

PM_{2.5} in Scotland: A Report for SEPA

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Experts in air quality management & assessment



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1 Executive Summary

- 1.1 The principal objective of this report is to provide SEPA with a better understanding of PM_{2.5} in Scotland. This will help SEPA determine how it can contribute to meeting European Union (EU) and UK exposure-reduction targets.
- 1.2 Information is provided on PM_{2.5} concentrations and sources in Scotland, based on an examination of monitoring data and the application of regional models to examine concentrations arising from Scotland's emissions. Account has been paid to both primary and secondary PM_{2.5}. In addition to examining concentrations throughout Scotland, specific attention has been paid to concentrations in Grangemouth, where a PM_{2.5} monitor is located near to an industrial area, and more generally to emissions from sources regulated by SEPA. Exposure to PM_{2.5} has been evaluated in relation to health criteria, including the new Daily Air Quality Index used to provide information to the public on their exposure to high levels of PM_{2.5}. Finally, opportunities for SEPA to contribute to the reduction of exposure to PM_{2.5} are evaluated. The following is a summary of the findings:

Concentrations

- PM_{2.5} concentrations have been measured using FDMS monitors at six locations within the Scottish Automatic Urban Network (SAUN) and the Scottish Automatic Rural Network (SARN), since 2009. Data from these monitors have been analysed in various ways and the findings are presented in this report.
- The 1-hour mean PM_{2.5} data for 2011 have been analysed in relation to the pathway that the air has followed for the 96-hours prior to its arrival at three monitoring station, Edinburgh St Leonard's, Grangemouth and Auchencorth Moss. The patterns for all three sites are similar, with the lowest PM_{2.5} concentrations in the Edinburgh region being associated with air from the Atlantic. The highest PM_{2.5} concentrations are associated with air brought in from northern and eastern Europe. There is also evidence of some elevation of PM_{2.5} concentrations for air that has passed over England. These higher concentrations associated with air from Europe and England will mainly represent secondary PM formed from precursor emissions from these regions.
- Analysis of the 1-hour mean PM_{2.5} concentrations at all sites shows that emissions from bonfires and fireworks, associated with the Bonfire Night celebrations, can be responsible for short-term exposure to very high concentrations of PM_{2.5}.
- PM_{2.5} episodes related to stagnant winter conditions, which allow a build-up of local emissions, can last several days and represent an important contribution to the annual mean concentrations.



- Predictions from the European Monitoring and Evaluation Programme (EMEP) (regional scale) model show a strong trend of declining concentrations of total-PM_{2.5}, primary-PM_{2.5}, sulphate, nitrate and ammonium from south to north across the UK, largely reflecting the greater distance from the major source areas in continental Europe.
- The national Pollution Climate Mapping (PCM) model predictions show annual mean background PM_{2.5} concentrations across Scotland in 2011 that range from 3 to 11 µg/m³, with the highest values in the urban areas, especially Glasgow and Edinburgh. The general pattern is of higher concentrations across the central belt of Scotland, and up the east coast to Aberdeen, with the lowest concentrations being in northwest Scotland.
- The highest measured concentrations of PM_{2.5} are those at the kerbside site in Glasgow. The monitor is within 1 m of the kerb of a busy street with a canyon like setting, in the centre of Glasgow.

Emissions

- No single source dominates the emission of primary PM_{2.5} from within Scotland. The three most important sources are road transport, domestic combustion and industry, which together account for 70% of the total.
- Total emissions from within Scotland of primary-PM_{2.5}, and the precursor gases, nitrogen oxides, sulphur dioxide and ammonia, have all declined over the period 1990-2009.
- Three sources dominate emissions of primary-PM_{2.5} from Scotland's industrial and power station sector, which together account for 61% of the total; these sources are power stations, mobile and stationary combustion sources in manufacturing industries, and construction.
- SEPA regulates the power station emissions sector, as well as sources within the industrial, waste and agricultural sectors. These sectors together account for around 40% of primary-PM_{2.5} emissions in Scotland, most being from the industrial sector. Emissions of the precursor gases in Scotland are dominated by power stations in the case of sulphur dioxide (67%), power stations and road traffic in the case of nitrogen oxides (27% and 30% respectively), and agriculture in the case of ammonia (86%).

Sources

- Many sources contribute to PM_{2.5} concentrations in Scotland.
- The background concentrations derived from the PCM model for the three urban background and one industrial PM_{2.5} monitoring sites in 2011, show:

Secondary PM_{2.5} is dominant at all four sites, accounting for around 30-40% of the total modelled background $PM_{2.5}$ concentrations;



Road traffic, which includes vehicle exhaust, brake and tyre wear, and road abrasion, is an important contributor to modelled background $PM_{2.5}$ concentrations at all sites, but especially in Glasgow (27%) and less so in Aberdeen (10%);

Industry is an important contributor to modelled background $PM_{2.5}$ concentrations at some sites, being most significant in Aberdeen (16%) and less so in Edinburgh (3%). Industry accounts for 10% of the modelled background $PM_{2.5}$ concentration at the Grangemouth industrial site;

Off-road mobile machinery can also be important, ranging from a 5% contribution to the modelled background $PM_{2.5}$ concentration in Aberdeen, to a 14% contribution in Grangemouth;

Dust, at both urban and rural sites accounts for some 7-10% of the modelled background $PM_{2.5}$ concentrations (dust will arise from a range of sources including soils and construction activities; while dust encompasses PM up to 75 µm, it also includes $PM_{2.5}$). These emissions are likely to be difficult to control; and

Long-range transport of primary-PM_{2.5} (which covers sources more than 10 km away from the grid square) makes a minimal contribution at all sites (3-4%).

- Point (industrial) sources contribute 0.1-0.3 µg/m³ to PM_{2.5} concentrations within a large area across the central belt of Scotland, between Glasgow and Edinburgh, rising to 3- 4 µg/m³ in two 1x1 km grid squares within the Grangemouth industrial complex, according to the PCM model.
- Results from the EMEP model confirm the analysis of PM_{2.5} concentrations in relation to air mass trajectories, showing that PM_{2.5} concentrations across the UK are significantly associated with emissions of the precursors of secondary PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia arising from outside of the UK, and to a lesser extent with primary-PM_{2.5} emissions.
- The EMEP4UK modelling shows that PM_{2.5} concentrations in Scotland are dominated by PM_{2.5} components derived from emissions outside of Scotland.
- The monitoring results for nitrate and sulphate provide evidence of an urban source of both components of PM, each showing an urban background enhancement of ~0.3 μg/m³ at the Edinburgh St Leonard's site in 2010.
- There is evidence from the analysis of the monitoring data for Glasgow that road traffic can be an important source of PM_{2.5}, with concentrations at the Glasgow Kerbside site being substantially higher than those at the nearby Glasgow Centre site.
- The diurnal pattern of PM_{2.5} and nitrogen oxides concentrations at the Glasgow Kerbside monitoring site shows that exhaust emissions are not the only source of traffic-related PM_{2.5}. It is likely that brake and tyre wear, and resuspension materials from the road surface, also make an important contribution. This is consistent with the results of the national PCM model, which



show that non-exhaust traffic emissions account for 23% of the road traffic contribution to background $PM_{2.5}$ concentrations at the Glasgow Centre site.

