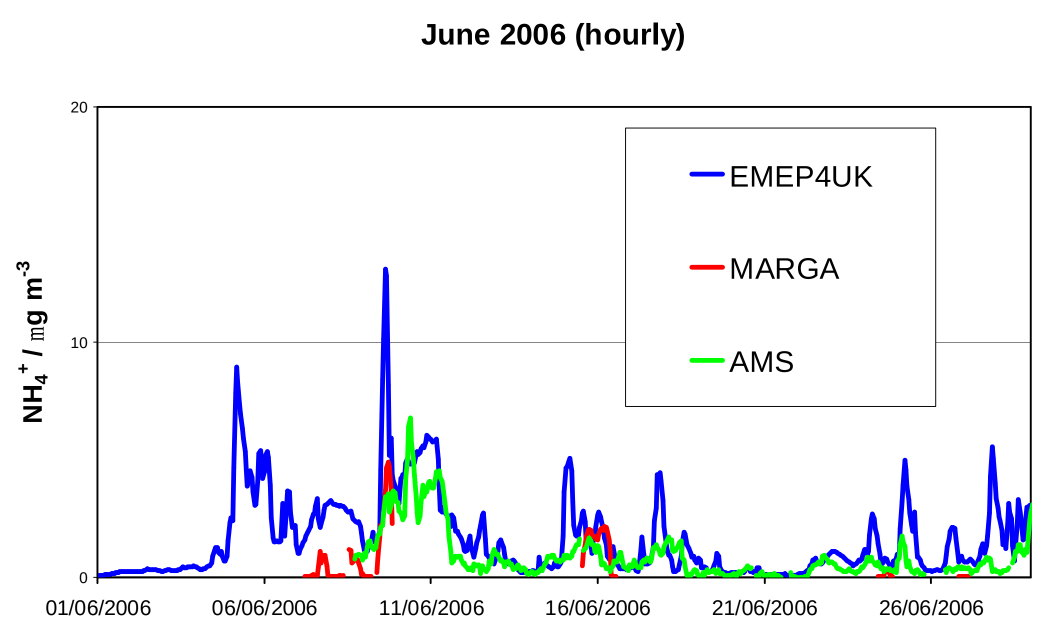

Figure 75 Time series of hourly simulated ammonium aerosol at Auchencorth rural site (blue trace) compared with measurements made at the same site by a Measurement of AeRosols and Gases (MARGA) instrument (red trace) and by an aerosol mass spectrometer (AMS) (green trace). Source: Vieno *et al.* (2009)

.

9. POLICY IMPLICATIONS, RECOMMENDATIONS AND DELIVERY

This Section evaluates the policy implications of the findings set out in this report, recommends additional work that should be carried out and then identifies those responsible for delivering the required reductions in exposure.

9.1 Policy Implications

Analyses of measured concentrations in relation to the EU and UK standards have shown that the key drivers for action to control exposure to $PM_{2.5}$ will be the exposure-reduction targets. The UK target is for a 15% reduction in annual mean $PM_{2.5}$ concentrations at background locations across the major urban areas, while the EU target will be either 10% or 15%. These will represent required reductions of about 1.5 to 2 ug/m^3 between 2010 and 2020. If these reductions are to be achieved from those urban sources giving rise to the urban background enhancement of around 3-6 ug/m^3, then these sources would need to be reduced by some 25-67%. As an alternative, the required reduction could be achieved by reducing the secondary PM contribution that accounts for ~30-50% of urban background $PM_{2.5}$ (around 4-6 ug/m^3). The required reduction would represent a decrease in the secondary $PM_{2.5}$ contribution of 25-50%. If both the urban primary and the regional secondary contributions are tackled equally, the percentage reductions would essentially be halved for each sector, but would still be challenging.

The EU limit values that were established to ensure there are no hot-spots with excessive concentrations are unlikely to be exceeded in the UK. The PM_{10} objectives and limit values have been shown to be more stringent than the $PM_{2.5}$ objectives and limit values, thus the PM_{10} objectives and limit values will drive policies to reduce exposure in hot spots, which will help drive down $PM_{2.5}$ concentrations at these locations.

The response of secondary PM concentrations to changes in precursor gas emissions is less than proportional. This implies that greater reductions in precursor gas emissions are required than might at first sight be the case. The chemistry of secondary inorganic PM formation is such that reductions in ammonia emissions should be more effective at reducing PM concentrations than equivalent reductions in sulphur dioxide and nitrogen oxides. The chemistry of secondary organic PM formation is not currently understood in sufficient detail (including the relationship of SOA formation from different NMVOC precursors in varying pollution climates) to know which source(s) of NMVOC reductions could be most appropriately targeted to reduce PM concentrations.

9.2 Recommendations

Recommendations for further work have been identified as a result of the material set out in this report. They are designed to provide a better understanding of $PM_{2.5}$ sources and concentrations so as to allow appropriate control strategies to be developed. Given the requirements to meet the exposure reduction targets by 2020, these recommendations should be taken to apply to work that should be undertaken over the next few years.

1. Secondary $PM_{2.5}$ has been identified as a major component of $PM_{2.5}$ in the UK. This will require control of emissions of the precursor gases: sulphur dioxide, nitrogen oxides, ammonia and VOCs. These arise variously from industrial, domestic, road traffic and agricultural sources. The extent to which UK and other EU emissions of these precursors contribute to $PM_{2.5}$ in the UK is, however, currently not well understood. This is especially the case for the roles of ammonia and VOCs, as well as the non-proportionalities between changes in emissions and subsequent changes in concentrations. **It is recommended that modelling studies are carried out to establish the contributions of UK and other EU emissions of precursor gases to annual mean $PM_{2.5}$ concentrations, and to determine how these contributions will respond to changes in emissions. This will help guide the development of the most cost-effective control programme.**

2. Secondary $PM_{2.5}$ is estimated to account for some 30-50% of the $PM_{2.5}$ in urban areas, of which secondary organic aerosol is an important component but whose exact composition and contributing sources are still very poorly understood. **It is recommended that a programme of field, chamber and modelling studies is carried out to establish the contributions from anthropogenic and 'natural' sources of organic carbon to $PM_{2.5}$, and how these will respond to changes in emissions.**
3. There is still a poor understanding of emissions of primary $PM_{2.5}$ and their contributions to urban background concentrations. This includes the roles of agricultural dust, resuspended road dust, sea salt, all industrial sources (including biomass installations) and domestic cooking, as well as the effectiveness of different Euro standards for motor vehicle emissions. **It is recommended that further work is carried out on quantifying emissions, in particular from the road transport sector under 'real-world' driving conditions, and from industry. The contributions to urban background concentrations from other sources should be investigated using PM speciation studies. Such studies should then be used to inform the source contributions via modelling.**
4. Air quality legislation will focus attention on predicting annual mean concentrations of $PM_{2.5}$ in urban background locations. This will require modelling of local and regional sources. Currently modelling for policy purposes is confined to the semi-empirical PCM model. It is considered necessary to expand upon this modelling capability. **It is recommended that improved deterministic modelling approaches are developed at the national and urban scale, with robust treatments of atmospheric dynamics, chemistry and aerosol processes, so as to give size and composition-resolved information on airborne concentrations. This will enable reliable projections to be made of future concentrations, to complement the findings of more empirical models.**
5. The national monitoring network has been expanded over the last two years. It is unlikely that many additional sites will be set up by local authorities, given the current economic climate and bearing in mind that there is no requirement for them to assess $PM_{2.5}$ concentrations. There are now 65 sites in the national network, the majority being at urban background locations. There is a significant deficiency of rural monitoring sites, with only 3 stations covering the UK. Rural sites are essential to provide evidence of the urban increment, and to define the extent to which changes in urban concentrations are due to changes in local emissions or to changes in regional background concentrations. A better quantification of rural concentrations is also essential to allow models to be verified, as the regional component should be considered separately from the urban increment. **It is recommended that, as a minimum, 7 additional $PM_{2.5}$ monitors be set up at rural background sites. These could usefully be collocated with existing ozone monitors at: Yarner Wood (SW England), Lough Navar (Northern Ireland), Weybourne (East Anglia), High Muffels (NE England), Ladybower (N England), Narberth (Wales), Aston Hill (Central England).**
6. Considerable resources are devoted to the measurement of PM in the UK. It is important that sufficient resources are also directed to the analysis of the results of the monitoring that is being carried out. This should include the use of the new software tools encompassed in the OpenAir software package, as used for this report. Such analysis will improve understanding of the sources and behaviour of PM, and thereby facilitate the development of the most effective control strategies. **It is recommended that sufficient resources are provided to ensure that the results of the PM monitoring programmes are subject to thorough analysis on an on-going basis.**
7. Current understanding of the quantitative source attribution of airborne PM derives from both source inventory/dispersion model studies and from receptor modelling in which airborne particle concentration/composition data are used to infer source contributions. Given the uncertainties in the emissions of a number of primary $PM_{2.5}$ components and weaknesses in chemistry/transport models for the prediction of secondary $PM_{2.5}$, there needs to be a greater emphasis upon source apportionment

through receptor-modelling studies. Currently, such studies are an active area of research, and approaches based upon chemical mass balance models and multivariate statistical techniques are widely applied but require further enhancement. For example, better information on source chemical profiles in the European context is required for chemical mass balance modelling, and further studies are needed to elucidate which measurements would be most effective in enhancing source discrimination in the multivariate statistical method approaches. Additionally, the most advanced software methods for the latter have yet to be applied to UK data, and currently available multi-component data sets for the United Kingdom are probably inadequate for use of the more advanced procedures. **It is recommended that a programme of work is established to support source apportionment of annual mean $PM_{2.5}$ at urban background locations in different areas of the UK.**

8. The House of Commons Environmental Audit Committee produced a report in March 2010 on air quality (Environmental Audit Committee, 2010). One of its recommendations was: *"The Government must urgently explore how planning guidance can be strengthened and applied to reduce air pollution."* Given that the controls on $PM_{2.5}$ need to be applied across urban areas, there is a clear role for the planning system to play in helping meet the $PM_{2.5}$ exposure-reduction targets. This could be by way of requiring all new developments to consider incorporating measures to reduce $PM_{2.5}$ emissions. **It is therefore recommended that consideration is given to ways of using the planning system to require reductions in $PM_{2.5}$ emissions to be incorporated into the planning of all new developments.**

9.3 Delivery

The policy implications set out above identify a need to deliver reductions in urban background concentrations of $PM_{2.5}$ and recommendations have been made as to further work to help develop the most effective control strategies. The additional work and subsequent control strategies will need to be developed and delivered at several levels. The UK government will play a key role, as will the Scottish Government, the Welsh Assembly Government and the Northern Ireland Executive, while the Mayor for London will have a role to play in London. The Environment Agency, the Scottish Environment Protection Agency and the Environment Agency Northern Ireland will also have an important role to play in relation to industrial, waste and some agricultural sources. At a more local level, local authorities throughout the UK will have a role to play in implementing measures locally.

The **UK Government** and the **devolved administrations** will need to:

- establish the monitoring, modelling and assessment strategy to define sources contributing to urban background $PM_{2.5}$;
- ensure the necessary work is carried out to define as accurately as possible what sources are contributing to the urban background concentrations in each of the agglomerations that form the monitoring network for compliance. This will cover emission inventories for both primary $PM_{2.5}$ and precursor emissions of secondary $PM_{2.5}$ together with source apportionment studies involving both speciated monitoring and modelling;
- ensure that models are applied to the determination of urban background concentrations in each of the agglomerations where compliance monitoring is being carried out, or a representative sample of these across the UK;
- predict the changes in concentrations between 2010 and 2020 arising from measures already in place, i.e. a business as usual scenario; and
- develop a cost-effective package of additional measures to be applied to fill any short-fall in the predicted concentration reduction.

The **Environment Agencies** should be expected to:

- contribute to the best possible emission inventories for:

  - primary $PM_{2.5}$ from processes, including fuel combustion for energy and fugitive emissions, and waste operations regulated by the agencies;
  - secondary $PM_{2.5}$ that arises from emissions of SO_2, NO_x, VOCs and NH_3 from industrial, waste and agricultural operations regulated by the agencies;
- assist with modelling the contributions of primary emissions of $PM_{2.5}$ and emissions that contribute to secondary $PM_{2.5}$ from industrial, agricultural and waste operations to different urban areas; and
- assist with identifying measures that could be introduced to reduce primary emissions of $PM_{2.5}$ and emissions that contribute to secondary $PM_{2.5}$ from industrial and waste operations.

Local Authorities could be expected to:

- help implement national measures to reduce primary $PM_{2.5}$ emissions. This could include requiring measures to control primary $PM_{2.5}$ emissions from new developments.
- help develop controls for $PM_{2.5}$ from new developments; and
- help develop amendments to the Clean Air Act to control emissions from biomass boilers.

10. REFERENCES

Abbott, J., Barker, N., Coleman, P., Howes, P., Stewart, R., Leonard, A. and Collings, A (2007) *Review of the potential impacts on air quality from increased wood fuelled biomass use in London.* Report to London Councils, AEA/ENV/R/2495/Issue 2, December 2007, available at: http://www.londoncouncils.gov.uk/London%20Councils/LondonbiomassreportIssue33010%5B1%5D.pdf

Abbott, J., Stewart, R., Fleming, S., Stevenson, K., Green, J. and Coleman, P. (2008) *Measurement and modelling of fine particulate emissions (PM_{10} & $PM_{2.5}$) from wood-burning biomass boilers*. Report to The Scottish Government, CR/2007/38, September 2008, available at: http://www.scotland.gov.uk/Resource/Doc/243574/0067768.pdf.

Abu-Allaban, M., Gillies, J.A., Gertler, A. W., Clayton, R. and Proffitt, D. (2003) *Tailpipe, resuspended road dust, and brake wear emission factors from on-road vehicles*. Atmospheric Environment, 37(1), 5283-5293.

ACEA, Alliance, EMA and JAMA. (2006) *Worldwide Fuel Charter*. Fourth Edition, September 2006.

Allan, J.D., Williams, P.I., Morgan, W.T., Martin, C.L., Flynn, M.J., Lee, J., Nemitz, E., Phillips, G.J., Gallagher, M.W. and Coe, H. (2010) *Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities.* Atmospheric Chemistry and Physics , 10, 647-668.

Anderson, H.R., Bremner, S.A., Atkinson, R.W., Harrison, R.M., Walters, S. (2001) *Particulate matter and daily mortality and hospital admissions in the West Midlands conurbation of the United Kingdom: Associations with fine and coarse particles, black smoke and sulphate*. Occupational and Environmental Medicine, 58, 504-510.

AQEG (2004) *Nitrogen dioxide in the United Kingdom*. Air Quality Expert Group, Defra, March 2004, London.

AQEG. (2005) *Particulate matter in the United Kingdom. Report of the air quality expert group*. Defra, London, UK.

AQEG. (2007) *Trends in primary nitrogen dioxide in the UK. Report of the air quality expert group*. Defra, London, UK.