Grangemouth

- Analysis of the monitoring data shows that local industrial sources are not having a major influence on the annual mean PM_{2.5} concentrations at Grangemouth. The PCM modelling results, on the other hand, suggest a more significant contribution.
- The monitoring data at the Grangemouth monitoring site show a clear relationship between sulphur dioxide arising from a point source on the lneos Refinery site and $PM_{2.5}$ concentrations. This relationship suggests that industrial sources that give rise to sulphur dioxide are adding around 2% to the annual mean $PM_{2.5}$ concentrations (~0.2 µg/m³) at the monitoring site. The PCM model, on the other hand suggests that point sources in the Grangemouth area are contributing up to 3.8 µg/m³ to the annual mean $PM_{2.5}$ background concentrations, just to the east of the Ineos Refinery, and around 0.5 µg/m³ in the grid square in which the monitor is located. The PCM model also shows general industrial sources contributing a further 1.1 µg/m³ to annual mean $PM_{2.5}$ concentrations in this grid square. Thus industrial sources contribute around 20% of the modelled annual mean $PM_{2.5}$ concentrations in the grid square in which the monitor is located. Without further work, the true contribution of industrial sources to $PM_{2.5}$ concentrations at the Grangemouth monitoring site must remain unclear.
- Analysis of the monitoring data shows that emissions from the Longannet power station make a negligible contribution to sulphur dioxide concentrations at Grangemouth and hence to PM_{2.5}.
- The emissions of sulphur dioxide and nitrogen oxides from the industrial sources in the Grangemouth area will though contribute to the formation of secondary PM_{2.5} (as sulphate and nitrate). The EMEP4UK model suggests that a 30% reduction in sulphur dioxide from all sources in Scotland (67% is from power stations) would reduce PM_{2.5} by around 0.1 µg/m³ in the Grangemouth area (where the maximum impact is seen).

Health Protection Criteria

- The EU has set a Limit Value for PM_{2.5} to protect the public from exposure to excessive concentrations in hot-spots. The Limit Value concentration of 25 µg/m³ (as an annual mean, to be met by 2015) is currently being met at all monitoring sites in Scotland, even at the Glasgow Kerbside site which records the highest concentrations (23.1 µg/m³ in 2010).
- The Scottish Government has set an objective of an annual mean of 12 µg/m³ (which is to be met by 2020). This objective does not apply at kerbside locations, but does apply at the façade of residential properties alongside busy roads. It is considered likely that PM_{2.5} concentrations will exceed this objective, especially in urban areas where concentrations are already elevated, unless emissions are reduced. This objective is not in Regulations, and there are no statutory



obligations on SEPA or the local authorities to assess against this objective, or to work towards meeting it.

- The Government has recently started to use a Daily Air Quality Index to report 24-hour mean PM_{2.5} concentrations to the public to allow sensitive members of the public to take appropriate action to reduce potential health risks. The monitoring data for Scotland (2009-2011) show that for the majority of the time the 24-hour mean PM_{2.5} concentrations are in the Low Band. Nevertheless, there are occasions when 24-hour mean PM_{2.5} concentrations in urban areas, especially at the kerbside of busy roads, rise into the High and Very High Bands, which can be taken to be an indicator of unacceptable air quality.
- The EU has set a target to reduce exposure to PM_{2.5} concentrations in urban areas within each Member State. This target is to reduce PM_{2.5} concentrations, expressed as a three-year average of concentrations measured at a number of pre-defined urban monitoring locations, over the period 2010 to 2020. The target for the UK will be either a 10% or 15% reduction over this period, depending on the 2010 baseline (measured as the average concentration over the period 2009-2011), in relation to a 13 µg/m³ threshold (a 10% reduction is required if below 13 µg/m³ and a 15% reduction if above). Meeting this target will be the best way to reduce health impacts from PM_{2.5}.

Control Requirements and Opportunities

EMEP Modelling

- Reducing UK PM_{2.5} concentrations requires reductions in emissions of precursor gases arising from outside of the UK, at least as much as within the UK.
- The greatest leverage on Scotland's PM_{2.5} concentrations will arise from reductions of Scotland's primary-PM_{2.5} emissions, rather than those emissions in Scotland that contribute to secondary PM_{2.5}.
- A 30% reduction in all of Scotland's anthropogenic emissions (principally primary-PM_{2.5}, sulphur dioxide, nitrogen oxides and ammonia) is predicted to reduce PM_{2.5} concentrations by no more than 11% (~0.4 μg/m³), and in most areas by considerably less.
- Aside from two hot-spots in the mid Forth Estuary and Aberdeen, reductions in shipping emissions would have a negligible impact on PM_{2.5} concentrations in Scotland. A 30% reduction in all shipping emissions is predicted to result in a reduction of less than 2% in PM_{2.5} concentrations across Scotland.

General

• The seasonal pattern of PM_{2.5} concentrations at the Edinburgh, Grangemouth and Glasgow monitoring sites, suggests that control strategies to reduce annual average concentrations



might be most effective if they focus on reducing emissions from local sources during the winter period.

SEPA as Regulator

- The application of Best Available Techniques (BAT) is the key instrument available to SEPA to limit emissions from industrial installations.
- There are a number of drivers that allow regulators (including SEPA) to go beyond BAT:

The National Emissions Ceilings Directive (2001/81/EC) (NECD);

The Large Combustion Plant Directive (2001/80/EC);

The Industrial Emissions Directive (2010/75/EU) (IED). This will incorporate the Integrated Pollution Prevention and Control Directive (2008/1/EC) (IPPC Directive);

The Ambient Air Quality Directive (2008/50/EC);

The Habitats Directive (79/409/EEC); and

The Scottish Planning System.

• The review of these drivers has led to the following observations:

The NECD set national emission ceilings, to be achieved by 2010, which are applied at Member State level. The NECD is currently being reviewed, with the outcome due in 2013; new ceilings are expected to be established, which are likely to include PM and possibly black carbon. The NECD has helped drive down total emissions from the UK without direct reference to BAT;

The National Emissions Reduction Plan (NERP) has been developed to help implement the Large Combustion Plant Directive. For those plant that opted to be part of the NERP, it is possible that the limits applied to the emissions for a specific plant could be significantly less than the Emission Limit Value that represents BAT;

When establishing a permit for an industrial installation under the IED, the regulator is required to set the conditions which can be achieved by the use of BAT. However, the regulator may impose tighter conditions if this is necessary to meet air quality standards;

The Ambient Air Quality Directive is important, as the regulator may require the operator to take additional action if an installation is making a significant contribution to the exceedence of an air quality limit value;

The regulator must carry out an Appropriate Assessment when determining an environmental permit application that is likely to have a significant effect on a designated site under the Habitats Regulations. The requirement to take into account the findings of an Appropriate Assessment, when determining permit applications, could clearly lead to a requirement to go beyond BAT to allow the process to operate; and



The regulators have an important role as statutory or recommended consultees in the spatial and transport planning processes. Regulators have historically not actively engaged at the planning stage, but their input could be used to help drive down emissions.

 The review presented in this report shows that controls that go beyond the requirements of BAT can be, and are being, applied on the grounds of health and environmental protection. However, it would be very difficult to demonstrate that an individual installation was making a significant contribution to PM_{2.5} concentrations in urban areas (where the exposure-reduction target applies), in order to justify controls that extend beyond BAT to help meet the exposurereduction reduction targets.



2 Introduction

- 2.1 The Scottish Environment Protection Agency (SEPA) has commissioned Air Quality Consultants Ltd to provide an assessment of PM_{2.5} in Scotland. This is to follow on from the report on *PM_{2.5} in the UK* prepared for the environment agencies under the auspices of the Scotland & Northern Ireland Forum for Environmental Research (SNIFFER) (Laxen et al., 2010). The principal objective of the project is to provide SEPA with a better understanding of PM_{2.5} in Scotland such that it can determine how it can best contribute to meeting the exposure reduction targets that have been set in EU and UK legislation.
- 2.2 The project arises from a specific recommendation of the SNIFFER report, which suggested that "SEPA...should contribute to the understanding of primary and secondary emissions of PM_{2.5} from industrial, agricultural and waste sources that they regulate, as well as assist in identifying measures to limit primary and secondary emissions."
- 2.3 The project has been designed to address five tasks:

Task 1 provides information on sources of $PM_{2.5}$. It involves a review of available information and generation of new information by analysing monitoring data using OpenAir software and by running the EMEP 4UK model.

Task 2 collates information on emissions of PM and precursor gases from SEPA regulated processes.

Task 3 reviews opportunities to control emissions from processes regulated by SEPA.

Task 4 carries out a detailed examination of monitoring data for the Grangemouth industrial site to help understand the role of local sources.

Task 5 comments on the health implication of emissions from SEPA regulated sources, including an analysis of monitoring data in relation to the new Daily Air Quality Index introduced by Defra following a review by the Committee on the Medical Effects of Air Pollutants (COMEAP).

- 2.4 The report has been structured to address these five tasks although the order has been changed for presentational reasons. Where appropriate, further details are provided in the Appendices to this report.
- 2.5 The work has been carried out by Air Quality Consultants Ltd, with support from Prof. Martin Bigg of the University of the West of England, Bristol, Dr Mathew Heal, of the University of Edinburgh and Massimo Vieno of the Centre for Ecology and Hydrology.



3 Assessment Criteria for PM_{2.5}

3.1 To help understand the concentrations presented in this report it is worth setting out the criteria used to assess PM_{2.5}. These are shown in Table 1 and apply in different ways. The only mandatory criterion is the EU Limit Value of 25 μg/m³, which applies from 2015. This applies effectively at any location with public exposure, i.e. it will include roadside locations and locations close to industrial operations. The annual mean objective of 12 μg/m³ is to apply in Scotland from 2020. The objective is not set in Regulations, and there are no obligations on SEPA or the local authorities to assess against this objective or work towards meeting it. The exposure-reduction criteria (both UK and EU) apply to the average concentration measured across a network of specified urban background sites over a 3-year period. The requirement is on the UK Government to work to meet the exposure-reduction criterion. The EU average exposure indicator (AEI) obligation applies at urban background locations; there is no evidence of any risk that this obligation, which applies from 2015, will be exceeded anywhere in Scotland.

Application	Averaging Period	Criterion	To be Achieved by
UK ^a	Annual Mean	Objective of 25 µg/m ³	2020
	3-year Running Mean	Objective of 15% reduction in concentrations measured at urban background sites	Between 2010 and 2020 ^b
Scotland ^a	Annual Mean	Objective of 12 µg/m ³	2020
EU °	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 μ g/m ³	2020
	3-year Average Exposure Indicator (AEI ^d)	Target reduction in AEI (ranging from 0% to 20% depending on initial AEI ^e)	Between 2010 and 2020
	3-year Average Exposure Indicator (AEI)	Obligation of 20 µg/m ³	2015

Table 1: Criteria for PM ₂

^a these criteria were set in the 2007 update to the Air Quality Strategy for England, Scotland, Wales and Northern Ireland (Defra, 2007).

^b in practice the start value will be defined by the years 2009-2011. It is not clear yet which three years will form the end value.

^c these criteria are set in the European Union Directive on ambient air quality and cleaner air for Europe (2008/50/EC).

- ^d the AEI applies across a pre-defined set of urban background sites.
- ^e the target reduction is 0% for initial AEI of 8.5 μ g/m³; 10% for initial AEI of 8.5 to <13 μ g/m³; 15% for initial AEI of 13 to <18 μ g/m³; 20% for AEI of 18 to <22 μ g/m³ and whatever reduction is required to achieve 18 μ g/m³ for initial AEI >22 μ g/m³.



3.2 In addition to the criteria set out above, the Scottish Government has adopted a Daily Air Quality Index for $PM_{2.5}^{1.}$. This is a running 24-hour mean updated every hour, thus there can be 8760 values in a year. The thresholds for the index are set out in Table 2. To provide some context, the annual mean concentrations in Scotland fall in the Low Band, with an Index of 1 or 2, while a single 24-hour concentration of 70 µg/m³, which is in the Very High Band, with an Index of 10, would contribute 0.19 µg/m³ to the annual mean. Although the health focus for objectives and limit values is on annual mean concentrations, it is recognised that high short-term (24-hour mean) concentrations also give rise to immediate health effects, so it is relevant to have this Index in place.

Band	Index	Running 24-hour Mean (μg/m³) ^a		
Very High	10	70 or more		
	9	65 - 69		
High	8	59 - 64		
	7	53 - 58		
	6	47 - 52		
Moderate	5	42 - 46		
	4	35 -31		
	3	24 - 34		
Low	2	12 - 23		
	1	0 - 11		

Table 2: Daily Air Quality Index bandings for PM_{2.5}

^a unrounded values are rounded before comparison, so 64.49 µg/m³ is index 8 and 64.51 µg/m³ is index 9.

¹ Further details at <u>www.scottishairquality.co.uk/about.php?n_action=standards#</u>. The index also covers ozone, nitrogen dioxide, sulphur dioxide and PM₁₀.



4 **PM and Precursor Gas Emissions**

Data Sources

- 4.1 A number of data sources have been utilised to inform this section:
 - Total emissions from Scotland have been obtained from AEA, who run the National Atmospheric Inventory on behalf of Defra and the devolved administrations. These emissions data cover the period 1990 to 2009. They cover direct PM emissions and the emissions of the precursor gases, nitrogen oxides, sulphur dioxide and ammonia. In the case of PM, original data sets provided by AEA were for PM₁₀. AEA therefore also provided annual PM_{2.5}:PM₁₀ factors for each of the emission categories for each of the years, to allow a PM_{2.5} inventory to be derived in a fashion compatible with the national inventory.
 - Industrial emissions from processes permitted by SEPA have been obtained from the Scottish Pollutant Release Inventory (SPRI) website². The focus has been on 2009, being the year with most complete data, and the year for which the Scottish inventory was available. The emissions are those reported by the industrial operators in their annual returns. In the case of PM, data can be reported as PM_{total}, PM₁₀ or PM_{2.5}, the choice being up to the operator. None of the operators reported PM_{2.5} data. Some reported PM₁₀ only, some PM_{total} only, and some both. The end result is that there is no consistent set of emissions for PM. In order to derive a single data set, and as the greatest number of returns was for PM₁₀, it was decided to augment the PM₁₀ data set with the PM_{total} values. This will give an exaggerated PM₁₀ emission total, as in most cases the PM₁₀ emissions should be less than the PM_{total} emissions.