Areskoug, H., Johansson, C., Alesand, T., Hedberg, E., Ekengena, T., Vesely, V., Wideqvist, U. and Hansson, H-C. (2004) *Concentrations and sources of PM_{10} and $PM_{2.5}$ in Sweden*. ITM Report 110. Institute of Applied Environmental Research, Stockholm University. ISSN 1103-341X.

Atkinson, R.W., Anderson, H.R., Sunyer, J., Ayres, J., Baccini, M., Vonk, J.M., Boumghar, A., Forastiere, F., Forsberg, B., Touloumi, G., Schwartz, J., and Katsouyanni, K. (2001) *Acute effects of particulate air pollution on respiratory admissions: results from APHEA 2 project. Air pollution and Health: a European Approach.* American Journal of Respiratory and Critical Care Medicine, 164, 1860-1866.

Atkinson, R.W., Fuller, G.W., Anderson, H.R., Harrison, R.M., Armstrong, B. (2010) *Associations between urban ambient particle metrics and health: a time series analysis.* Epidemiology, 2010: in press.

BAA (2008) *Stansted Generation 2 Airport Project: Environmental Statement Volume 4.* Air Quality. BAA

Beelen, R., Hoek, G., van den Brandt, P.A., Goldbohm, R.A., Fischer, P., Schouten, L.J., Jerrett, M., Hughes, E., Armstrong, B., Brunekreef, B. (2008) *Long-term effects of traffic-related air pollution on mortality in a Dutch cohort (NLCS-AIR study).* Environmental Health Perspectives, 116, 196-202.

Bell, M.L., Ebisu, K., Peng, R.D., Samet, J.M., Dominici, F. (2009) *Hospital admissions and chemical composition of fine particle air pollution*. American Journal of Respiratory and Critical Care Medicine, 179, 1115-1120.

Berdowski, J.J.M., Mulder, W., Veldt, C., Visschedijk, A. J. H., and Zandveld, P. Y. J. (1997) *Particulate matter emissions* (*PM_{10} - $PM_{2.5}$ - $PM_{0.1}$*) *in Europe in 1990 and 1993.* TNO Report TNO_MEP - R 96/472. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, Apeldoorn, The Netherlands.

Bolling, A.K., Pagels, J., Yttri, K.E., Barregard, L., Sallsten, G., Schwarze, P.E. and Boman, C. (2009) *Health effects of residential wood smoke particles: the importance of combustion conditions and physicochemical particle properties*. Particle & Fibre Toxicol., 6, 29.

Brown, A.S., Yardley, R.E., Quincey, P.G. and Butterfield, D.M. (2005) *Ambient air particulate matter: quantifying errors in gravimetric measurements.* NPL Report DQL-AS 015.

Brunekreef, B., and Forsberg, B. (2005) *Epidemiological evidence of effects of coarse airborne particles on health*. European Respiratory Journal, 26, 309-318.

Brunekreef, B., Beelen, R., Hoek, G., Schouten, L., Bausch-Goldbohm, S., Fischer, P., Armstrong, B., Hughes, E., Jerrett, M., and van den Brandt, P. (2009) *Effects of long-term exposure to traffic-related air pollution on respiratory and cardiovascular mortality in the Netherlands*. The NLCS-AIR Study, HEI Research Report 139.

Bukowiecki, N., Gehrig, R., Hill, M., Lienemann, P., Zwicky, C. N., Buchmann, B., Weingartner, E., and Baltensperger, U. (2007) *Iron, manganese and copper emitted by cargo and passenger trains in Zurich* (*Switzerland): Size-segregated mass concentrations in ambient air*. Atmospheric Environment, 41, 878–889.

Burnett, R.T., Brook, J., Dann, T., Delocla, C., Philips, O., Cakmak, S., Vincent, R., Goldberg, M.S. and Krewski, D. (2000) *Association between particulate- and gas-phase components or urban air pollution and daily mortality in eight Canadian cities*. Inhalation Toxicology, 12 (Suppl. 4), 15-39.

Cadle, S. H., and Williams, R. L. (1978) *Gas and particle emissions from automobile tires in laboratory and field studies.* Rubber Chemistry and Technology, 52, 146-158.

CAEP (2004) *Report of the committee on aviation environmental protection, sixth meeting Montréal*. **2 – 12 February 2004 (Doc 9836, CAEP/6)**

Calderón-Garciduenas, L., Reed, W., Maronpot, R. R., Henriquez-Roldán, C., Delgado-Chávez, R., Calderón-Garciduenas, A., Dragustinovis, I., Franco-Lira, M., Aragóon-Flores, M., Solt, A.C., Altenburg, M., Torres-Jardón, R. and Swenberg, J.A. (2004) *Brain inflammation and Alzheimer's-like pathology in individuals exposed to severe air pollution*. Toxicologic Pathology. 32**,** 650-658.

Calderón-Garciduenas, L., Solt, A. C., Henriquez-Roldán, C. Torres-Jardón, R., Nuse, B., Herritt, L., Villarreal-Calderón, R., Osnaya, N., Stone, I., García, R., Brooks, D.M., González-Maciel, A., Reynoso-Robles, R., Delgado-Chávez, R. and Reed, W. (2009)

Long-term air pollution exposure is associated with neuroinflammation, an altered innate immune response, disruption of the blood-brain barrier, ultrafine particulate deposition, and accumulation of amyloid beta-42 and alpha-synuclein in children and young adults. Toxicologic Pathology, 36**,** 289-310.

Cardina, J. A. (1974) *Particle size determination of tire tread rubber in atmospheric dusts*. Rubber Chemistry and Technology, 47, 1005-1010.

Caseiro, A., Bauer, H., Schmidl, C., Pio, C.A. and Puxbaum H. (2009) *Wood burning impact on* PM_{10} *in three Austrian regions.* Atmospheric Environment, 43, 2186-2195.

CEN (1999) *Air quality – determination of the* PM_{10} *fraction of suspended particulate matter. Reference method and field test procedure to demonstrate equivalence of measurement methods.* (Available through the British Standards Institute (BSI) as BS EN12341).

CEN (2005) *Air Quality – Determination of the* $PM_{2.5}$ *fraction of suspended particulate matter. Reference method and field test procedure to demonstrate equivalence of measurement methods.* (Available through the British Standards Institute (BSI) as BS EN14907).

Cha, S., Carter, P. and Bradow, R. L. (1983) *Simulation of automobile brake wear dynamics and estimation of emissions*. SAE Transactions Paper 831036, Society of Automotive Engineers, Warrendale, Pennsylvania.

Chemel, C., Sokhi, R.S., Dore, A.J., Sutton, P., Vincent. K.J., Griffiths, S.J., Hayman, G.D., Wright, R., Baggaley, M., Hallsworth, S., Prain, D. and Fisher, B.E.A. (2010) *Predictions of UK Regulated Power Station Contributions to Regional Air Pollution and Deposition: A Model Comparison Exercise*. In preparation.

Chemel, C. (2010) *Personal communication.* National Centre for Atmospheric Science, Weather Directorate.

Charron, A. and Harrison, R.M. (2005) *Fine (*$PM_{2.5}$*) and coarse (*$PM_{2.5\text{-}10}$*) particulate matter on a heavily trafficked London highway: sources and processes.* Environmental Science and Technology, 39, 7768-7776.

Chen, L.C. and Lippmann, M. (2009) *Effects of metals within ambient air particulate matter* (*PM) on human health*. Inhalation Toxicology, 21, 1-31.

Cho, S.H., Tong, H.Y., McGee, J.K., Baldauf, R.W., Krantz, Q.T., and Gilmour, M.I. (2009) *Comparative toxicity of size-fractionated airborne particulate matter collected at different distances from an urban highway*. Environmental Health Perspectives, 117, 1682-1689.

Chow, J.C., Fairley, D., Watson, J.G., DeMandel, R., Fujita, E.M., Lowenthal, D.H., Lu, Z., Frazier, C.A., Long, G., Cordova, J. (1995) *Source spportionment of wintertime* PM_{10} *at San Jose, California.* Journal of Environmental Engineering, 121, 378-387.

Clancy, L., Goodman, P., Sinclair, H. and Dockery, D.W. (2002) *Effects of air-pollution control on death rates in Dublin, Ireland: an intervention study.* Lancet, 360, 1210.

COMEAP. (2009) *Long-term exposure to air pollution: effect on mortality.* A report by the Committee on the Medical Effects of Air Pollutants.

Dahl, A., Gharibi, A., Swietlicki, E., Gudmundsson, A., Bohgard, M., Ljungman, A., Blomqvist, G. and Gustafsson, M. (2006) *Traffic-generated emissions of ultrafine particles from pavement-tire interface*. Atmospheric Environment, 40, 1314–23.

Dannis, M. L. (1974) *Rubber dust from the normal wear of tyres*. Rubber Chemistry and Technology, 47, 1011-1037.

Defra (2007) *The air quality strategy for England, Scotland, Wales and Northern Ireland,* July 2007, Defra, London, available at: http://www.defra.gov.uk/environment/quality/air/airquality/strategy/index.htm.

Defra, (2009) *Local air quality management – technical guidance LAQM.TG(09).* February 2009, Defra, available at: http://www.defra.gov.uk/environment/quality/air/airquality/local/guidance/policy.htm

Defra (2010a) *Background maps*, available at: http://laqm1.defra.gov.uk/review/tools/background.php

Defra (2010b) *Valuing the overall impacts of air pollution*. March 2010, Defra, London, available at: http://www.defra.gov.uk/environment/quality/air/airquality/panels/igcb/documents/100303-aq-valuing-impacts.pdf

Defra (2010c) *NO_x to NO_2 calculator*, available at: http://laqm1.defra.gov.uk/review/tools/monitoring/calculator.php

De Leeuw, F. and Horálek, J. (2009) *Assessment of the health impacts of exposure to $PM_{2.5}$ at a European level*. ETC/ACC Technical Paper 2009/1. The European Topic Centre on Air and Climate Change (ETC/ACC), Bilthoven, The Netherlands.

Denier van der Gon, H. and Hulskotte, J. (2009) *Methodologies for estimating shipping emissions in the Netherlands - a documentation of currently used emission factors and related activity data.* Report 500099012, Netherlands Environmental Assessment Agency, Bilthoven. ISSN: 1875-2322.

Derwent, R., Witham, C., Redington, A., Jenkins, M., Stedman, J., Yardley, R. and Hayman, G. (2009) *Particulate matter at a rural location in southern England during 2006: Model sensitivities to precursor emissions.* Atmospheric Environment, 43, 689-696.

Derwent, D., Fraser, A., Abbott, J., Jenkin, M., Willis, P. and Murrells, T. (2010) *Evaluating the performance of air quality models.* Issue 3, June 2010, Defra, available at: http://www.airquality.co.uk/reports/list.php

DfT (2006) *Project for the sustainable development of Heathrow - Report of the Air Quality Technical Panels*, Department for Transport, London.

Diaz-Sanchez, D., Garcia, M.P., Wang, M., Jyrala, M., and Saxon, A. (1999) *Nasal challenge with diesel exhaust particles can induce sensitization to a neoallergen in the human mucosa.* Journal of Allergy and Clinical Immunology 104, 1183-1188.

Diociaiuti, M., Balduzzi, M., De Berardis, B., Cattani, G., Stacchini, G., Ziemacki, G., Marconi, A., and Paoletti, L. (2001) *The two $PM_{2.5}$ (fine) and $PM_{2.5-10}$ (coarse) fractions: Evidence of different biological activity*. Environmental Research Section A, 86, 254-262.

Dockery, D.W., Pope, C.A., Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G., and Speizer, F.E. (1993) *An association between air pollution and mortality in six U.S. cities*. New England Journal of Medicine, 329, 1753-1759.

Dominicci, F., Peng, R.D., Bell, M.L., Pham, L., McDermott, A., Zeger, S.L., and Samet, J.M. (2006) *Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases.* Journal of the American Medical Association, 295, 1127-1134.

Donaldson, K., *Mills, N., MacNee, W., Robinson, S., Newby, D.* (2005) *Role of inflammation in cardiopulmonary health effects of PM*. Toxicology and Applied Pharmacology, 207, 483-488.

EC (2004) *Second position paper on particulate matter.* CAFÉ Working Group on Particulate Matter, available at: http://ec.europa.eu/environment/archives/cafe/pdf/working_groups/2nd_position_paper_pm.pdf

EC Working Group (2009) *Guide to the demonstration of equivalence of ambient air monitoring methods*. January 2010, available at: http://ec.europa.eu/environment/air/quality/legislation/pdf/equivalence.pdf

EEA (2009) *EMEP/EEA air pollutant emission inventory guidebook.* Technical report 9/2009, June 2009. Available at: http://www.eea.europa.eu/publications/emep-eea-emission-inventory-guidebook-2009

Elder, A.C.P., Gelein, R., Finkelstein, J.N., Cox, C., and Oberdorster, G. (2000) *Pulmonary inflammatory response to inhaled ultrafine particles is modified by age, ozone exposure, and bacterial toxin.* Inhalation Toxicology 12, 227-246.

Environment Agency (2009a) *Study of ambient air quality at Scunthorpe, 6 August 2008 to 15 April 2009*. Technical Report NMA/TR/2009/04, July 2009, Environment Agency.

Environment Agency (2009b) *Port Talbot steelworks* PM_{10} *permit review.* January 2009, Environment Agency.

Environmental Audit Commission (2010) *Air quality, Fifth Report of Session 2009-10*. Vol. 1, March 2010, The Stationery Office, London

Fauser, P. (1999) *Particulate air pollution with emphasis on traffic generated aerosols*. Riso-R-1053(EN), Riso National Laboratory, Roskilde, Denmark.

Favez, O., Cachier, H., Sciare, J., Sarda-Esteve, R. and Martinon, L. (2009) *Evidence for a significant contribution of wood burning aerosols to* $PM_{2.5}$ *during the winter season in Paris, France.* Atmospheric Environment., 43, 3640-3644.

Finlayson-Pitts, B.J. and Pitts, J.N. (2000) *Chemistry of the upper and lower atmosphere,* Academic Press.

Fridell, E., Steen, E., and Peterson, K. (2008) *Primary particles in ship emissions*. Atmospheric Environment, 42, 1160–1168.

Gaffney, P., Bode, R. and Murchison, L. (1995) PM_{10} *emission inventory improvement program for California.* Air Resources Borar, 2020L Street, Sacramento, CA.95814.

Garg, B. D., Cadle, S. H., Mulawa, P. A., Groblicki, P. J., Laroo, C., and Parr, G. A. (2000) *Brake wear particulate matter emissions.* Environmental Science and Technology, 34 (21), 4463-4469.