Scotland's Emissions – All Sources

4.2 Scotland's emissions of PM_{2.5}, PM₁₀, nitrogen oxides, sulphur dioxide and ammonia in 2009 from a range of source sectors are summarised in Table 3. The emissions of primary-PM_{2.5} from all sources in Scotland in 2009 are shown in Figure 1, with the trend in these emissions over the period 1990 to 2009 shown in Figure 2. There is no single source that dominates the emissions from Scotland of primary-PM_{2.5}. The three most important sources are residential combustion (23%), road transport (24%) and industry (22%), accounting for ~70% of the total. Power stations and off-road mobile sources are also important (15.6%), followed by waste and agriculture (10.1%). Shipping is not an important source nationally (3.3%), but will be more significant in a few locations, e.g. Aberdeen. Total emissions of primary-PM2.5 have declined over the period 1990-2009. Emissions of the precursor gases in Scotland are dominated by power stations in the case of sulphur dioxide (67%), power stations and road traffic

² <u>http://www.sepa.org.uk/air/process_industry_regulation/pollutant_release_inventory.aspx.</u>



in the case of nitrogen oxides (27% and 30% respectively), and agriculture in the case of ammonia (86%).

4.3 SEPA regulates the power station emissions sector, as well as sources within the industrial, waste and agricultural sectors. These sectors together account for around 40% of primary-PM_{2.5} emissions. The waste sector covers emissions from incineration, largely from small scale waste burning, while the agricultural sector is dominated by emissions from poultry operations (based on emissions for PM₁₀ in Scotland cited in the Appendices to the report by (MacCarthy et al., 2010).

Source Sector	Emissions (kt/yr)				
Source Sector	PM _{2.5}	PM ₁₀	NO _x	SO ₂	NH ₃
Residential	1.63	2.82	4.0	7.4	0.40
Commercial/institutional	0.04	0.07	1.6	0.4	<0.01
Power stations	0.54	0.89	28.0	43.6	0.09
Industrial operations	1.54	2.76	16.2	10.4	0.07
Off-road mobile	0.53	0.67	14.4	1.2	0.01
Road transport	1.67	2.27	31.1	0.1	0.92
Rail	0.03	0.07	3.5	0.1	-
Shipping	0.21	0.23	5.1	2.2	-
Aviation	0.01	0.01	0.8	0.1	-
Waste	0.38	0.38	0.1	<0.1	1.61
Agriculture	0.31	1.43	<0.1	-	30.35
Other	0.13	0.25	0.1	0.1	1.97
Total	7.02	11.84	105.0	65.5	35.42

Table 3:Emissions of PM2.5, PM10, nitrogen oxides, sulphur dioxide and ammonia from
Scotland in 2009.

4.4 Trends for precursor emissions of nitrogen oxides, sulphur dioxide and ammonia from Scotland are set out in Figure A7.1, Figure A7.2 and Figure A7.3 in Appendix A7. Total emissions of nitrogen oxides, sulphur dioxide and ammonia from Scotland have all declined over the period 1990-2009. The greatest reduction has been for sulphur dioxide, especially from power stations, although these remained the dominant source in 2009 (Figure A7.2). Nitrogen oxides emissions have also declined significantly, due mainly to the decline in emissions from power stations and road transport. The recent decline in road transport emissions of nitrogen oxides is, however, likely to be exaggerated, as the real world emissions from diesel vehicles have not declined as much as suggested by the Euro standards for new vehicles (Carslaw et al., 2011). Ammonia



emissions are dominated by the agricultural sector and they have only declined slightly over the 20-year period.



Figure 1: Apportionment of PM_{2.5} emissions from Scotland in 2009.



1990 1991 1992 1993 1994 1995 1996 1997 1998 1999 2000 2001 2002 2003 2004 2005 2006 2007 2008 2009

Figure 2: Trends in emissions of PM_{2.5} from Scotland 1990-2009 (kt/year)



Scotland's Emissions – Industrial and Power Stations

4.5 The industrial and power station emissions of primary-PM_{2.5} from Scotland for 2009 are shown in greater detail in Figure 27. Three sources dominate emissions of primary-PM_{2.5} from Scotland's industrial and power stations sector, accounting for 61% of the total: power stations, mobile and stationary combustion sources in manufacturing industries, and construction. Also important are quarrying and mining and other mineral products (19%), with the remainder (20%) being from a wide range of industrial activities.



Figure 3: Industrial sector emissions of PM_{2.5} in 2009, derived from NAEI PM₁₀ values

4.6 The Scottish Pollutant Release Inventory (SPRI) has records of PM emissions from 85 industrial sites for 2009, although two returns have not been agreed with the operator. Operators are required to report emissions over 1,000 kg/yr (1 t/yr) for PM₁₀ and PM_{2.5} and 10,000 kg/yr (10 t/yr) for total PM³. None of the operators reported PM_{2.5} emissions. Where both PM₁₀ and total PM are reported (11 plant in 2009), PM₁₀ formed between 31% and 91% of total PM. The locations and emissions of PM₁₀ from these industrial sites in 2009 are shown in Figure 4. The majority (75%) have emissions below 20 t/yr, and many of these are poultry farms. There are four sites emitting more than100 t/yr, with the maximum 459 t/yr being Longannet power station. The majority of sources are in the central belt of Scotland, continuing up the east coast to Aberdeen.

³ Scottish Pollutant Release Inventory (SPRI) Schedule to Reg. 26(2) Notice, available at: <u>http://www.sepa.org.uk/air/process_industry_regulation/pollutant_release_inventory/what_is_spri.aspx</u>





Figure 4: Point source emissions of PM₁₀ in 2009 (kg/year).



Ratios of Emissions of PM_{2.5} to Other Pollutants

4.7 PM_{2.5} emissions as a proportion of primary emissions of PM₁₀, nitrogen oxides, sulphur dioxide and ammonia are summarised by source sector in Table 4. The PM_{2.5}:PM₁₀ ratios are highly variable, although there must be considerable uncertainty as to the quality of some of the PM emissions data. The ratios to other pollutants are also highly variable between sectors. For instance, road transport is clearly a more important source of PM_{2.5} than it is of sulphur dioxide (PM_{2.5}:SO₂ ratio 22:1), while for power stations sulphur dioxide is more important (PM_{2.5}:SO₂ ratio 0.01:1). Some of these ratios are used in subsequent data analysis in Chapter 5 and 6.

Source Sector	Ratio of PM _{2.5} to				
Source Sector	PM ₁₀	NO _x	SO ₂	NH ₃	
Residential	0.578	0.405	0.219	4.107	
Commercial/institutional	0.586	0.025	0.097	481.914	
Power stations	0.607	0.019	0.012	6.200	
Industrial operations	0.558	0.095	0.148	22.945	
Off-road mobile	0.789	0.037	0.441	41.268	
Road transport	0.735	0.054	22.335	1.807	
Rail	0.380	0.007	0.234	-	
Shipping	0.947	0.042	0.099	-	
Aviation	0.760	0.008	0.117	-	
Waste	1.000	3.637	8.617	0.237	
Agriculture	0.216	-	-	0.010	
Other	0.547	1.280	2.161	0.068	
Total	0.593	0.067	0.107	0.198	

 Table 4:
 Ratios of emissions of PM_{2.5} to other pollutants in Scotland in 2009 by source sector



5 Sources and Concentrations of PM_{2.5} and its Components

Analysis of Measured Concentrations

Data Sources and Data Summary

- 5.1 A number of data sources have been utilised to inform this section:
 - Measured 1-hour mean concentrations of PM_{2.5}, at six sites operated as part of the Scottish Automatic Urban Network (SAUN) and Scottish Automatic Rural Network (SARN)⁴. These sites measure other pollutants, and data for PM₁₀, nitrogen oxides, nitrogen dioxide, sulphur dioxide, carbon monoxide and ozone have been used as appropriate;
 - Measured 1-hour mean concentrations of black carbon, at two SAUN sites, operated as part of the UK Black Carbon Network⁵;
 - Measured monthly mean concentrations of ammonium (NH₄⁺), from five sites operated as part of the national ammonia monitoring network (NAMN)⁶. The samplers used are estimated to have a cut-off of around 4.5 μm (Tang et al., 2009) and therefore fall between PM_{2.5} and PM₁₀;
 - Measured monthly mean concentrations of nitrate (NO₃⁻), sulphate (SO₄²⁻) and chloride (Cl⁻), from five sites operated as part of the national Acid Gases and Aerosols Network (AGANET)⁷. The samplers used are estimated to have a cut-off of around 4.5 μm (Tang et al., 2009) and therefore fall between PM_{2.5} and PM₁₀;
 - Meteorological data (wind speed and wind direction) specific to each SAUN and SARN site, derived from the forecasting model run by AEA and available from the air quality archive since mid-2010⁸.
- 5.2 The focus has been on data for the period 2009-2011. Some of the 2011 data are provisional, but a visual examination of the data suggests this is unlikely to have a significant impact on the outcome⁹. In the presentations of concentrations by time-of-day and month-of-the-year, results have been removed for those days when there was evidence of PM_{2.5} peaks due to Bonfire Night (noon on 5 November to noon on 8 November 2009; noon on 5 November to noon on 7 November 2010; and noon on 4 November to noon on 6 November 2011). This has been done to ensure that

⁴ <u>http://www.scottishairquality.co.uk</u>

⁵ <u>http://uk-air.defra.gov.uk/networks/network-info?view=ukbsn</u>

⁶ <u>http://pollutantdeposition.defra.gov.uk/networks</u>

⁷ <u>http://pollutantdeposition.defra.gov.uk/networks</u>

⁸ <u>http://uk-air.defra.gov.uk/data/data_selector</u>

⁹ A subsequent check carried out in April 2012 confirmed that the ratified data were essentially unchanged from those used in this report.



these episodes do not have an undue influence on the more general diurnal and seasonal patterns (further information on Bonfire Night episodes is provided in paragraph 5.15 chapter 5).

5.3 Many of the data plots have been generated using the OpenAir software package, versions 0.5-22 (Carslaw and Ropkins, 2012) using the R language versions 2.13.0 and 2.14.1 (R Development Core Team, 2012).

PM_{2.5}

- 5.4 PM_{2.5} is currently measured at six sites across Scotland:
 - three are *urban background*: Aberdeen Errol Place, located to the north of the city centre;
 Edinburgh St Leonard's, located to the southeast of the city centre; Glasgow Centre, located on the southern side of the city centre;
 - one is *rural*: Auchencorth Moss, located 10 miles to the south of Edinburgh;
 - one is *kerbside*: **Glasgow Kerbside**, located 1 m from the kerb of a busy canyon street, Hope Street, in the centre of Glasgow; and
 - one is *industrial*: **Grangemouth**, located to the southwest of an industrial area.

The locations of these sites are shown in Figure A1.1 in Appendix A1. The 1-hour mean data for these sites are presented in Figure A2.1 in Appendix A2. It is clear from these plots that the highest overall concentrations are seen at the Glasgow Kerbside site and the lowest concentrations at Auchencorth Moss. This is also evident in the annual mean data for all sites set out in Table 5.

5.5 Closer examination of the data for Auchencorth Moss reveals that caution should be applied to these results. Figure A2.2 shows the concentrations at Auchencorth Moss and Edinburgh St Leonard's for 2010 and 2011 on an expanded scale (-10 to 50 µg/m³). This makes clear that a large number of the Auchencorth Moss 1-hour mean concentrations are negative, lying between 0 and minus 4 µg/m³. The appearance is of a negative offset in the data, which suggests that the concentrations at Auchencorth Moss are too low. The reason that these data have not been rejected, is that the data ratification process allows negative concentrations down to minus 4 µg/m³. The retention of negative concentrations as potentially genuine is a feature of the FDMS instruments, and occurs when volatile material is lost from the filters. It does, however, have important implications; it is useful to estimate the urban increment by subtracting the rural (regional) background from the urban background measurements. If the rural background concentrations are, in reality, too low, then the estimated urban increment will be too high. Issues with zero off-sets in FDMS analysers have been recognised by AEA and are being investigated (Eaton and Stacey, 2011)¹⁰. These concerns about the data at Auchencorth Moss are

¹⁰ Further evidence as to the uncertainty of the Auchencorth Moss data is provided in other sections of this report.



considered to relate just to an offset, which will affect absolute concentrations, but not the relative concentrations. The analyses of the data for patterns in relation to time and to wind speed and direction are therefore still considered to be valid.

Site	Site Type	Annual Mean (μg/m³)		ıg/m³)	
		2009	2010	2011	
	PM _{2.5}				
Aberdeen Errol Place	Urban Background	((6.9)) ^a	(7.1) ^b	8.5	
Edinburgh St Leonard's	Urban Background	8.5	9.2	12.3	
Glasgow Centre	Urban Background	11.7	12.4	10.1	
Glasgow Kerbside	Kerbside	((19.7))	23.1	22.5	
Grangemouth	Industrial	8.6	11.0	10.8	
Auchencorth Moss	Rural	3.4	((4.0))	4.0	
	PM ₁₀				
Aberdeen Errol Place	Urban Background	14.8	13.3	13.7	
Edinburgh St Leonard's	Urban Background	((17.6))	14.2	15.4	
Glasgow Centre	Urban Background	(25.1)	((23.1))	16.7	
Glasgow Kerbside	Kerbside	24.3	28.6	((21.5))	
Grangemouth	Industrial	12.5	14.4	14.1	
Auchencorth Moss	Rural	((6.6))	((7.1))	(7.0)	
Black Carbon					
Edinburgh St Leonard's	Urban Background	n/a ^c	1.2	n/a	
Glasgow Centre	Urban Background	n/a	2.7	n/a	
Nitrogen Oxides					
Aberdeen Errol Place	Urban Background	44.8	((36.6))	38.2	
Edinburgh St Leonard's	Urban Background	36.6	46.6	35.1	
Glasgow Centre	Urban Background	83.8	103.5	59.7	
Glasgow Kerbside	Kerbside	269.3	308.2	242.3	
Grangemouth	Industrial	32.5	(43.8)	24.5	

Table 5:	Annual mean PM _{2.5} , PM ₁₀ , black carbon, nitrogen oxides and sulphur dioxide
	concentrations, 2009-2011



Continued

Site	Site Site Type Annual		al Mean (µ	Mean (µg/m³)	
		2009	2010	2011	
Sulphur Dioxide					
Edinburgh St Leonard's	Urban Background	3.1	4.3	3.0	
Glasgow Centre	Urban Background	3.5	3.7	2.4	
Grangemouth	Industrial	7.6	12.0	6.5	

^a values in double brackets have <75% data capture.

^b values in single brackets have between 75% and 90% data capture.

^c n/a = not available.

Black Carbon

- 5.6 Black carbon is currently measured at two sites in Scotland:
 - Edinburgh St Leonard's, located to the southeast of the city centre; and
 - Glasgow Centre, located on the southern side of the city centre.
- 5.7 Annual mean results for these sites are presented Table 5. The concentrations are only available for one year (2010) and show an annual mean concentration at the Glasgow Centre site which is 2.2 times greater than that at Edinburgh St Leonard's, which is identical to the ratio for nitrogen oxides at the two sites (see below), suggesting similar sources for both pollutants; probably road traffic, together with other combustion sources (such as domestic and commercial heating).