Gavett, S.H., Madison, S.L., Dreher, K.L., Winsett, D.W., McGee, J.L., Costa, D.L. (1997) *Metal and sulfate composition of residual oil fly ash determines airway hyperreactivity and lung injury in rats*. Environmental Research, 72, 162-172

Gelencser, A., May, B., Simpson, D., Sanchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Caseiro, A., Pio, C. and Legrand, M. (2007*) Source apportionment of* $PM_{2.5}$ *organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin*. Journal of Geophysical Research 112, D23S04, doi:10.1029/2006JD008094.

Gerlofs-Nijland, M.E., Dormans, J.A.M.A., Bloemen, H.J.T., Leseman, D.L.A.C., Boere, A.J.F., Kelly, F.J., Mudway, I.S., Jimenez, A.A., Donaldson, K., Guastadisegni, C., Janssen, N.A.H., Brunekreef, B., Sandstrom, T., and Cassee, F.R. (2007) *Toxicity of coarse and fine particulate matter from sites with contrasting traffic profiles.* Inhalation Toxicology, 19, 1055-1069.

Gerlofs-Nijland, M.E., Rummelhard, M., Boere, A. J. F., Leseman. D.L.A.C., Duffin, R., Schins, R.P.F., Borm, P.J.A., Sillanää, M, Salonen, R.O. and Cassee, F.R. (2009) *Particle induced toxicity in relation to transition metal and polycyclic aromatic hydrocarbon contents*. Environmental Science & Technology , 43**,** 4729-4736.

Gietl, J.K., Lawrence, R., Thorpe, A.J. and Harrison, R.M. (2010) *Identification of brake wear particles and derivation of a quantitative tracer for brake dust at a major road*. Atmospheric Environment, 44, 141-146.

Goldberg, M.S., Burnett, R.T., and Bailar, J.C., Brook, J., Bonvalot, Y., Tamblyn, R., Singh, R., Valois, M-F. and Vincent, R. (2001) *The association between daily mortality and ambient air particle pollution in Montreal, Quebec 2.* Cause-specific mortality. Environmental Research, 86, 26-3.

Grahame, T.M., and Schlesinger, R. (2005) *Evaluating the health risk from secondary sulfates in eastern North American regional ambient air particulate matter.* Inhalation Toxicology, 17, 15-27.

Grahame, T.J., and Schlesinger, R. B. (2007) *Health effects of airborne particulate matter: Do we know enough to consider regulating specific particle types or sources?* Inhalation Toxicology, 19, 457-48.

Grahame, T.J., and Schlesinger, R. B. (2010) *Cardiovascular health and particulate vehicular emissions: a critical evaluation of the evidence*. Air Quality Atmospheric Health, 3, 3-27.

Granum, B., Gaarder, P.I., and Lovik, M. (2001) *IgE adjuvant effect caused by particles - immediate and delayed effects.* Toxicology 156, 149-159.

Green, D. (2007) *The King's College London volatile correction model for* PM_{10} *- development, testing and application.* Report KCLERG\MT\FDMS\EQ prepared for Defra, May 2007.

Grice, S.E., Lingard, J.J.N., Stedman, J. R., Cooke, S.L., Yap, F.W., Kent, A.J., Bush, T.J., Vincent, K.J. and Abbott, J. (2010) *UK air quality modelling for annual reporting 2008 on ambient air quality assessment under Council Directives 96/62/EC, 1999/30/EC and 2000/69/EC.* AEA/ENV.R.2859 Issue 1, July 2010, AEA, Harwell.

Happo, M.S., Hirvonen, M.R., Hälinen, A.I., Jalava, P.I., Pennanen, A.S., Sillanpää, M., Hillamo, R. and Salonen, R.O. (2008) *Chemical compositions responsible for inflammation and tissue damage in the mouse lung by coarse and fine particulate samples from contrasting air pollution in Europe*. Inhalation Toxicology, 20**,** 1215-1231.

Harrison D (2006) *UK equivalence programme for monitoring of particulate matter*. Report BV/AQ/AD202209/DH/2396, June 2006, Available at: www.airquality.co.uk/archive/reports/list.php

Harrison, D. (2010) Personal communication, Bureau Veritas.

Harrison, R.M., Yin, J., Mark, D., Stedman, J., Appleby, R.S., Booker, J. and Moorcroft, S. (2001) *Studies of the coarse particle (2.5-10 μm) component in UK urban atmospheres.* Atmospheric Environment, 35, 3667-3679.

Harrison, R.M., Jones, A.M. and Lawrence, R.G. (2003) *A pragmatic mass closure model for airborne particulate matter at urban background and roadside sites*. Atmospheric Environment, 37, 4927-4933.

Harrison, R.M., Smith, D.J.T., and Kibble, A.J. (2004) *What is responsible for the carcinogenicity of* $PM_{2.5}$*?* Occupational and Environmental Medicine, 61, 799-805.

Hayes, E., and Chatterton, T. (2009) *An independent review of monitoring measures undertaken in Neath Port Talbot in respect of particulate matter (*PM_{10}*)*. Report prepared for Welsh Assembly Government. Available at: http://wales.gov.uk/topics/environmentcountryside/epq/airqualitypollution/airquality/southwaleszone/porttalbotpm10/;jsessionid=PMbsLKJTk4p1GBLXK1WhTDS64GWXcc6nQgycsBHB66zhxzqw1H2r!200562741?lang=en

Heal, M. R., Hibbs, L. R., Agius, R. M. and Beverland, I. J. (2005) *Interpretation of variations in fine, coarse and black smoke particulate matter concentrations in a Northern European city*. Atmospheric Environment, 39, 3711-3718.

Heal, M. R., Naysmith, P., Cook, G. T., Xu, S., Raventos Duran, T. and Harrison, R. M. (2010) *Application of 14C analyses to source apportionment of carbonaceous* $PM_{2.5}$ *in the UK*, Atmospheric Environment, submitted.

Health Effects Institute (2003) *Revised analyses of time-series studies of air pollution and health.* Special Report, Health Effects Institute, Boston, MA.

Hedley, A.J., Wong, C.M., Thach, T.Q., Ma, S., Lam, T.H., and Anderson, H.R. (2002) *Cardiorespiratory and all-cause mortality after restrictions on sulphur content of fuel in Hong Kong: an intervention study*. Lancet, 360, 1646-1652.

Heywood, J. B. (1988) *Internal combustion engine fundamentals.* McGraw-Hill , New York

Hickman, A.J., McCrae, I.S., Cloke, J. and Davies, G.J. (2002) *Measurement of roadside air pollution dispersion*. Project report PR SE/445/02 TRL Ltd, Crowthorne.

Hildemann, L.M., Markowski, G.R. and Cass, G.R. (1991) *Chemical composition of emissions from urban sources of fine organic aerosol,* Environmental Science and Technology, 25, 744-759.

Holland, M., Watkiss, P., Pye, S., de Oliveria, A. and Van Regemorter, D (2005) *Cost-benefit analysis of the thematic strategy on air pollution*. Report AEAT/ED48763001/Thematic Strategy, Issue 1. Available at: http://ec.europa.eu/environment/archives/cafe/general/keydocs.htm#integrated.

Horálek, J., de Smet, P., de Leeuw, F., Denby, B., Kurfurst, P. and Swart, R. (2008) *European air quality maps for 2005 including uncertainty analysis*. ETC/ACC Technical

Paper 2007/7. Available online: http://air-climate.eionet.europa.eu/reports/ETCACC_TP_2007_7_spatAQmaps_ann_interpol.

IGCB (2006) *An economic analysis to inform the air quality strategy review consultation*. Third report of the Interdepartmental Group on Costs and Benefits, PB 11252b, Department for Environment, Food and Rural Affairs, London.

Iijima, A., Sato, K., Yano, K., Tago, H., Kato, M., Kimura, H. and Furuta, N. (2007) *Particle size and composition distribution analysis of automotive brake abrasion dusts for the evaluation of antimony sources of airborne particulate matter*. Atmospheric Environment, 41, 4908–19.

IMO (2008): MEPC.176(58) *Amendments to the annex of the protocol of 1997 to amend the international convention for the prevention of pollution from ships, 1973, as modified by the protocol of 1978 relating thereto (Revised MARPOL Annex VI).* International Maritime Organisation, London.

ISO (2009) *Stationary source emissions - determination of PM_{10}/$PM_{2.5}$ mass concentration in flue gas - measurement at low concentrations by use of impactors*. ISO 23210:2009, www.iso.org/iso/iso_catalogue/catalogue_ics/catalogue_detail_ics.htm?csnumber=53379

Jaecker-Voirol, A. and Pelt, P. (2000) *PM_{10} emission inventory in Ile de France for transport and industrial sources: PM_{10} re-suspension, a key factor for air quality.* Environmental Modelling & Software 15, 575-580.

Jalava, P.I., Salonen, R.O., Pennanen, A.S., Sillanpää, M., Hälinen, A.I., Happo, M.S., Hillamo, R., Brunekreef, B., Katsouyanni, K., Sunyer, J. and Hirvonen, M.R. (2007) *Heterogeneities in inflammatory and cytotoxic responses of RAW 264.7 macrophage cell line to urban air coarse, fine, and ultrafine particles from six European sampling campaigns*. Inhalation Toxicology, 19, 213-225.

Janssen, N.A., Brunekreef, B., van Vliet, P., Aarts, F., Meliefste, K., Harssema, H. and Fischer, P. (2003) *The relationship between air pollution and heavy traffic and allergic sensitization, bronchial hyperresponsiveness, and respiratory symptoms in Dutch schoolchildren.* Environmental Health Perspectives, 111, 1512-1518.

Jenkin, M.E., Murrells, T.P. and Passant, N.R. (2000) The temporal dependence of ozone precursor emission: estimation and application. Report to DETR, AEA//R/ENV/0355/Issue 1, November 2000, available at: http://www.airquality.co.uk/reports/empire/AEAT_ENV_0355_v2.pdf

Johansson, C. (2002) *Source contributions of PM in Sweden – implications for abatement strategies.* Institute of Applied Environmental Research (ITM), Stockholm. Presentation at Presentation at 'Particles in the size of 2.5 to 10 microns in urban areas' workshop, in support of the Clean Air for Europe (CAFE) programme of DG ENV in Berlin, Germany, November 4-6, 2002

Katsouyanni, K., Touloumi, G., Samoli, E., Gryparis, A., Le Tertre, A., Monopolis, Y., Rossi, G., Zmirou, D., Ballester, F., Boumghar, A., Anderson, H.R., Wojtyniak, B., Paldy, A., Braunstein, R., Pekkanen, J., Schindler, C. and Schwartz, J. (2001) *Confounding and effect modification in the short-term effects of ambient particles on total mortality: Results from 29 European cities within the APHEA2 project.* Epidemiology 12, 521-531.

Kleeman, M.J., and Cass, G.R. (1999) *Effect of emissions control strategies on the size and composition distribution of urban particulate air pollution.* Environmental Science and Technology, 33, 177-189.

Ko, F.W.S., Tam, W., and Wong, T.W., Lai, C.K.W., Wong, G.W.K., Leung, T.F., Ng, S. and Hui, D.S.C. (2007) *Effects of air pollution on asthma hospitalization rates in different age groups in Hong Kong*, Clinical and Experimental Allergy, 37**,** 1312-1319.

Koenig, J.Q. Larson, T.V., Hanley, Q.S., Rebolledo, V., Dumler, K., Checkoway, H., Wang, S.Z., Lin, D. and Pierson, W.E. (1993) *Pulmonary function changes in children associated with fine particulate matter*. Environmental Research, 63, 26-38.

Krämer, U., Herder, C., Sugiri, D., Strassburger, K., Schikowski, T., Ranft, U., Rathmann, W. (2010). *Traffic-related air pollution and incident type 2 diabetes: results from the SALIA cohort study*. Environmental Health Perspectives, 118, 1273-1279.

Kunzli, N., Mudway, I.S., Gotschi, T., Shi, T.M., Kelly, F.J., Cook, S., Burney, P., Forsberg, B., Gauderman, J.W., Hazenkamp, M.E., Heinrich, J., Jarvis, D., Norback, D., Payo-Losa, F., Poli, A., Sunyer, J., and Borm, P.J.A. (2006) *Comparison of oxidative properties, light absorbance, and total and elemental mass concentration of ambient $PM_{2.5}$ collected at 20 European sites*. Environmental Health Perspectives, 114, 684-690.

Laden, F., Neas, L.M., Dockery, D.W., and Schwartz, J. (2000) *Association of fine particulate matter from different sources with daily mortality in six US cities*. Environmental Health Perspectives, 108, 941-947.

Laden, F., Schwartz, J., Spelzer, F.E. and Dockery, D.W. (2006) *Reduction in fine particulate air pollution and mortality – extended follow-up of the Harvard six cities study*. American Journal of Respiratory and Critical Care Medicine, 173, 667-672.

Laxen, D. and Moorcroft, S. (2005) *Options for an exposure-reduction approach to air quality management in the UK and EU for non-threshold pollutants.* Air Quality Consultants Ltd, January 2005. Available from www.aqconsultants.co.uk

Lipfert, F.W., Baty, J.D., Miller, J.P., and Wyzga, R.E. (2006) *$PM_{2.5}$ constituents and related air quality variables as predictors of survival in a cohort of U. S. military veterans*. Inhalation Toxicology, 18, 645-657.

Lohrer, W. and Mierheim, L. W. (1983). *Staub-Reinhalt.* Luft, 43, 78-83.

Madden, M.C., Richards, J.H., Dailey, L.A., Hatch, G.E., and Ghio, A.J. (2000) *Effect of ozone on diesel exhaust particle toxicity in rat lung.* Toxicology and Applied Pharmacology 168, 140-148.

Matthijsen and ten Brink, H. M. (2007) *$PM_{2.5}$ in the Netherlands - consequences of the new European air quality standards*. MNP Report 500099001/2007. The Netherlands Environmental Assessment Agency (MNP), Bilthoven, The Netherlands.

McDonnell, W.F., Nishino-Ishikawa, N., Petersen, F.F., Chen, L.H., and Abbey, D.E. (2000) *Relationships of mortality with the fine and coarse fractions of long-term ambient PM_{10} concentrations in nonsmokers*. Journal of Exposure Analysis and Environmental Epidemiology, 10, 427-436.

Mills, N.L., Tornqvist, H., Robinson, S.D., Gonzalez, M., Darnley, K., MacNee, W., Boon, N.A., Donaldson, K., Blomberg, A., Sandstrom, T., and Newby, D. E. (2005*) Diesel exhaust inhalation causes vascular dysfunction and impaired endogenous fibrinolysis*. Circulation, 112, 3930-3936.

Mills, N.L., Donaldson, K., Hadoke, P.W., Boon, N.A., MacNee, W., Cassee, F.R., Sandstrom, T., Blomberg, A., and Newby, D.E. (2008) *Adverse cardiovascular effects of air pollution*. Nature Clinical Practice Cardiovascular Medicine, 6, 36.

Moosmüller, H., Gillies, J., Rogers, F., DuBois, D., Chow, J., Watson, J. and Langston, R. (1998) *Particulate emission rates for unpaved shoulders along a paved road*. Journal of the Air and Waste Management Association. 48, 398-407.

Mosleh, M., Blau, P.J. and Dumitrescu, D. (2004) *Characteristics and morphology of wear particles from laboratory testing of disk brake materials.* Wear 256:1128–34.

Murrells, T.P. (2010) *Personal communication*. AEA, Harwell.

NAEI (2010) *National Atmospheric Emissions Inventory*. At: http://www.naei.org.uk/

Oberdorster, G., Gelein, R.M., Ferin, J., and Weiss, B. (1995) *Association of particulate air pollution and acute mortality - Involvement of ultrafine particles.* Inhalation Toxicology 7, 111-124.

Official Journal (1999). *Directive 1999/13/EC of the European Parliament and of the Council on limitation of emissions of VOCs due to the use of organic solvents in certain activities & installations.* OJEC, OJ L85, 1999.

Official Journal (2000). *Directive 2000/76/EC of the European Parliament and of the Council on waste incinerators.* OJEC, OJ L332, 2000.

Official Journal (2001). *Directive 2001/80/EC of the European Parliament and of the Council on limitation of emissions of certain pollutants into the air from large combustion plant.* OJEC, OJ L309, 2001.

Official Journal (2001). *Directive 2001/81/EC of the European Parliament and of the Council on National Emissions Ceilings for certain atmospheric pollutants.* OJEC, OJ L309, 2001.

Official Journal (2004). *Directive 2004/42/EC of the European Parliament and of the Council on limitation of emissions of VOCs due to use of organic solvents in certain paints & varnishes & vehicle refinishing products.* OJEC, OJ L143, 2004.

Official Journal (2009). *Directive 2001/81/EC of the European Parliament and of the Council concerning Integrated Pollution Prevention and Control.* OJEC, OJ L24, 2008.

Ostro, B., Broadwin, R., Green, S., Feng, W.Y., and Lipsett, M. (2006) *Fine particulate air pollution and mortality in nine California counties: Results from CALFINE*, Environmental Health Perspectives, 114**,** 29-33.

Peng, R.D., Bell, M.L., Geyh, A.S., McDermott, A., Zeger, S.L., Samet, J.M. and Dominici, F. (2009) *Emergency admissions for cardiovascular and respiratory diseases and the chemical composition of fine particle air pollution*. Environmental Health Perspectives, 117, 957-963.

Peters, A., *Dockery, D.W., Muller, J.E. and Mittleman, M.A*. (2001) *Increased particulate air pollution and the triggering of myocardial infarction.* Circulation, 103, 2810-2815.

Pierson, W. R. and Brachaczek, W. W. (1974) Airborne *particulate debris from rubber tyres*. Rubber Chemistry and Technology, 47, 1275-1299.

Pope, C.A., Thun, M.J., Namboodiri, M.M., Dockery, D.W., Evans, J.S., Speizer, F.E. and Heath, C.W.(1995) *Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults*. American Journal of Respiratory and Critical Care Medicine, 151, 669-674.

Pope, C.A. Burnett, R.T., Thun, M.J., Calle, E.E. Krewski, D., Ito, K. and Thurston, G.D. (2002) *Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution*. Journal of the American Medical Association, 287, 1132-1141.

Pope, C.A. (2007) *Mortality effects of longer term exposures to fine particulate air pollution: Review of recent epidemiological evidence*. Inhalation Toxicology, 19 (Suppl. 1), 33-38.

Pope, C.A., Ezzati, M. and Dockery, D.W. (2009) *Fine-particulate air pollution and life expectancy in the United States*. New England Journal of Medicine, 360, 376-386.

Pregger, T., Friedrich, R. (2002) *Emission sources of PM_{10} (coarse and fine fraction) in Germany.* Universität Stuttgart, Stuttgart. Presentation at 'Particles in the size of 2.5 to 10 microns in urban areas' workshop, in support of the Clean Air for Europe (CAFE) programme of DG ENV in Berlin, Germany, November 4-6, 2002.

Putaud, J.P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Flentje, H., Fuzzi, S., Gehrig, R., Hansson, H. C., Harrison, R. M., Herrmann, H., Hitzenberger, R., Hüglin, C., Jones, A. M., Kasper-Giebl, A., Kiss, G., Kousa, A., Kuhlbusch, T. A. J., Löschau, G., Maenhaut, W., Molnar, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz, M., Puxbaum, H., Querol, X., Rodriguez, S., Salma, I., Schwarz, J., Smolik, J., Schneider, J., Spindler, G., ten Brink, H., Tursic, J., Viana, M., Wiedensohler, A. and Raes. F. (2010).*A European aerosol phenomenology - 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe*. Atmospheric Environment, 44, 1308-1320.

Puxbaum, H., Caseiro, A., Sanchez-Ochoa, A., Kasper-Giebl, A., Claeys, M., Gelencser, A., Legrand, M., Preunkert, S. and Pio, C. (2007) *Levoglucosan levels at background sites in Europe for assessing the impact of biomass combustion on the European aerosol background.* Journal of Geophysical Research, 112, D23S05.

Quincey, P. (2007) *A relationship between black smoke index and black carbon concentration,* Atmospheric Environment, 41, 7964-7968.

Quincey, P. (2010) Personal communication, National Physical Laboratory.

Rauterberg-Wulff, A. (1998) *Beitrag des Reifen- und Bremsenabriebs zur Russimmission an Strassen*, Dissertation am Fachgebiet Luftreinhaltung der Technischen Universitat Berlin.

Rauterberg-Wulff, A. (1999) *Determination of emission factors for tyre wear particles up to 10µm by tunnel measurements*. Proceedings of 8th International Symposium 'Transport and Air Pollution', Graz, Austria, 31 May - 2 June 1999. Technical University Graz. Institute for Internal Combustion Engines and Thermodynamics.

Reiss, R., Anderson, E.L., Cross, C.E., Hidy, G., Hoel, D., McClellan, R., and Moolgavkar, S. (2007) *Evidence of health impacts of sulphate and nitrate-containing particles in ambient air*. Inhalation Toxicology, 19, 419-449.

RoTAP, (2009) *Review of Transboundary Air Pollution (RoTAP) Acidification, eutrophication, ground level ozone and heavy metals in the UK.* Draft Report for Defra and the Devolved Administrations, available at: http://www.rotap.ceh.ac.uk/documents

Routledge, J.C, Manney, S., Harrison, R.M., Ayres, J.G., and Townend, J.N. (2006) *Effect of inhaled sulphur dioxide and carbon particles on heart rate variability and markers of inflammation and coagulation in human subjects*. Heart, 92, 220-227.

Sanders, P. G., Xu, N., Dalka, T. M., and Maricq, M. M. (2003) A*irborne brake wear debris: Size distributions, composition, and a comparison of dynamometer and vehicle tests.* Environmental Science and Technology, 37, 4060-4069.

Sandradewi, J., Prevot, A.S.H., Alfarra M.R., Szidat, S., Wehrli, M.N., Ruff, M., Weimer, S., Lanz, V.A., Weingartner, E., Perron, N., Caseiro, A., Kasper-Giebl, A., Puxbaum, H., Wacker, L. and Battensperger, U. (2008) *Comparison of several wood smoke markers and source apportionment methods for wood burning particulate mass.* Atmospheric Chemistry and Physics Discussion, 8, 8091-8118.

Sarnat, J.A., Marmur, A., Klein, M., Kim, E., Russell, A.G., Sarnat, S.E., Mulholland, J.A., Hopke, P.K. and Tolbert, P.E. (2008) *Fine particle sources and cardiorespiratory morbidity: an application of chemical mass balance and factor analytical source-apportionment methods*. Environmental Health Perspectives, 116, 459-466.

Schauer, J.J., Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.r. Simoneit B.R.T. (1996) *Source apportionment of airborne particulate matter using organic compounds as tracers,* Atmospheric Environment, 30, 3837-3855.

Schins, R.P.F., Lightbody, J.H., Borm, P.J.A., Shi, T.M., Donaldson, K., and Stone, V. (2004) *Inflammatory effects of coarse and fine particulate matter in relation to chemical and biological constituents,* Toxicology and Applied Pharmacology, 195**,** 1-11.

Schlesinger, R.B. (2007) *The health impact of common inorganic components of fine particulate matter* (*$PM_{2.5}$) in ambient air: A critical review*. Inhalation Toxicology, 19, 811-832.

Seaton, A., Godden, D., MacNee, W., and Donaldson, K. (1995) *Particulate air pollution and acute health effects*. Lancet, 345, 176-178.

Simpson, D., Fargerli, H., Jonson, J. E., Tsyro, S. and Wind, P. (2003) *Transboundary acidification, eutrophication and ground level ozone in Europe. Part 1. Unified EMEP model description*. EMEP/MSC-W Report 1/2003, The Norwegian Meteorological Institute, Oslo, Norway.

Stationery Office (1999). *Pollution Prevention and Control Act, 1999.*

Stedman (2010) *Personal communication*. AEA, Harwell.

Szidat, S., Jenk, T. M., Gaggeler, H. W., Synal, H. A., Fisseha, R., Baltensperger, U., Kalberer, M., Samburova, V., Reimann, S., Kasper-Giebl, A. and Hajdas, I. (2004) *Radiocarbon (C-14)-deduced biogenic and anthropogenic contributions to organic carbon (OC) of urban aerosols from Zurich, Switzerland*, Atmospheric Environment 38, 4035-4044.

Szidat, S., Jenk, T. M., Synal, H. A., Kalberer, M., Wacker, L., Hajdas, I., Kasper-Giebl, A. and Baltensperger, U. (2006) *Contributions of fossil fuel, biomass-burning, and biogenic emissions to carbonaceous aerosols in Zurich as traced by ^{14}C*, Journal of Geophysical Research 111, D07206, doi:10.1029/2005JD006590.

Szidat, S., Ruff, M., Perron, N., Wacker, L., Synal, H. A., Hallquist, M., Shannigrahi, A. S., Yttri, K. E., Dye, C. and Simpson, D. (2009) *Fossil and non-fossil sources of organic* *carbon (OC) and elemental carbon (EC) in Goteborg, Sweden*, Atmospheric Chemistry and Physics 9, 1521-1535.

Tainio, M., Tuomisto, J.T., Pekkanen, J., Karvosenoja, N., Kupiainen, K., Porvari, P., Sofiev, M., Karppinen, A., Kangas, L. and Kukkonen, J. (2010) *Uncertainty in health risks due to anthropogenic primary fine particulate matter from different source types in Finland*. Atmospheric Environment, 44**,** 2125-2132.

Thorpe, A.J., Harrison, R.M., Boulter, P.G. and McCrae, I.S. (2007) *Estimation of particle resuspension source strength on a major London road.* Atmospheric Environment, 41, 8007–8020.

Thorpe, A. and Harrison, R. M. (2008) *Sources and properties of non-exhaust particulate matter from road traffic: A review*. Science of the Total Environment, 400, 270-282.

Tsai, F,C., Apte, M.G., and Daisey, J.M.(2000) *An exploratory analysis of the relationship between mortality and the chemical composition of airborne particulate matter*. Inhalation Toxicology, 12, 121-135.

Tzamkiozis, T., Ntziachristos, L. and Samaras, Z. (2010) *Diesel passenger car PM emissions: From Euro 1 to Euro 4 with particle filter*. Atmospheric Environment, 44, 909-916.

UIC (2006) *Exhaust emission tests for diesel traction engines*. Leaflet 624, 3rd edition. ISBN 2-7461-1040-7, Union Internationale des Chemins de Fer, Paris.

UNECE (1998) *Protocol to the 1979 convention on long-range transboundary air pollution on heavy metals*. Available at: http://www.unece.org/env/lrtap/full%20text/1998.Heavy.Metals.e.pdf

UNECE (1999) *Protocol to the 1979 convention on long-range transboundary air pollution to abate acidification, eutrophication and ground level ozone.* Available at: http://www.unece.org/env/lrtap/full%20text/1999%20Multi.E.Amended.2005.pdf

USEPA (1995). *Compilation of air pollution emission factors.* Vol.1 and Vol.2, AP-42, 5th edition.

USEPA (2006) *Emission factor documentation for AP-42, section 13.2.1 paved roads*. 11/06. Available at http://www.epa.gov/ttnchie1/ap42/ch13/final/c13s0201_2006.pdf

USEPA (2009) *Integrated Science Assessment for Particulate Matter*. EPA/600/R-08/139F, United States Environmental Protection Agency, available at: http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=216546

USEPA (2010) *Chemical mass balance (CMB) model.* United States Environmental Protection Agency, available at: http://www.epa.gov/scram001/receptor_cmb.htm

Vieno, M., Dore, A. J., Wind, P., Di Marco, C., Nemitz, E., Phillips, G., Tarrason, L. and Sutton, M. A. (2009b) *Application of the EMEP unified model to the UK with a horizontal resolution of 5 x 5 km*^{2}. Atmospheric Ammonia, 367-372.

Vieno, M., Dore, A. J., Stevenson, D. S., Doherty, R., Heal, M. R., Reis, S., Hallsworth, S., Tarrason, L., Wind, P., Fowler, D., Simpson, D. and Sutton, M. A. (2010a) *Modelling surface ozone during the 2003 heat wave in the UK*. Atmospheric Chemistry and Physics, 7963-7978.

Vieno, M. (2010b) Personal Communication. University of Edinburgh.

Wahlin, P., Berkowicz, R. and Palmgren, F. (2006) *Characterisation of traffic-generated particulate matter in Copenhagen.* Atmospheric Environment, 40, 2151–9.

Wessels, A., Birmili, W., Albrecht, C., Hellack, B., Jermann, E., Wick, G., Harrison, R.M., and Schins, R.P.F. (2010) *Oxidant generation and toxicity of size-fractionated ambient particles in human lung epithelial cells*. Environmental Science & Technology, 44, 3539-3545.

Whiting, R. (2004) *Emission factors programme Task 9 – Review of particulate matter emissions from industrial processes*. Rept. AEAT/ENV/R/1714 Issue 1, June 2004, available at: http://www.airquality.co.uk/reports/cat08/0407081216_Task9_report_issue_1.pdf

Wichmann, H.E., and Peters, A. (2000) *Epidemiological evidence of the effects of ultrafine particle exposure*. Philosophical Transactions of the Royal Society of London Series A 2000, 358**,** 2751-2768.

Wilson, W., and Suh, H. (1997) *Fine particles and coarse particles: concentration relationships relevant to epidemiological studies*. Journal of the Air and Waste Management Association, 47, 1238-1249.

WHO (2003) *Health aspects of air pollution with particulate matter, ozone and nitrogen dioxide. Report on a WHO Working Group.* EUR/03/5042688, World Health Organisation, Bonn.