Nitrogen Oxides and Sulphur Dioxide

5.8 Nitrogen oxides concentrations are not measured at Auchencorth Moss, but are available for the other five sites (Table 5). The nitrogen oxides concentrations are lowest at Grangemouth, an industrial site, and highest at Glasgow Kerbside. The concentrations are significantly higher at the Glasgow Centre site than at Edinburgh St Leonard's and Aberdeen Errol Place. This indicates a more substantial road traffic component at the Glasgow Centre site, although there may also be a contribution from heating plant for the domestic and commercial premises in the area.

Nitrate, Sulphate, Ammonium and Chloride

5.9 Concentrations of nitrate, sulphate, ammonium and chloride at four rural and one urban background site are summarised in Table 6 for 2010 (more recent data are not available). The locations of the sites are shown in Figure A1.1 in Appendix A1. The Edinburgh monitor is the only urban site in Scotland in the Network, and is located alongside the PM_{2.5} monitor at the SAUN Edinburgh St Leonard's site.



5.10 Annual mean concentrations of nitrate, sulphate, ammonium and chloride over the period of measurements at the AGANET sites, 2000-2010, are shown in Figure A3.1 in Appendix A3.

Table 6:	Annual mean nitrate,	sulphate and ammonium	concentrations, 2010.
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Site	Site Type	Annual Mean Concentration (µg/m³)			
Nitrate					
Edinburgh	Urban Background	1.27			
Bush LHS	Rural	1.02			
Glensaugh	Rural	0.55			
Eskdalemuir	Rural	0.74			
Carradale	Rural	0.64			
	Sulphate				
Edinburgh	Urban Background	0.77			
Bush LHS	Rural	0.50			
Glensaugh	Rural	0.37			
Eskdalemuir	Rural	0.45			
Carradale	Rural	0.60			
Ammonium					
Edinburgh	Urban Background	0.47			
Bush LHS	Rural	0.39			
Glensaugh	Rural	0.23			
Eskdalemuir	Rural	0.32			
Carradale	Rural	0.31			
Chloride					
Edinburgh	Urban Background	1.62			
Bush LHS	Rural	1.11			
Glensaugh	Rural	0.96			
Eskdalemuir	Rural	0.90			
Carradale	Rural	1.94			

5.11 The results show the following features:

The rural chloride concentration was around 2 µg/m³ on the west coast, and around 1 µg/m³ elsewhere in 2010, with evidence of an urban enhancement of around 0.5 µg/m³ in Edinburgh. If all the chloride was present as salt (NaCl) then these values would represent ~3.3 µg/m³,



~1.6 μ g/m³ and ~0.8 μ g/m³ of salt respectively (mostly sea salt, with some contribution from winter use of de-icing salt, especially in urban areas);

- The nitrate concentrations in 2010 were higher than those of sulphate, especially at the Edinburgh St Leonard's urban site;
- There appears to be an enhancement of ~0.3 µg/m³ of both nitrate and sulphate at the Edinburgh St Leonard's site in 2010, suggesting an urban source of both nitrate and sulphate (estimated by subtracting the Bush LHS concentration from the Edinburgh St Leonard's concentration);
- The calculation of the ionic balance suggests most of the sulphate and nitrate can be accounted for by ammonium sulphate and ammonium nitrate, with the total of ammonium+sulphate+nitrate in 2010 being 2.5 µg/m³ in Edinburgh and 1.1-1.9 µg/m³ in rural areas. To turn these values into a contribution to PM, it is necessary to take into account water of hydration by multiplying by 1.29 (see paragraph A5.7 in Appendix A5). The total contribution of ammonium+sulphate+nitrate in 2010 then becomes 3.2 µg/m³ in Edinburgh and 1.4-2.5 µg/m³ in rural areas (Note: this applies approximately to the PM_{4.5} fraction);
- There was a strong decline in nitrate at the Edinburgh St Leonard's site over the five years 2006-2010, which was not apparent at the rural sites;
- Sulphate and ammonium concentrations show the strongest evidence of a decline over the ten years (2000-2010) at all sites, although there was a peak in 2003.

Analysis of Measured Concentrations

Relationship Between Pollutants

- 5.12 The correlations between the 1-hour mean measured pollutant concentrations and the modelled wind speeds at the five SAUN sites and one SARN sites in 2011, are summarised in Figure 5. The findings in relation to PM_{2.5} are:
 - A high positive correlation with PM₁₀, being strongest (r of 90 to 93%) at Auchencorth Moss, Glasgow Centre, Glasgow Kerbside and Grangemouth, but weaker at Edinburgh St Leonard's (r of 83%) and Aberdeen (r of 69%). This is not unexpected, as a large part of PM₁₀ is PM_{2.5};
 - A medium positive correlation with nitrogen oxides (r of 29% to 44%) at four sites, rising to higher correlation at Glasgow Kerbside (r of 69%). The latter is **evidence that road traffic is a source of both PM**_{2.5} and nitrogen oxides;
 - A slight negative correlation with wind speed (r of -4% to -31%), indicating greater dilution at higher wind speeds. A negative relationship with wind speed would be expected to apply to local sources, and the slight correlation suggests that local sources of PM_{2.5} are not dominant;



- A slight negative correlation with ozone (r of -7% to-31), probably driven by higher winter PM_{2.5} and lower winter ozone concentrations;
- A slight positive correlation with sulphur dioxide (r of 8% to 28%), which is not unexpected, as sulphur dioxide is an indicator of combustion sources, which will also contribute PM_{2.5}, although the slight correlation suggests that local combustion sources of PM_{2.5} are not dominant.
- 5.13 The relationship between annual mean PM_{2.5} and nitrogen oxides concentrations at the five SAUN sites in 2009-2011 is shown in Figure 6. The slope of the best fit line is 0.054, with an intercept of 7.4 µg/m³. The latter is effectively the concentration of PM_{2.5} unconnected with combustion sources. If this relationship is reflective of that of local emissions then it implies that the ratio of PM_{2.5}:NO_x in the emission is 0.054. This PM_{2.5}:NO_x ratio may be compared with that for total Scottish emissions of 0.067, and for road transport sources (including PM_{2.5} from brake and tyre wear and road abrasion, which are likely to be particularly important local contributors) of 0.054 (Table 4). Although this agreement between the measured ratio and the ratio for road traffic emissions appears to support a dominant role for road transport nitrogen oxides is likely to have been underestimated in 2009 (Carslaw et al., 2011), in which case the emissions ratio for traffic sources should be lower than that shown in Table 4.
- 5.14 Residential sources are also important (Table 3) and the PM_{2.5}:NO_x ratio in this case is 0.405 (Table 4), which is much higher than the ratio of 0.054 inferred from Figure 6. Residential emissions will, however, cover a number of different fuels, each with different PM_{2.5}:NO_x ratios. Gas is likely to dominate in the areas around the monitoring sites, and the ratio for domestic gas combustion is 0.022. Burning oil is likely to be the next most important, at 0.032. Solid fuels, although less extensively used, do have much higher ratios: (coal 1.83, wood 8.03), so it will not require many domestic solid-fuel sources to increase the ratio significantly. Indeed, one wood burning source in 250 properties, the rest using gas, would generate a ratio of 0.054, with the equivalent for coal being one source in 60 properties.





Figure 5: Correlations between measured 1-hour concentrations and modelled wind speed at five SAUN sites and one SARN site in Scotland, 2011. Numbers are r values expressed as a percentage, with the direction of the correlation shown with a + or – sign. The shapes and colours convey the same information visually.