WHO (2004) *Meta-analysis of time-series studies and panel studies of particulate matter and ozone*, Available from World Health Organisation at: http://www.euro.who.int/document/e82792.pdf

WHO (2005) *Air quality guidelines global update 2005: particulate matter, ozone, nitrogen dioxide and sulfur dioxide*. Available from World Health Organisation at: http://www.euro.who.int/Document/E90038.pdf

Yin, J. and Harrison, R.M. (2008) *Pragmatic mass closure study for* $PM_{1.0}$, $PM_{2.5}$ *and* PM_{10} *at roadside, urban background and rural sites*. Atmospheric Environment, 42, 980-988.

Yin, J., Harrison, R.M., Chen, Q., Rutter, A. and Schauer, J.J. (2010) *Source apportionment of fine particles at urban background and rural sites in the UK atmosphere.* Atmospheric Environment, 44, 841-851.

Zanobetti, A., Schwartz, J., Samoli, E., Gryparis, A., Touloumi, G., Peacock, J., Anderson, R.H., Le Tertre, A., Bobros, J., Celko, M., Goren, A., Forsberg, B., Michelozzi, P., Rabczenko, D., Hoyos, S.P., Wichmann, H.E. and Katsouyanni, K. (2003) *The temporal pattern of respiratory and heart disease mortality in response to air pollution.* Environmental Health Perspectives, 111, 1188-1193.

Zimmer, R.A., Reeser, W.K. and Cummins, P. (1992) *Evaluation of* PM_{10} *emission factors for paved streets.* In: Chow, J.C. and Ono, D.M. (Eds.), PM_{10} standards and non-traditional particulate source controls, pp311-323.

11. ABBREVIATIONS

Components of PM

Ba	Barium
Ca^{2+}	Calcium
Cl^-	Chloride
CM	Carbonaceous matter
Cu	Copper
EC	Elemental carbon
Fe	Iron
HNO_3	Nitric acid
K^+	Potassium
Mg^{2+}	Magnesium
Na^+	Sodium
NH_3	Ammonia
NH_4^+	Ammonium
NMVOCs	Non-methane volatile organic compounds
NO_x	Nitrogen oxides
NO_3^+	Nitrate
OC	Organic carbon
OH	Hydroxyl radical
OM	Organic matter
PM	Particulate matter
$PM_{0.1}$	Particulate matter less than 0.1 micrometres aerodynamic diameter
PM_1	Particulate matter less than 1 micrometres aerodynamic diameter
PM_{10}	Particulate matter less than 10 micrometres aerodynamic diameter
$PM_{2.5}$	Particulate matter less than 2.5 micrometres aerodynamic diameter
POA	Primary organic aerosol
PTFE	Polytetrafluoroethylene
Sb	Antimony
SO_2	Sulphur dioxide
SO_4^{2-}	Sulphate
SOA	Secondary organic aerosol
TC	Total carbon
THC	Total hydrocarbons
VOCs	Volatile organic compounds

Units

°C	degrees Celsius
kW	kilowatt
MO	Monin-Obukhov length – a measure of atmospheric stability
ppm	parts per million
$\mu g/m^3$	microgrammes per cubic metre
µm	micrometre

Organisations and Countries

AQEG	Air Quality Expert Group
CEN	European Committee for Standardisation
COMEAP	Committee on the Medical Effects of Air Pollution
Defra	Department for Environment Food and Rural Affairs
DfT	Department for Transport
EAC	Environmental Audit Committee of the House of Commons
EC	European Commission
EU	European Union
EU27	The 27 countries forming the EU
IGCB	Interdepartmental Group on Costs and Benefits
ISO	International Standards Organisation
UK	United Kingdom
USEPA	United States Environmental Protection Agency
WHO	World Health Organisation

Other

AEI	Average Exposure Index
AQMA	Air Quality Management Area
AURN	Automatic Urban and Rural Network
CAFE	Clean Air for Europe
COPD	Chronic obstructive pulmonary disease
ELR	European Load Response
ESC	European Stationary Cycle
ETC	European Transient Cycle
EUDC	Extra-Urban Driving Cycle
FDMS	Filter dynamics measurement system
HDV	Heavy Duty Vehicle
IPPC	Integrated Pollution Prevention and Control
LAPC	Local Air Pollution Control
LAQM	Local Air Quality Management
LAQN	London Air Quality Network
LCP	Large combustion plant
LTO	Landing/take-off
MCERTS	Monitoring Certification Scheme
NAEI	National Atmospheric Emissions Inventory
NEC	National emissions ceilings
NEDC	New European Driving Cycle
NERP	National Emissions Reduction Plan
NIAQN	Northern Ireland Air Quality Network
NRMM	Non-road mobile machinery
TEN	Time Extension Notice
TEOM	Tapered element oscillating microbalance
UDC	Urban Driving Cycle
VCM	Volatile Correction Model

12. APPENDICES

12.1 Appendix 1: Populations Exceeding Particular Concentrations of $PM_{2.5}$

The following Tables have been provided by Stedman (2010)

Table A 1.1 Total background area (km^2) and population in area exceeding annual mean $PM_{2.5}$ concentrations of 12 and 20 $\mu g/m^3$ in 2010.

	>12 $\mu g/m^3$		**>20 $\mu g/m^3$**	
	Area (km^2)	**Population**	**Area (km^2)**	**Population**
London	1,145	6,569,583	0	0
Rest of England	1,901	2,811,868	0	0
Scotland	0	0	0	0
Wales	5	4,846	0	0
Northern Ireland	0	0	0	0
Total	3,051	9,386,297	0	0
Percentage	1.2%	16.0%	0.0%	0.0%

Table A 1.2 Total background area (km^2) and population in area exceeding annual mean $PM_{2.5}$ concentrations of 12 and 20 $\mu g/m^3$ in 2015.

	>12 $\mu g/m^3$		**>20 $\mu g/m^3$**	
	Area (km^2)	**Population**	**Area (km^2)**	**Population**
London	573	3,970,684	0	0
Rest of England	466	593,075	0	0
Scotland	0	0	0	0
Wales	2	915	0	0
Northern Ireland	0	0	0	0
Total	1,041	4,564,673	0	0
Percentage	0.4%	7.8%	0.0%	0.0%

Table A 1.3 Total background area (km2) and population in area exceeding annual mean $PM_{2.5}$ concentrations of 12 and 20 µg/m^3 in 2020.

	>12 µg/m^3		>20 µg/m^3	
	Area (km^2)	**Population**	**Area (km^2)**	**Population**
London	271	2,141,860	0	0
Rest of England	144	183,819	0	0
Scotland	0	0	0	0
Wales	2	915	0	0
Northern Ireland	0	0	0	0
Total	417	2,326,594	0	0
Percentage	0.2%	4.0%	0.0%	0.0%

Table A 1.4 Total road length (km) exceeding an annual mean $PM_{2.5}$ concentration of 20 µg/m^3 in 2010, 2015 and 2020.

	2010	2015	2020
London	16.9	0.0	0.0
Rest of England	0.0	0.0	0.0
Scotland	0.0	0.0	0.0
Wales	0.0	0.0	0.0
Northern Ireland	0.0	0.0	0.0
Total	16.9	0.0	0.0

Table A 1.5 Population-weighted mean[a] PM$_{2.5}$ concentrations (µg/m^3) in 2010, 2015 and 2020.

	2010	2015	2020	Percentage Reduction (2010 – 2020)
Scotland	5.531	5.280	5.146	6.96%
Wales	8.257	7.991	7.847	4.96%
Northern Ireland	6.377	6.145	6.018	5.63%
Inner London	14.137	13.435	13.111	7.26%
Outer London	13.439	12.909	12.669	5.73%
Rest of England	10.596	10.225	10.026	5.38%
UK	10.307	9.928	9.731	5.59%

[a] The population-weighted mean concentration represents the average concentration exposure of each population sector to PM$_{2.5}$. The population-weighted mean concentration cannot be directly compared with the AEI, as this is based on an average PM$_{2.5}$ concentration measured over three years, at urban background sites.

12.2 Appendix 2: Emission Factors for Transport Sector

Table A 2.1 EU Emission standards for passenger cars[30]

Tier	TA date	Emission standard (g/km)				
		CO	HC	HC+NO$_x$	NO$_x$	PM
Compression Ignition (Diesel)						
Euro 1	1992.07	2.72 (3.16)	-	0.97	-	0.14
Euro 2, IDI	1996.01	1.0	-	0.7	-	0.08
Euro 2, DI	1996.01	1.0	-	0.9	-	0.10
Euro 3	2000.01	0.64	-	0.56	0.50	0.05
Euro 4	2005.01	0.50	-	0.30	0.25	0.025
Euro 5	2009.09	0.50	-	0.23	0.18	0.005[b]
Euro 6	2014.09	0.50	-	0.17	0.08	0.005[b]
Positive Ignition (Gasoline)						
Euro 1	1992.07	2.72 (3.16)	-	0.97	-	-
Euro 2	1996.01	2.2	-	0.5	-	-
Euro 3	2000.01	2.30	0.20	-	0.15	-
Euro 4	2005.01	1.0	0.10	-	0.08	-
Euro 5	2009.09	1.0	0.10	-	0.06	0.005[a,b]
Euro 6	2014.09	1.0	0.10	-	0.06	0.005[a,b]

[a] Applicable only to vehicles using DI engines.

[b] Proposed to be changed to 0.003 g/km using the PMP measurement procedure.

TA – Type approval

DI – Direct injection

IDI – Indirect injection

---

[30] Adapted from http://www.dieselnet.com/

Table A 2.2 EU emission standards for heavy-duty diesel engines[31].

			Emission standard				
Tier	**TA date**	**Test**	**CO (g/kWh)**	**HC (g/kWh)**	**NO_x (g/kWh)**	**PM (g/kWh)**	**Smoke (m^{-1})**
Euro I	1992, < 85 kW	ECE R-49	4.5	1.1	8.0	0.612	
	1992, > 85 kW		4.5	1.1	8.0	0.36	
Euro II	1996.10		4.0	1.1	7.0	0.25	
	1998.10		4.0	1.1	7.0	0.15	
Euro III	*1999.10, EEVs only*	*ESC & ELR*	*1.5*	*0.25*	*2.0*	*0.02*	*0.15*
	2000.10	ESC & ELR	2.1	0.66	5.0	0.10	0.8
Euro IV	2005.10	ESC & ELR	1.5	0.46	3.5	0.02	0.5
Euro V	2008.10	ESC & ELR	1.5	0.46	2.0	0.02	0.5
Euro VI	2013.01	ESC & ELR[a]	1.5	0.13	0.4	0.01	

[a] World harmonised test cycles to be introduced in implementing legislation.

---

[31] Adapted from http://www.dieselnet.com/

Table A 2.3 Type approval emission standards for motorcycles (two-wheel, three-wheel).

EU Directive	Engine type	Mass/ capacity	TA date	Limit values (g/km)		
				CO	HC	NO_x
97/24/EC Stage I	2-stroke	All	17/6/1999	8	4	0.1
	4-stroke	All		13	3	0.3
97/24/EC Stage II (amended in Directive 2002/51/EC)	2-stroke	<150cc	01/04/2003	5.5	1.2	0/3
		>150cc		5.5	1	0.3
	4-stroke	<150cc		5.5	1.2	0.3
		>150cc		5.5	1	0.3
97/24/EC Stage III (amended in Directive 2002/51/EC)	2-stroke	<150cc	01/01/2006	2	0.8	0.15
		>150cc		2	0.3	0.15
	4-stroke	<150cc		2	0.8	0.15
		>150cc		2	0.3	0.15

Table A 2.4 Stage III A standards for inland waterway vessels[32].

Cat.	Displacement (dm^3 per cylinder)	Date	g/kWh		
			CO	NO_x+HC	PM
V1:1	D ≤ 0.9; P > 37 kW	2007.01	5.0	7.5	0.40
V1:2	0.9 < D ≤ 1.2	2007.01	5.0	7.2	0.30
V1:3	1.2 < D ≤ 2.5	2007.01	5.0	7.2	0.20
V1:4	2.5 < D ≤ 5	2009.01	5.0	7.2	0.20
V2:1	5 < D ≤ 15	2009.01	5.0	7.8	0.27
V2:2	15 < D ≤ 20; P ≤ 3300 kW	2009.01	5.0	8.7	0.50
V2:3	15 < D ≤ 20; P > 3300 kW	2009.01	5.0	9.8	0.50
V2:4	20 < D ≤ 25	2009.01	5.0	9.8	0.50
V2:5	25 < D ≤ 30	2009.01	5.0	11.0	0.50

Table A 2.5 Stage III standards for rail traction engines[33].

Category	Net Power (kW)	Date	CO (g/kWh)	HC (g/kWh)	HC+NO_x (g/kW)h	NO_x (g/kW)h	PM (g/kWh)
Stage III A							
RC A	130 < P	2006.01	3.5	-	4.0	-	0.2
RL A	130 ≤ P ≤ 560	2007.01	3.5	-	4.0	-	0.2
RH A	P > 560	2009.01	3.5	0.5*	-	6.0	0.2
Stage III B							
RC B	130 < P	2012.01	3.5	0.19	-	2.0	0.025
R B	130 < P	2012.01	3.5	-	4.0	-	0.025

---

[32] http://www.dieselnet.com/standards/eu/nonroad.php

[33] http://www.dieselnet.com/standards/eu/nonroad.php

12.3 Appendix 3: Summary of $PM_{2.5}$ Monitoring Sites Open in 2010

Table A 3.1 Summary of $PM_{2.5}$ monitoring sites open in 2010 (as of August). Two site names are shown where they apply to the same site in two networks.