Figure 6: Relationship between PM_{2.5} and annual mean nitrogen oxides concentrations at five SAUN sites 2009-2011 (data sets with >75% data capture). Excludes Auchencorth, for which there are no nitrogen oxides data.

Episodes

5.15 The 1-hour mean concentrations of PM_{2.5} show clear evidence of occasional high levels (see Figure A2.1 in Appendix A2). One clearly identified cause of episodes is Bonfire Night, when fireworks and bonfires contribute to PM_{2.5} emissions. Bonfire Night occurs at a time of year when there are often still, calm conditions that allow concentrations to build up. An example is presented in Figure 7 for Bonfire Night in 2011. The 1-hour mean concentrations rose to almost 300 µg/m³ at some sites and the high concentrations persisted for several hours. Indeed, the concentrations at Grangemouth persisted long enough to give a Very High concentration in terms of the Daily Air Quality Index, i.e. a running 24-hour mean concentration >70 µg/m³ (see paragraph 7.4). Bonfire Night can clearly be responsible for short-term exposure to very high concentrations of PM_{2.5}.





Figure 7: Plots of 1-hour $PM_{2.5}$ concentrations $\mu g/m^3$ at five SAUN sites and one SARN site during an episode on Bonfire Night 2011.

5.16 Episodes during the autumn and winter months are also well known in the UK, occurring when a high pressure system is dominant and stagnant atmospheric conditions prevail, with little vertical mixing and low wind speeds. These conditions can allow local emissions to build up over several days. If conditions are particularly cold then local emissions from a heating plant for example will



also be much higher. Such conditions arose during December 2010, when extreme (for the UK) cold persisted for a number of days across much of the country.

5.17 During the six day period 19-25 December temperatures were below freezing throughout the day and wind speeds were low in Glasgow, especially on 22 December (dropping to -12°C at night time). Both temperatures and wind speed increased on 26 December, bringing the episode to an end. High PM concentrations were measured at the Glasgow Centre site during this period Figure 8. The main part of the PM episode was due to PM_{2.5}, with little in the coarse PM_{2.5-10} fraction; for example, on 22 December the 1-hour mean PM_{2.5} concentrations rose to around 130 µg/m³, while the PM_{coarse} concentrations rose to just over 20 µg/m³. Also shown in Figure 8 are the nitrogen oxides and sulphur dioxide concentrations, which followed a similar pattern to the PM, although the elevation in nitrogen oxides (maximum over 1000 µg/m³) was considerably higher than that of sulphur dioxide (maximum under 30 μ g/m³). All four pollutants show a broad peak in concentrations centred on the afternoon and early evening (higher concentrations occurred between around 09:00 and 22:00h, peaking between 17:00 and 18:00h). A number of sources will have contributed to these peaks, including road traffic and domestic and commercial combustion sources. However, it is also known that this site is located in a public square, which is used at this time of year for a Christmas fair, with the stall using diesel generators, and this very local source will have contributed to the high concentrations (but to an unknown extent). The average concentration over the 7 days of the episode 19 to 25 December 2010 was 40.6 µg/m³ at Glasgow Centre. At Glasgow Centre, this episode contributed 0.8 µg/m³ to the annual mean, amounting to 6.3% of the annual mean of 12.4 µg/m³. PM_{2.5} episodes related to stagnant winter conditions, which allow a build-up of local emissions, can last several days and represent a not insignificant contribution to the annual mean concentrations.





Figure 8: PM₁₀, PM_{2.5}, PM_{2.5-10}, sulphur dioxide and nitrogen oxides concentrations over 8 days, 19-26 December 2010, at Glasgow Centre (μg m⁻³).



Variations by Time-of-Day

- 5.18 PM_{2.5} concentrations averaged by hour-of-the-day over the years 2009-2011, for all sites, except Glasgow Kerbside, are presented in Figure 9. The equivalent values for nitrogen oxides are presented in Figure 10. There are some broad similarities and some significant differences as discussed below.
- 5.19 Auchencorth Moss defines the pattern for regional background PM_{2.5} concentrations, with departures from this pattern representing local influences. The Auchencorth Moss pattern is one of a steady decrease during the morning to a minimum in the middle of the day (~11:00-12:00h), before a steady increase to a broad peak in the evening/early night (~18:00-24:00). The range though is small (~1.5 µg/m³). This diurnal pattern is likely to be a reflection of the semi-volatile nature of ammonium nitrate, which will evaporate during the day-time and condense during the night-time. If this explanation of the minor diurnal pattern is correct, then without it there may be no diurnal variation in PM_{2.5} concentrations at this rural site, which is consistent with the uniform regional concentration that would be expected for secondary PM_{2.5}, with a very limited input from local sources that give rise to a diurnal pattern, as seen at urban sites.
- 5.20 The diurnal patterns of PM_{2.5} concentrations at Edinburgh St Leonard's and Aberdeen Errol Place are reasonably similar (although the absolute concentrations are different). The range is higher than at Auchencorth Moss (~2-2.5 µg/m³) (Figure 9), however, the diurnal variation is still fairly limited in comparison with the diurnal range for nitrogen oxides (Figure 10). The PM_{2.5} concentrations are lowest during the early morning (~04:00h), rising to a broad morning peak (~07:00-11:00h), which occurs slightly earlier at Aberdeen Errol Place¹¹, followed by a further broad evening peak (~17:00-22:00).
- 5.21 The diurnal pattern of PM_{2.5} concentrations at Glasgow Centre is notably different. The minimum is still during the early morning (~04:00h), but the day-time concentrations are much higher, giving a much more significant diurnal range than at the other sites (~5 µg/m³) (Figure 9). This monitor is in the commercial heart of Glasgow and appears to be affected to a much greater extent by local emissions. These emissions are likely to be from a range of sources, including road traffic exhaust, road traffic brake and tyre wear, road traffic re-suspension of deposited materials, domestic and commercial combustion sources, including those for heating and cooking, and construction activities. A more significant role of local combustion sources at the Glasgow Centre site is also evident in the much higher nitrogen oxides concentrations (Figure 10). Notably though, the nitrogen oxides concentrations decrease during the day from a morning peak, while those of PM_{2.5} remain high. This difference in nitrogen oxides and PM_{2.5} patterns suggests a

¹¹ The earlier peak at Aberdeen Errol Place is seen more prominently in the nitrogen oxides results (Figure 10), the peak occurring around an hour earlier than at the other sites (07:00-08:00h *cf.* 08:00-09:00h). This suggests a different traffic pattern in the city, which may be related to the port activities.


disconnection between the two pollutants, and that not all of the $PM_{2.5}$ is associated with combustion sources.

5.22 The diurnal range of $PM_{2.5}$ concentrations at Grangemouth is more limited than at the other urban sites, being similar to that at Auchencorth Moss (~1.5 µg/m³), and the pattern is also somewhat different. As with the other sites, there is no clear relationship between the nitrogen oxides pattern and that for $PM_{2.5}$.



Figure 9: Plots of PM_{2.5} concentrations (µg/m³) averaged by hour-of-the-day (adjusted to local time) at five SAUN sites 2009- 2011. The shaded areas show the 95% confidence interval.





Figure 10: Plots of nitrogen oxides concentrations (μg/m³) average by hour-of-the-day (adjusted to local time) at four SAUN sites 2009- 2011. The shaded areas show the 95% confidence interval.