Site Name	Network	Site type group	PM_{10} monitor
Aberdeen	AURN	Background	Yes
Auchencorth Moss	AURN	Rural	Yes
Belfast Centre (Belfast Centre)	AURN (NIAQN)	Background	Yes
Birmingham Tyburn	AURN	Background	Yes
Birmingham Tyburn Roadside	AURN	Roadside	Yes
Blackpool Marton	AURN	Background	No
Bristol St Paul's	AURN	Background	Yes
Bury Roadside	AURN	Roadside	Yes
Camden (Camden Swiss Cottage)	AURN (LAQN)	Roadside	Yes
Camden Kerbside	AURN	Roadside	Yes
Cardiff Centre	AURN	Background	Yes
Carlisle Roadside	AURN	Roadside	Yes
Chepstow A48	AURN	Roadside	Yes
Chesterfield	AURN	Background	Yes
Chesterfield Roadside	AURN	Roadside	Yes
Coventry Memorial Park	AURN	Background	No
Derry (Derry Brooke park)	AURN (NIAQN)	Background	Yes
Eastbourne	AURN	Background	Yes
Edinburgh St Leonards	AURN	Background	Yes
Glasgow Centre	AURN	Background	Yes
Glasgow Kerbside	AURN	Roadside	Yes
Grangemouth	AURN	Industrial	Yes
Haringey Roadside (Haringey – Haringey Town Hall)	AURN (LAQN)	Roadside	Yes
Harwell	AURN	Rural	Yes
Hull Freetown	AURN	Background	Yes

Site Name	Network	Site type group	PM_{10} monitor
Leamington Spa	AURN	Background	Yes
Leeds Headingley Kerbside	AURN	Roadside	Yes
Leeds Centre	AURN	Background	Yes
Leicester Centre	AURN	Background	Yes
Liverpool Speke	AURN	Background	Yes
London Bexley (Bexley - Slade Green FDMS)	AURN (LAQN)	Background	No
London Bloomsbury (Camden – Bloomsbury)	AURN (LAQN)	Background	Yes
London Eltham (Greenwich – Eltham)	AURN (LAQM)	Background	Yes
London Harlington (Hillingdon – Harlington)	AURN (LAQN)	Background	Yes
London Harrow Stanmore (Harrow – Stanmore)	AURN (LAQN)	Background	Yes
London Marylebone Road (Westminster – Marylebone Road)	AURN (LAQM)	Roadside	Yes
London N. Kensington (Kensington and Chelsea – North Ken)	AURN (LAQN)	Background	Yes
London Teddington (Richmond - National Physical Laboratory)	AURN (LAQN)	Background	No
Manchester Piccadilly	AURN	Background	No
Middlesbrough	AURN	Industrial	Yes
Newcastle Centre	AURN	Background	Yes
Newport	AURN	Background	Yes
Norwich Lakenfields	AURN	Background	Yes
Nottingham Centre	AURN	Background	Yes
Oxford St Ebbes	AURN	Background	Yes
Plymouth Centre	AURN	Background	Yes
Port Talbot Margam	AURN	Industrial	Yes
Portsmouth	AURN	Background	Yes
Preston	AURN	Background	No
Reading New Town	AURN	Background	Yes

Site Name	Network	Site type group	PM_{10} monitor
Rochester Stoke	AURN	Rural	Yes
Salford Eccles	AURN	Industrial	Yes
Saltash Roadside	AURN	Roadside	Yes
Sandy Roadside	AURN	Roadside	Yes
Sheffield Centre	AURN	Background	Yes
Southampton Centre	AURN	Background	Yes
Southend-on-Sea	AURN	Background	No
Stanford-le-Hope Roadside	AURN	Roadside	Yes
Stoke-on-Trent Centre	AURN	Background	Yes
Storrington Roadside	AURN	Roadside	Yes
Sunderland Silksworth	AURN	Background	No
Swansea Roadside	AURN	Roadside	Yes
Tower Hamlets Roadside (Tower Hamlets – Blackwall)	AURN (LAQN)	Roadside	Yes
Warrington	AURN	Background	Yes
Wirral Tranmere	AURN	Background	
Wigan Centre	AURN	Background	No
York Bootham	AURN	Background	Yes
Bexley - Erith	LAQN	Background	Yes
Greenwich - Millennium Village	LAQN	Background	Yes
Greenwich - Westhorne Avenue	LAQN	Roadside	Yes
Greenwich - A206 Burrage Grove	LAQN	Roadside	Yes
Greenwich - Plumstead High Street	LAQN	Roadside	Yes
Kensington and Chelsea - Cromwell Road	LAQN	Roadside	Yes
Lisburn Dunmurry High School	NIAQN	Background	No

AURN = Automatic Urban and Rural Network

NIAQN = Northern Ireland Air Quality Network

LAQN = London Air Quality Network

12.4 Appendix 4: Measured Annual Mean Urban Background $PM_{2.5}$ in 2009

Table A 4.1 Summary of annual mean $PM_{2.5}$ concentrations at urban background sites in 2009.

Site	Annual Mean (µg/m³)
Scotland	
Edinburgh St Leonards[a]	8.5
Glasgow Centre[a]	11.7
Northern Ireland	
Belfast Centre[a]	(12)[b]
Lisburn Dunmurray High School	15.5
England (excl. London)	
Birmingham Tyburn[a]	13.6
Blackpool Marton[a]	(9)
Bristol St Paul's[a]	13.3
Chesterfield[a]	13.6
Coventry Memorial Park[a]	(13)
Hull Freetown[a]	14.0
Leamington Spa[a]	12.9
Leeds Centre[a]	13.7
Liverpool Speke[a]	(12)
Manchester Piccadilly[a]	(12)
Newcastle Centre[a]	11.0
Nottingham Centre[a]	14.1
Oxford St Ebbes[a]	11.4
Portsmouth[a]	11.3
Preston[a]	(12)
Reading New Town[a]	12.3
Sheffield Centre[a]	(14)
Southampton Centre[a]	13.4
Southend-on-Sea	13.2
Stoke-on-Trent Centre[a]	14.8

Site	Annual Mean (µg/m^3)
Warrington[a]	11.6
Wigan Centre[a]	(14)
Wirral Tranmere[a]	(9)
York Bootham[a]	12.0
Wales	
Cardiff Centre[a]	12.5
Newport	11.9
London	
Bexley Slade Green[a]	13.7
Greenwich Eltham[a]	17.6
Greenwich Millenium Village	15.5
Harrow Stanmore [a]	(13)
Bloomsbury[a]	16.3
North Kensington[a]	13.9
Teddington[a]	13.2

[a] these sites form part of Defra's network of sites for calculating the AEI

[b] values in brackets have data capture between 75 and 90%

12.5 Appendix 5: Summary of $PM_{2.5}$:PM_{10} Ratios in 2009

Table A 5.1 Summary of $PM_{2.5}$:PM_{10} ratios in 2009. Only for sites with >75% data capture for each pollutant.

Site	$PM_{2.5}$ (µg/m^3)	PM_{10} (µg/m^3)	$PM_{2.5}$:PM_{10} Ratio
Urban Background			
Belfast	12.3	20.1	0.610
Birmingham	13.6	20.4	11.7
Bristol	13.3	18.9	0.676
Cardiff	12.5	18.4	0.736
Chesterfield	13.7	17.6	0.822
London Bloomsbury	16.3	19.4	0.891
Glasgow	11.7	25.1	0.447
Hell	14.0	20.9	0.705
Leamington Spa	12.9	19.8	0.678
Leeds	13.7	21.1	0.662
Liverpool	11.8	16.2	0.735
Newcastle-upon-Tyne	11.0	15.2	0.750
Newport	11.9	21.5	0.545
Oxford St Ebbes	11.4	17.3	0.629
Reading	12.3	16.5	0.743
Southampton	13.4	18.5	0.727
Warrington	11.6	18.7	0.605
Roadside/Kerbside			
Carlisle Roadside	11.3	18.5	0.791
London Marylebone Road	21.0	35.3	0.649
Sandy Roadside	15.1	20.2	0.791
Industrial			
Grangemouth	8.6	12.9	0.661
Port Talbot	8.1	24.7	0.332
Salford Eccles	14.0	17.1	0.908

12.6 Appendix 6: Monitoring Sites Divided into 4 UK Areas

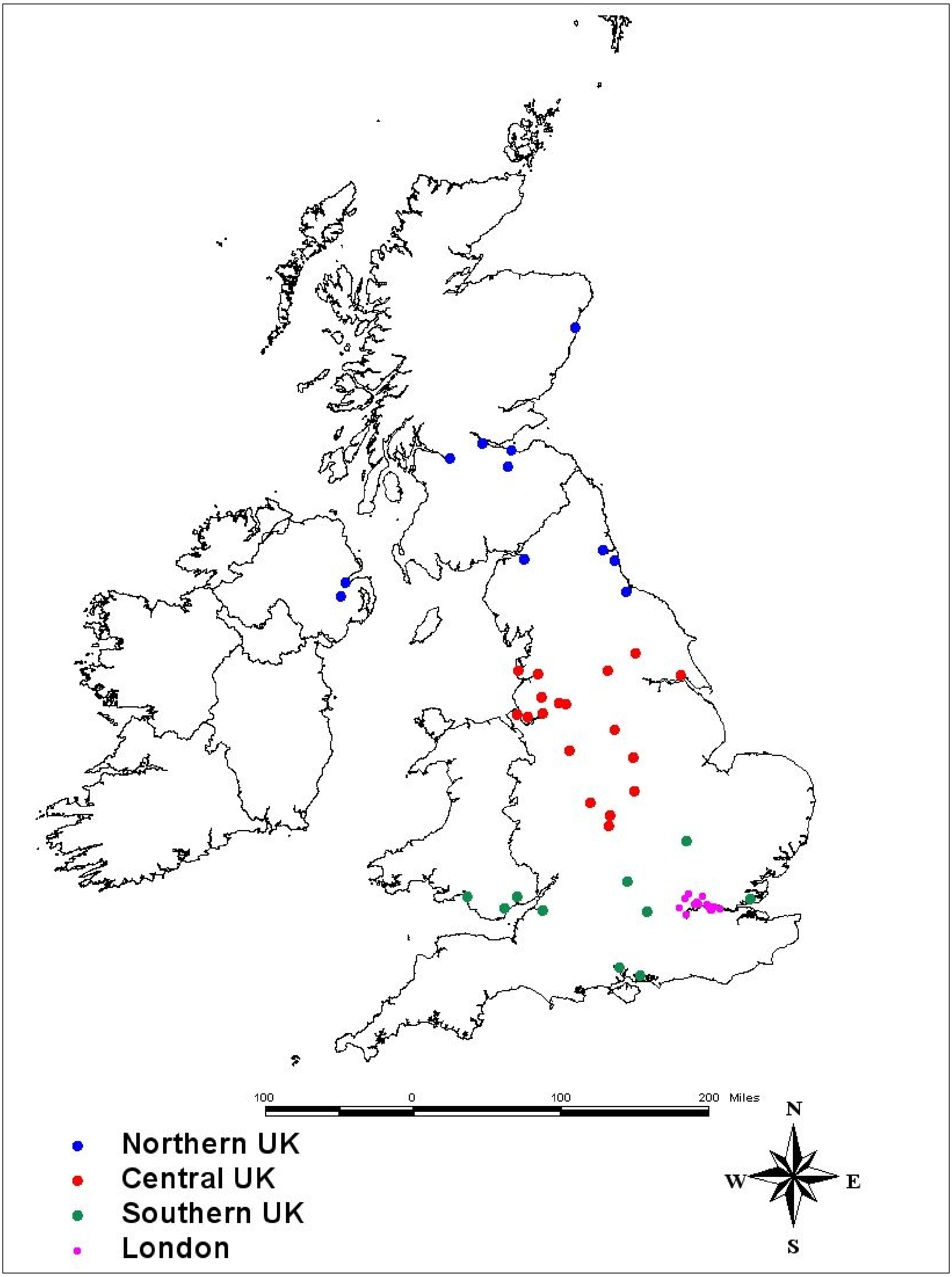

Figure A 6.1 Monitoring sites with >75% data capture in 2009, divided into four geographic regions of the UK.

12.7 Appendix 7: Summary Information on Models

Table A 7.1 Brief summary of key models of Interest

Regional-Scale Models
EMEP From a policy perspective, the most important air quality model for addressing PM across Europe (including the UK) is the Unified Eulerian European Monitoring and Evaluation Programme (EMEP) chemical transport model. This is because of this model's strategic role in the analyses that underpin the Convention on Long-Range Transboundary Air Pollution (CLRTAP). The EMEP model has been used extensively to quantify the non-linear relationships between secondary inorganic PM and SO_2, NO_x and NH_3 emissions, and for O_3. The model has 20 vertical levels (using pressure level coordinates), with a lowest layer of ~92 m, and the top of the domain at 100 hPa. The horizontal grid is a polar stereographic projection, true at 60° north, with grid-cells approximately 50 km x 50 km. Meteorological data with a 3-h resolution are used from PARLAM-PS, a dedicated version of the High Resolution Limited Area Model (HIRLAM) weather prediction model. National emissions are distributed across the EMEP grid cells and distributed vertically according to European Environment Agency Selected Nomenclature for Air Pollution (SNAP) code delineations.
EMEP4UK EMEP4UK is a nested UK version of the EMEP model running at a 50 km resolution over Europe and at a 5 km resolution over the UK. EMEP4UK has the advantage (over other UK-based statistical models) of using real-time meteorology (derived from either HIRLAM or the US Weather Research Forecast (WRF) model), enabling the interactions between emissions, meteorology, concentrations and deposition to be addressed at a fine spatial scale.
CMAQ The Community Multiscalar Air Quality (CMAQ) Eulerian chemical transport model is an open source development project of the US EPA. CMAQ was also designed to have multi-scale capabilities so that separate models were not needed for urban and regional scale air quality modelling. The target grid resolutions and domain sizes for CMAQ range spatially and temporally over several orders of magnitude. The CMAQ modelling system simulates various chemical and physical processes that are important for understanding atmospheric trace gas transformations and distributions. The CMAQ modelling system contains three sets of modelling components: meteorological, emissions and chemistry/transport.
NAME The Met Office's Lagrangian dispersion model, NAME (previously the Nuclear Accident ModeEl and now the Numerical Atmospheric dispersion Modelling Environment) simulates the release of atmospheric pollutants by releasing air parcels into a three-dimensional model atmosphere driven by three-dimensional meteorological data from the Met Office's Numerical Weather Prediction model (NWP). The air parcels are carried passively by the NWP wind fields and random walk techniques are used to simulate the local turbulent dispersion. In order to calculate the species concentrations required for the chemistry scheme, a three-dimensional grid is constructed over the model domain. The model is driven using EMEP emissions data on a 50 km grid, hence this resolution grid was also used in the horizontal for the chemistry calculations. Five vertical layers are used (0–100 m, 100–300 m, 300–800 m, 800–5000 m and 5000–20000 m). The model emits 13 primary species, including seven VOCs, that are then scaled to represent the full VOC emission inventory.
RAINS The Regional Air Pollution Information and Simulation (RAINS) model has been developed by the International Institute for Applied Systems Analysis to assess strategies to reduce acid deposition in Europe and Asia. It describes the pathways of emissions of sulphur dioxide, nitrogen oxides and ammonia. It is made up of three core modules: the emission-cost module; the acid deposition and ecosystem impact module; and the optimisation module (which is intended to identify the most cost effective emissions reduction measures to meet target deposition levels).

Regional-Scale Models

EMEP

From a policy perspective, the most important air quality model for addressing PM across Europe (including the UK) is the Unified Eulerian European Monitoring and Evaluation Programme (EMEP) chemical transport model. This is because of this model's strategic role in the analyses that underpin the Convention on Long-Range Transboundary Air Pollution (CLRTAP). The EMEP model has been used extensively to quantify the non-linear relationships between secondary inorganic PM and SO_2, NO_x and NH_3 emissions, and for O_3. The model has 20 vertical levels (using pressure level coordinates), with a lowest layer of ~92 m, and the top of the domain at 100 hPa. The horizontal grid is a polar stereographic projection, true at 60° north, with grid-cells approximately 50 km x 50 km. Meteorological data with a 3-h resolution are used from PARLAM-PS, a dedicated version of the High Resolution Limited Area Model (HIRLAM) weather prediction model. National emissions are distributed across the EMEP grid cells and distributed vertically according to European Environment Agency Selected Nomenclature for Air Pollution (SNAP) code delineations.

EMEP4UK

EMEP4UK is a nested UK version of the EMEP model running at a 50 km resolution over Europe and at a 5 km resolution over the UK. EMEP4UK has the advantage (over other UK-based statistical models) of using real-time meteorology (derived from either HIRLAM or the US Weather Research Forecast (WRF) model), enabling the interactions between emissions, meteorology, concentrations and deposition to be addressed at a fine spatial scale.

CMAQ

The Community Multiscalar Air Quality (CMAQ) Eulerian chemical transport model is an open source development project of the US EPA. CMAQ was also designed to have multi-scale capabilities so that separate models were not needed for urban and regional scale air quality modelling. The target grid resolutions and domain sizes for CMAQ range spatially and temporally over several orders of magnitude. The CMAQ modelling system simulates various chemical and physical processes that are important for understanding atmospheric trace gas transformations and distributions. The CMAQ modelling system contains three sets of modelling components: meteorological, emissions and chemistry/transport.

EURAD

The EURopean Air pollution Dispersion (EURAD) model system simulates the physical, chemical and dynamic processes which control emission, production, transport and deposition of atmospheric trace species. EURAD provides concentrations of these trace species in the troposphere over Europe and their removal from the atmosphere by wet and dry deposition. EURAD consists of three sub-models for the treatment of meteorology (NCAR/Pennstate University; MM5: Mesoscale model, Version 5), chemistry and transport (EURAD-CTM: Chemistry-Transport-Model) and emission (EURAD Emission Model). It has been applied to the simulation of a large number of air pollution episodes, focusing on ozone and other photo-oxidants, as well as aerosols. The model system has been applied to the assessment of emission changes as a contribution to the development of strategies for the reduction of air pollution levels in Europe. EURAD consists of a series of nested simulations. This enables consistent modelling of air quality from small (local) to large (regional) scales. Applications with coarse resolution usually cover most of Europe. They can be zoomed down to regions of the size of central Europe and fractions of it, *e.g.* countries. An extended System (CARLOS: Chemistry and Atmospheric transport in Regional and LOcal Scale) is under development which combines EURAD with a meteorological model of higher resolution so that finer structures of air pollutant fields can also be simulated. Daily forecasts of air quality, including aerosols, are generated within EURAD.

Regional-Scale Models

CHIMERE

The CHIMERE multi-scale chemical transport model is primarily designed to produce daily forecasts of ozone, aerosols and other pollutants and make long-term simulations (entire seasons or years) for emission control scenarios. CHIMERE runs over a range of spatial scales from the urban scale (100-200 km) to the regional scale (several thousand kilometres) with resolutions from 1-2 km to 100 km.

FRAME

The FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model is a Lagrangian atmospheric transport model used to assess the long-term annual mean deposition of reduced and oxidised nitrogen and sulphur over the United Kingdom. FRAME was developed initially to focus in particular on transport and deposition of reduced nitrogen. Recent developments in the treatment of sulphur and oxidised nitrogen mean that it may now be considered as a robust multi-chemical species tool. Variations of FRAME are now also used to simulate transport and deposition of heavy metals, particles, base cations, and greenhouse gases such as methane and nitrous oxide.

UK PCM

The UK Pollution Climate Mapping model is designed to calculate UK-wide maps of annual average pollutant concentration at sufficient speed that extensive scenario testing can be conducted. The background maps provide concentrations at 1 km resolution, and a single representative concentration for each road segment is used to represent a roadside increment.

The maps of background concentrations have been calculated by summing contributions from different sources:

- Secondary inorganic aerosol (derived by interpolation and scaling of measurements of SO_4^{2-}, NO_3^- and NH_4^+ at rural sites)
- Secondary organic aerosol (semi-volatile organic compounds formed by the oxidation of non-methane volatile organic compounds. Estimates derived from results from the HARM/ELMO model)
- Large point sources of primary particles (modelled using ADMS and emissions estimates from the National Atmospheric Emissions Inventory (NAEI))
- Small point sources of primary particles (modelled using the small points model and emissions estimates from the NAEI)
- Regional primary particles (from results from the TRACK model and emissions estimates from the NAEI and EMEP)
- Area sources of primary particles (modelled using a dispersion kernel and emissions estimates from the NAEI)
- Rural calcium rich dusts from re-suspension of soils (modelled using a dispersion kernel and information on land use)
- Urban calcium rich dusts from re-suspension of soils due to urban activity (estimated from a combination of measurements made in Birmingham and population density)
- Regional iron rich dusts from re-suspension (assumed to be a constant value, estimated measurements made in the vicinity of Birmingham)
- Iron rich dusts from re-suspension due to vehicle activity (modelled using a dispersion kernel land and vehicle activity data for heavy-duty vehicles)
- Sea salt (derived by interpolation and scaling of measurements of chloride at rural sites)
- Residual (assumed to be a constant value)

The concentrations of many of these components have been estimated separately for the fine and coarse fraction. This enables a consistent method to be adopted for estimation of PM$_{10}$ (the sum of the fine and coarse fractions) and PM$_{2.5}$ (fine fractions only). These component pieces are then aggregated to a single 1 km x 1 km background grid. An additional roadside increment is added for roadside locations.

UK PTM

The UK Photochemical Trajectory Model takes account of long range transport of pollutants to predict episodes of photochemically generated pollutants during the summer. It uses a simplified chemical model to predict the formation of ozone as air travels to the UK, and considers air mass back trajectories for the preceding 96 hours. The model can also assess the contribution of secondary PM.

HARM

The Hull Acid Rain Model (HARM) was developed to model acidifying pollutants in the UK. Model outputs are compared with sulphur and nitrogen deposition data using linear regression to predict deposition patterns for sulphur and oxidised and reduced nitrogen. Its limitations include the fact that it underestimates dry deposition of reduced nitrogen. The model has been used to help formulate UK strategies relating to emissions for acidifying pollutants. It does not provide a complete picture of $PM_{2.5}$.

OSRM

The Ozone Source-Receptor Model (OSRM) was developed to help define UK ozone policy, but includes some $PM_{2.5}$ precursor species. It is a Lagrangian trajectory model used to model photochemical ozone production within the UK. OSRM has a surface conversion module which enables modelling of vertical gradients. It does not provide a complete picture of $PM_{2.5}$.

STOCHEM

STOCHEM is the UK Met Office Global Three-Dimensional Lagrangian model. It is used to model a range of species (including greenhouse gases, oxidants and aerosols) whilst considering anthropogenic and climatic influences from pre-industrial times to the year 2100. The model describes the transport and chemistry of NO_x, CO, methane, VOCs SOx and ozone. It is a complex model and takes account of convective mixing processes, dry deposition, chemical reactions including thermal and photolytic processes, chemical kinetics and meteorological data including wind speed, temperature, humidity, cloud cover, precipitation, surface pressure and stratospheric ozone fields. However, its horizontal spatial resolution is very coarse (5° x 5°).It does not provide a complete picture of $PM_{2.5}$.

Derwent Chemical Model

Derwent *et al.* (2009) developed a chemical transformation model to simulate conversion of primary precursors into PM. They note that their model is simplistic, representing a first attempt only. PM formation is described using a kinetic approach to represent constraints on establishing a thermodynamic equilibrium. Photolysis takes account of instantaneous solar angles. Gas and aerosol dry deposition were counted, but wet deposition was not.

CAMx

The Comprehensive Air quality Model with extensions (CAMx) is an Eulerian photochemical dispersion model that allows for integrated 'one-atmosphere' assessments of gaseous and particulate air pollution over many scales ranging from sub-urban to continental. CAMx works with many different meteorological models (*e.g.*, MM5, RAMS, and WRF) and emission inputs developed using many emissions processors (SMOKE, CONCEPT, EPS, EMS). The US EPA has approved the use of CAMx for numerous ozone and PM State Implementation Plans throughout the U.S, and has used this model to evaluate regional mitigation strategies. CAMx is also used throughout the world to support a wide variety of research and regulatory activities.

REMSAD

The REgional Modelling System for Aerosols and Deposition (REMSAD) is used extensively in the US. It was originally intended as a screening tool – a model that could be run (quickly) for a continental-scale modelling domain (specifically the continental US) and for a full-year simulation period – to provide information (although not very detailed) on the distribution and composition of particulate matter, the deposition of pollutant species onto the surfaces of inland and coastal bodies of water, and the expected change in air quality and deposition that results from changes in emissions. All of these parameters were intended to be primarily represented in terms of seasonal or annual averages or deposition totals. The model has evolved into a more complex 'one atmosphere' modelling system that simulates the chemistry, transport, and deposition of airborne pollutants (with emphasis on PM, ozone, and mercury). The REMSAD model is capable of 'nesting' one or more finer-scale subgrids within a coarser overall grid. This two-way fully interactive nesting feature allows the user to apply higher resolution over selected source and/or receptor regions. The modelling system may be applied at scales ranging from a single metropolitan area to a continent containing multiple urban areas; although to date, most applications have focused on the continental-scale.

Urban-Scale Models

ADMS-Urban

The Atmospheric Dispersion Modelling System is produced by Cambridge Environmental Research Consultants. It can be used to examine emissions from 6000 sources simultaneously and output concentrations across wide areas. It allows line, point, area, volume, or jet sources to be included. ADMS-Urban incorporates the latest (quasi Gaussian) understanding of the boundary layer structure, with explicit calculation of key parameters. The model uses advanced algorithms for the height-dependence of wind speed, turbulence and stability to produce improved predictions. It allows up to 500 annual hourly emission profiles and up to 50 monthly emission profiles. Wet and dry deposition can be included, but secondary $PM_{2.5}$ formation cannot. Concentrations can be output on averaging times ranging from one hour to annual means. Emissions can be taken from any available source, but ADMS-Urban is often used in combination with the EMIT emissions model. EMIT is both a database that can be used to organise an emissions inventory, and an emissions calculator in its own right, containing a range of embedded emission factors.

The London Air Quality Model

The Kings College Environmental Research Group London Air Quality Model treats $PM_{2.5}$ as the sum of two components :

$PM_{2.5}$ related to NO_x:- $PM_{2.5}$ f(NO_x) (primary emissions)

$PM_{2.5}$ independent of NO_x (assumed to be secondary aerosol)

Rolling annual mean measurements of NO_x, PM_{10} and $PM_{2.5}$ have been taken at a number of co-located monitoring sites across London. Relationships between annual mean NO_x and $PM_{2.5}$ were established using linear regression. Time series' of the gradients and of the intercepts of the regression relationships were calculated. For $PM_{2.5}$, only regression relationships with an r^2 value of >0.75 were used. The annual mean concentrations of the $PM_{2.5}$ component that is dependent on NO_x can be calculated from the average of the gradients. The annual mean concentrations of the independent components can be calculated from the time series of the intercepts. A time series of daily means for each of the particulate components was calculated by applying the factors derived from the regression equations to the daily mean NO_x, and $PM_{2.5}$ measured at each of the sites. This allowed the calculation of the NO_x dependent components. The non-NO_x component was then calculated by subtraction. Predictions were then made at other locations. It is assumed that the daily mean NO_x independent components across the area are independent of site location. The daily mean variation of NO_x is required to derive the NO_x dependent components and this was predicted using (ADMS-based) dispersion modelling.

AIRVIRO

AIRVIRO is an integrated system for data handling, emission inventories and dispersion modelling. It includes a series of separate dispersion models: the Gauss model, the Grid model and the Canyon model. It thus offers an Eulerian flow and diffusion scheme as an alternative to a Gaussian model. AIRVIRO can be run at a range of resolutions, with the finest usually being a 20 m x 20 m grid. These averages are seldom useful for comparing predicted concentrations against limit values, but can be more useful for applications such as exposure reduction. AIRVIRO is currently being updated to incorporate the latest UK road vehicle EFT.

Local-Scale Models

ADMS-Roads, ADMS-4, ADMS-Screen, and ADMS-Airports.

These models share many of the features of ADMS-Urban, but allow fewer sources to be included and are thus tailored toward local-scale assessments. ADMS-Roads is specifically for roads assessments, but does allow limited inclusion of point, area and volume sources. ADMS-4 is intended for industrial modelling and allows line, point, area, volume, or jet sources to be modelled. ADMS-Airports is designed around the requirements of airport modelling, including features such as moving jet sources. ADMS-Screen is a screening model which does not take account of local meteorological conditions, but does account for regional meteorological trends, surface roughness, and the effects of a single building on dispersion of pollutants from a single point source. All of these models allow emissions to be entered by hand from any available source. In addition, ADMS-Roads includes inbuilt emission factors. The current road-vehicle emission factor dataset used in ADMS-Roads has been superseded and work is currently underway to incorporate the latest UK road vehicle EFT.

AERMOD

AERMOD is produced by the US EPA and is commonly used across the world. It is distributed free of charge, but Graphical User Interfaces, featuring additional functionality, have been produced commercially. It is a steady-state plume model that incorporates air dispersion based on boundary layer turbulence structure and scaling concepts, including treatment of both surface and elevated sources, and both simple and complex terrain effects (using a terrain pre-processor AERMAP). AERMOD can model multiple emission sources (vertical and horizontal point, area, volume and open pit) to predict concentrations at discrete receptor points and/or across contour grids. AERMOD assumes a Gaussian horizontal plume distribution. Treatment of vertical plume distribution depends on atmospheric stability. It uses hour-by-hour meteorological files from local measurements and features a meteorological data pre-processor (AERMET) that calculates the atmospheric parameters needed by the model. This can take account of local data such as direction-specific values of surface roughness etc. The model can be run with PRIME (Plume RIse Model Enhancements) which allows for building downwash. AERMOD can model deposition but is not constrained to conserve mass and this can lead to considerably more substance deposited than is released.

ISC

The Industrial Source Complex model is currently not one of the US EPA’s preferred regulatory models and is not widely used in the UK. The latest version of ISC is ISC3. It is a steady-state Gaussian plume model which can be used to assess pollutant concentrations from a wide variety of sources associated with an industrial complex. The model can account for: settling and dry deposition of particles; downwash; point, area, line, and volume sources; plume rise as a function of downwind distance; separation of point sources; and limited terrain adjustment. ISC3 operates in both long-term and short-term modes (ISCLT3 and ISCST3 respectively). PRIME can be used with ISC3.

SCREEN3

SCREEN3 is a screening version of ISC3. It is a single source Gaussian plume model which provides maximum ground-level concentrations for point, area, flare, and volume sources. It is based on the same steady-state Gaussian plume algorithms as ISC and is applicable for estimating ambient impacts out to a distance of about 50 km. SCREEN3 also includes algorithms for addressing building downwash influences, including the cavity recirculation region, and incorporates the Valley 24-hour screening algorithm for estimating complex terrain impacts.

CALPUFF

CALPUFF is a multi-layer, multi-species non-steady-state puff dispersion model that simulates the effects of time- and space-varying meteorological conditions on pollution transport, transformation and removal. CALPUFF can be applied on scales of tens to hundreds of km. It includes algorithms for subgrid scale effects (such as terrain impingement), as well as, longer range effects (such as pollutant removal due to wet scavenging and dry deposition, and chemical transformation). Anecdotal evidence from the US is that CALPUFF is a much better deposition model than AERMOD and is often used in preference where deposition is of concern over these spatial scales.

CALINE

The California Line Source Dispersion Model is currently available as CALINE3 and CALINE4. It is a roads model based on Gaussian equations, employing a mixing zone concept to characterize pollutant dispersion over the roadway. CALINE3 and CALINE4 use different methods for developing vertical and horizontal dispersion curves, but the differences are described by the model developers as 'fine tuning'. CALINE3 is recommended for US regulatory purposes, but CALINE4 is used in the UK – mainly via incorporation in the AAQuIRE and BREEZE Roads models. CAL3QHC is a CALINE3 based model with queuing and hot spot calculations, and with a traffic model to calculate delays and queues that occur at signalized intersections; CAL3QHCR is a more refined version based on CAL3QHC that requires local meteorological data.

BREEZE ROADS is an enhanced version of the CALINE4, CAL3QHC, and CAL3QHCR series of models that incorporates methods for estimating queue lengths and the contribution from idling vehicles.

AAQuIRE provides a simple, MS Access based, user interface for both CALINE4 and AERMOD. Historically, AAQuIRE has had issues 'dropping' sources from its input inventories and double-counting output sources. An awareness of these limitations, along with considerable care, is thus required when using AAQuIRE.

DMRB

The Highways Agency's Design Manual for Roads and Bridges is a simple screening tool for road traffic impacts. It includes PM$_{10}$ but not PM$_{2.5}$ and relies on emission factors which have now been superseded. Work is, however, underway to update the DMRB model using the latest UK road vehicle EFT and the update is likely to include PM$_{2.5}$.

CFD

Computational Fluid Dynamics provides a detailed mechanistic way of predicting dispersion and dilution in the immediate vicinity of a source. The computational demands of this method are such that it is not usually used to predict long-term concentrations. It is thus not of immediate interest to this review.

UK Road Vehicle EFT

The UK Road Vehicle Emission Factor Toolkit has recently been updated to V4.2. It is produced by Defra and is used in the majority of local-scale road traffic modelling studies. It uses speed/exhaust-emission equations issued by the DfT for specific vehicle categories. These are combined with existing and projected future vehicle fleet compositions from the NAEI. The EFT incorporates estimated emissions from brake and tyre wear but, excludes resuspension. The EFT is routinely used to predict emissions from specific sections of road according to the average speed of vehicles on that road. This is, however, an inappropriate use of the DfT emission factors, which relate to average drive cycle speeds. There are potentially large differences between, for example, the emission from a vehicle which has averaged 50 km/h across its entire journey, and the emission from a vehicle travelling at 50 km/h. Most modellers are aware of these issues but have no alternative than to use the EFT. An alternative might come from instantaneous emission factors, which have been successfully applied in some studies but there are still problems with applying these to most traffic studies.

Other Models

Other Models

The following models are also available for local-scale assessments, but are seldom used, particularly in the UK.

The **Complex Terrain Dispersion Model** Plus Algorithms for Unstable Situations (CTDMPLUS) is a refined point source Gaussian air quality model for use in all stability conditions for complex terrain. CTSCREEN is the screening version of CTDMPLUS.

Air Force Dispersion Assessment Model (ADAM) is a modified box and Gaussian dispersion model which incorporates thermodynamics, chemistry, heat transfer, aerosol loading, and dense gas effects.

DEGADIS simulates the atmospheric dispersion at ground-level of area-source dense gas (or aerosol) clouds released with zero momentum into the atmospheric boundary layer over flat, level terrain. The model describes the dispersion processes which accompany the ensuing gravity-driven flow and entrainment of the gas into the boundary layer.

RAPTAD is a 3-dimensional Lagrangian random puff model for pollutant transport and diffusion. It is used in the US for prediction of transport and diffusion processes for complex terrain, coastal regions, urban areas, and around buildings where conventional models fail.

The **HYbrid ROADway** Model (HYROAD) is used in the US. It integrates three historically individual modules that simulate the effects of traffic, emissions and dispersion. The traffic module is a micro-scale transportation model which simulates individual vehicle movements. The emission module uses speed distributions from the traffic module to determine composite emission factors; spatial and temporal distribution of emissions is based on the vehicle operation simulations. The dispersion module uses a Lagrangian puff formulation, along with a gridded non-uniform wind and stability field derived from traffic module outputs, to describe near-roadway dispersion characteristics. HYROAD is designed to determine hourly concentrations from vehicle emissions at receptor locations that occur within 500 meters of the roadway intersections.

Panache is an Eulerian (and Lagrangian for particulate matter), 3-dimensional finite volume fluid mechanics model designed to simulate continuous and short-term pollutant dispersion in the atmosphere, in simple or complex terrain.

The **Second-order Closure Integrated PUFF** Model (SCIPUFF) is a Lagrangian puff dispersion model that uses a collection of Gaussian puffs to predict three-dimensional, time-dependent pollutant concentrations. In addition to average concentrations, SCIPUFF provides a prediction of the statistical variance in the concentration field resulting from the random fluctuations in the wind field.

The **Shoreline Dispersion Model** (SDM) is a multiple-point Gaussian dispersion model that can be used to determine ground-level concentrations from tall stationary point source emissions near a shoreline.

The **Offshore and Coastal Dispersion** Model Version 5 (OCD) is a straight line Gaussian model developed to determine the impact of offshore emissions from point, area or line sources on the air quality of coastal regions. OCD incorporates overwater plume transport and dispersion as well as changes that occur as the plume crosses the shoreline. Hourly meteorological data are needed from both offshore and onshore locations.

Table A 7.2 Summary data on key models (adapted from ROTAP (2009) and AQEG (2005))

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
			Regional-Scale Models			
Unified EMEP	Daily, monthly and weekly variations of the emissions for each SNAP sector. Emitted species are: NH_3, NO_x, SO_2, NMVOC, PM_{10}, $PM_{2.5}$, CO	Eulerian	50 x 50 km	3-h resolution from PARLAM-PS	Includes ammonium chemistry, gas and aqueous, oxidation of SO_2 to sulphate, and VOC-NO_x-O_3 gaseous photochemistry.	Coarse output resolution. Uncertainties in physicochemistry describing secondary organic aerosol
EMEP4UK	As above	Eulerian	5 × 5 km	Real time data from NWP model	As above	Uncertainties in physicochemistry describing secondary organic aerosol
CMAQ	Annual anthropogenic emissions from EMEP (Europe) and NAEI (UK) by SNAP sectors, disaggregated using spatial surrogates and temporal profiles. Biogenic emissions computed online within the meteorological model. Point sources include stack parameters	Eulerian	European ~50 km, Regional ~5 km, Urban/Local ~1 km	MM5 or WRF or UM	Different chemical mechanisms (CB-IV, CB05, SAPRAC-99 and RADM2) and solvers (EBI, SMVGEAR) are available	Uncertainties in physicochemistry describing secondary organic aerosol

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
NAME	EMEP 50x50 km data	Lagrangian	50 x 50 km – 15 x15 km	from NWP model	Inorganic	Uncertainties in physicochemistry describing secondary organic aerosol
CHIMERE	-	-	1 km – 100 km	-	-	-
FRAME	Annual emissions NAEI SNAP sector (SO_2 & NO_x) Agricultural sector (NH_3) Point sources include stack parameters	Statistical Lagrangian	1 x 1 km or 5 x 5 km (UK) 50 x 50 km (Europe)	UKMO mapped annual precipitation. Statistical wind frequency and speed from radiosonde data	20 chemical species, pseudo first order reaction rates, gas and aqueous oxidation of SO_2 to sulphate, gas phase and particle formation nitrogen chemistry	No organic aerosol representation
UK PCM	NAEI SNAP sector, annual emissions (NO_x). Point sources include stack parameters.	Regression analysis based on ADMS calculations and monitoring data for urban background; simple formulation for representative calculation on road segments	1 x1 km	Hour-by-hour met data from met monitoring sites	Empirical relationship between urban decrement and local NO_x concentrations. No PM chemistry	Spatial resolution, simplistic treatment of local road sources and lack of PM chemistry. Largely empirical rather than physicochemical process based

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
UK PTM	Annual emissions with day-of-week and diurnal variations by SNAP sector NAEI and EMEP for SO_2, NO_x, NH_3, CO, CH_4 and VOCs, Isoprene from EMEP, Terpenes from GEIA 175 VOCs from 248 SNAP sectors	Lagrangian	10 x 10 km (UK) 50 x 50 km and 150 x 150 km (Europe)	UK Met Office NAME archive HYSPLIT/NCEP BADC/UK MO	MCM v3.1: 4414 species, 12871 reactions, 175 VOCs MCM CRIv2: 434 species, 1183 reactions, 115 VOCs CBM4: 36 species, 93 reactions, 8 VOCs	Lowest model layer is full boundary layer depth, *i.e.* no sub-boundary layer vertical resolution
HARM	Boundary values from STOCHEM. Annual emissions from EMEP grid, nested UK emissions from NAEI SNAP. Emitted species: SO_2, NO_x, NH_3, HCl, PM_{10}. Emission height into 3 layers based on data in inventories.	Statistical Lagrangian	Standard model 10 x 10 km for UK. Within EMEP 50 x 50 km	Annual gridded precipitation from U KMO, wind speed and frequency based on U KMO Unified model	24 trace constituent species, oxidation by O_3, OH and aqueous phase (for S). Nitrate production from NO_2 via HNO_3 and N_2O_5 and from ammonia via HNO_3	No organic aerosol representation

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
OSRM	Daily, monthly and weekly variations of the emissions (a) derived from the NAEI for the UK [and assigned to 8 OSRM sectors] and (b) taken from EMEP for non-UK sources. Emitted species are: NO_x, NMVOC, SO_2, CO. The emission of biogenic VOCs based on a bespoke emission potential inventory	Lagrangian	Notionally 10 km x 10 km	UK Met Office NAME archive	Mechanism has 70 species, 200 thermal and photochemical reactions and 12 emitted NMVOCs including isoprene. Derived from STOCHEM scheme with extra reactions added: (i) HONO chemistry; (ii) reactions of peroxy radicals with NO_3 and (iii) formation of organic nitrates. DMS chemistry replaced.	While some $PM_{2.5}$ precursors are considered, the model does not explicitly consider $PM_{2.5}$.
STOCHEM	Annual anthropogenic. Monthly varying natural and biomass burning. Biogenic VOC diurnal. Biogenic VOC and lightning NO_x linked to meteorology.	Global Lagrangian	5 degrees (~400 km at UK latitudes)	Coupled to Hadley Centre climate models or driven from archived Unified Model output	70 species, 12 emitted NMVOCs including isoprene, aqueous-phase S including DMS.	Very coarse spatial resolution. No organic aerosol representation
Derwent Chemical Model	Extension of UKPTM	Lagrangian	n/a	For each mid-afternoon of each day of 2006, a set of 30 equally probably and randomly selected 96-h air parcel trajectories from NAME.	Chemical kinetic approach based on NH_4NO_3-HNO_3-NH_3 system	Experimental model described by authors as ‘first attempt’

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
			Urban-Scale Models			
ADMS-Urban	Hour-by-hour emissions profiles – usually managed using the EMIT programme. Any relevant emissions (of conservative species) can be entered as line, point, area, volume, or jet sources.	Quasi Gaussian using h/M_{MO} parameterisation of boundary layer.	<1 m	Hour-by-hour met data from met monitoring sites	NO_x-O_3 chemistry but not PM formation	Frequently under-predicts road traffic impacts, but local verification usually not feasible for $PM_{2.5}$. Treatment of dispersion is simplistic, particularly in respect of dispersion around buildings.
London Air Quality Model	All emission types, set up for London	Regression analysis based on ADMS calculations and monitoring data	<1 m	Hour-by-hour met data from Heathrow Airport	No PM chemistry	Simplistic road traffic dataset gives more spatial detail in outputs than is available in inputs. Simplistic treatment of junctions. No inclusion of resuspension.
AIRVIRO	Any relevant emissions (of conservative species) can be entered as point, area, or line sources.	Eulerian and/ or Gaussian	500 x 500 m to 20 x 20 m	Hour-by-hour wind speed and direction	Not included	Coarse spatial resolution of outputs.

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
			Local-Scale Models			
ADMS (Roads, 4, *etc.*)	Hour-by-hour emissions profiles. Any relevant emissions (of conservative species) can be entered as line, point, area, volume, or jet sources (depending on which particular model is used).	Quasi Gaussian using h/M_{MO} parameterisation of boundary layer. Can account for the effects of building downwash, complex terrain, street canyons, and elevated roads.	<1 m	Hour-by-hour met data from met monitoring sites	NO_x-O_3 chemistry but not PM formation	Frequently under-predicts road traffic impacts, but local verification usually not feasible for $PM_{2.5}$. Treatment of dispersion is simplistic, particularly in respect of dispersion around buildings.
Aermod	Hour-by-hour variable. Vertical and horizontal point, area, volume and open pit.	Advanced Gaussian. Can account for the effects of building downwash and complex terrain.	<1 m	Hour-by-hour met data from met monitoring sites	No PM chemistry	Difficult to operate without 3rd-party GUI. Treatment of dispersion is simplistic, particularly in respect of dispersion around buildings.

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
Screen3	Fixed emissions. Single source scenarios. Point, area, volume and flare.	Gaussian. Can account for effects of simple or complex terrain.	na	Fixed emissions. Single source scenarios. Point, area, volume and flare.	Not included	Treatment of dispersion is simplistic, particularly in respect of dispersion around buildings.
ISC	Hour-by-hour variable. Vertical and horizontal point, area, volume, flare, area line and open pit.	Gaussian. Can account for effects of building downwash.	<1 m	Hour-by-hour met data from met monitoring sites	No PM chemistry	Treatment of dispersion is simplistic, particularly in respect of dispersion around buildings.
CALINE	Line sources only.	Gaussian. Can account for effects of elevated and depressed roads, and street canyons.	<1 m	Hour-by-hour met data from met monitoring sites	No PM chemistry	Developed for Californian roads and not ideally suited to narrower UK road environments, where it frequently under-predicts.
Breeze Roads	As for CALINE	As for CALINE	As for CALINE	As for CALINE	As for CALINE	As for CALINE
AAQuIRE	As for CALINE/AERMOD	As for CALINE /AERMOD	As for CALINE /AERMOD	As for CALINE /AERMOD	As for CALINE /AERMOD	As for CALINE/AERMOD

Model	Emissions	Transport / Dispersion	Horizontal Resolution	Meteorology	Chemistry	Key Limitations for $PM_{2.5}$ modelling
CFD	Infinitely variable	Computational fluid dynamics	<1 m	Short-term	No PM chemistry	Computational requirements usually prohibit modelling annual mean concentrations.