5.23 PM_{2.5} concentrations averaged by hour-of-the-day for Glasgow Kerbside are presented in Figure 11, together with the results for the nearby Glasgow Centre site. The equivalent values for nitrogen oxides are presented in Figure 12. The kerbside PM_{2.5} concentrations are roughly double those at the urban background site, but follow a similar pattern. The much higher PM_{2.5} concentrations at the kerbside are consistent with a major contribution from road traffic, however, this does not seem to be closely related to vehicle exhaust emissions, as nitrogen oxides concentrations at Glasgow Kerbside fall during the day (Figure 12), from a morning rush-hour peak (peaking at 08:00-09:00h), whereas the PM_{2.5} concentrations remain steady throughout the daytime (Figure 11). It is known that the numbers of heavy duty vehicles on the road, and hence exhaust emissions of nitrogen oxides, decrease during the day from a morning peak (Beevers et al., 2009). These observations suggest that road traffic <u>exhaust</u> emissions alone are not the dominant local source of PM_{2.5} at the Glasgow Kerbside monitoring site.





Figure 11: Plots of PM_{2.5} concentrations (µg/m³) averaged by hour-of-the-day (adjusted to local time) at Glasgow Centre and Glasgow Kerbside, 2009- 2011. The shaded areas show the 95% confidence interval.



Figure 12: Plots of nitrogen oxides concentrations (µg/m³) averaged by hour-of-the-day (adjusted to local time) at Glasgow Centre and Glasgow Kerbside, 2009- 2011. The shaded areas show the 95% confidence interval.



- 5.24 It is thus evident from the analysis of the data for the two sites in Glasgow that while road traffic is likely to be an important source of PM_{2.5}, vehicle exhausts are not the only contributor to this traffic source.
- 5.25 Another feature of the results in Figure 9 is the significantly higher PM_{2.5} concentrations at the Edinburgh St Leonard's and Grangemouth sites than at Auchencorth Moss, by some 5 to 6 μg/m³. This is the case even during the early morning 03:00-05:00h, when local emissions will be minimal, as seen in the nitrogen oxides patterns at these two sites Figure 10. The magnitude of this difference is puzzling. At face value the higher urban background concentrations suggest that there is a general source of PM_{2.5} making a uniform contribution throughout the day at both Edinburgh St Leonard's and Grangemouth (although Grangemouth is classified as industrial, later analysis in this report in Chapter 6, suggests a minimal contribution to PM_{2.5} concentrations from the industrial sources). It is not clear what this general source could be. On the other hand, the difference is more likely to be an artefact of the data and in particular the apparent negative offset at Auchencorth Moss, leading to artificially low concentrations at this site (see paragraph 5.5).

Seasonal Variations

- 5.26 PM_{2.5} concentrations averaged by month-of-the-year over the years 2009-2011, for all sites except Glasgow Kerbside, are presented in Figure 13. These plots show higher concentrations during the winter and lower concentrations during the summer at the three sites in Central Scotland (Edinburgh St Leonard's, Glasgow Centre and Grangemouth). This seasonal pattern is not really evident at the Aberdeen Errol Place (urban background) and Auchencorth Moss (rural) sites. The difference between the two groups of sites suggests that conditions during the winter months cause a build-up of PM in larger urban areas. This winter build-up is likely to be due to a combination of increased local emissions from combustion sources (especially for heating) coupled with weather conditions that restrict the dispersal of local emissions. This pattern will also be affected, to some extent, by the semi-volatile nature of ammonium nitrate (there will be less particulate nitrate in the summer due to greater volatilisation), and by the frequency with which air masses arrive from continental Europe (they tend to be less frequent during the summer months, which will contribute to the lower summer concentrations). The seasonal pattern of $PM_{2.5}$ in urban areas suggests that control strategies to reduce annual average concentrations might be most effective if they focus on reducing emissions from local sources during the winter period.
- 5.27 It would appear from the reduced seasonal pattern at Aberdeen Errol Place that there are fewer local sources affecting PM_{2.5} concentrations at this site.
- 5.28 The results in Figure 13 once again show that the concentrations at Auchencorth Moss are significantly lower than those at the other sites, even during the summer, when local sources appear to be playing a lesser role. For example, the $PM_{2.5}$ concentrations during August are essentially the same at the four urban sites (at ~6-7 µg/m³), while the concentration at Auchencorth



Moss is around 4-5 μ g/m³ lower (at ~2 μ g/m³). This provides further evidence to question the quality of the data from the FDMS monitor at Auchencorth Moss (see paragraph 5.5 for further discussion on this).



Figure 13: Plots of PM_{2.5} concentrations (µg/m³) averaged by month-of-the-year (adjusted to local time) at four SAUN sites and one SARN site, 2009- 2011. The shaded areas show the 95% confidence interval.

Relationship with Wind Speed and Direction

- 5.29 The 1-hour mean PM_{2.5} concentrations have been analysed in relation to wind speed and wind direction using OpenAir to produce bivariate polar plots. The results for 2011 are shown in Figure 14. In these plots the annual means are broadly represented by the green colours, with the blues representing concentrations below the mean, and the yellows and reds above the mean. There are some broad similarities and some significant differences as discussed below.
- 5.30 The common feature of all the plots is that the higher concentrations are associated with winds in the southeast sector and mostly at higher speeds of 10-15 m/s. This common feature will represent the influx of secondary PM_{2.5} being transported from precursor sources elsewhere in the UK and Europe (see next section on trajectory analysis for more details of this transport).
- 5.31 One important difference between the sites is that the Glasgow Kerbside site has much higher concentrations, both overall and with winds from the east and southeast. The local sources



affecting this kerbside site are evident in the higher concentrations with lower wind speeds. These high concentrations are still most clearly associated with winds from the east and southeast, but they are probably a reflection of recirculation in the street canyon. The monitor is on the east side of a road that runs north/south, and recirculation in the canyon will bring the road traffic emission to the monitor with easterly winds. There is also an east-west tunnel 40 m to the south of the monitoring site, which runs for 100 m under the Central Station, that might be having some influence.

- 5.32 Another interesting feature is the generally lower PM_{2.5} concentrations at Auchencorth Moss. There is no evidence of a significant contribution from the Edinburgh urban area to the north, but more notably even when there are strong winds from the southwest, which will generally bring clean air to all sites, the concentrations are lower at Auchencorth Moss. This is likely to relate to the issues of data quality from the FDMS monitor at Auchencorth Moss, as discussed in paragraphs 5.5.
- 5.33 The polar plot for Aberdeen Errol Place show particularly high concentrations associated with winds from the southeast through to south that have a high wind speed (~15 m/s). The most likely explanation for these high concentrations is that they show a contribution from shipping using Aberdeen docks, located 1.4-2.2 km upwind in this direction. Higher winds are known to bring plumes down to the ground closer to the source, and the source is likely to be the ships engines which discharge some 20-30 m above the ground. Winds from this direction at higher speeds will be relatively infrequent, so this source will not make a large contribution to the annual mean concentrations at this site (southerly winds blowing from the docks towards the Errol Place monitoring site will be reasonably frequent, but not at the higher speeds relevant to this analysis; see the wind rose at http://www.metoffice.gov.uk/climate/uk/es/print.html.





Figure 14: Polar plots of $PM_{2.5}$ concentrations at the five SAUN sites and one SARN site in 2011, as a function of wind speed and wind direction. The concentrations are shown on the colour scale in $\mu g/m^3$. The wind speeds increase from 0 m/s at the centre to 20 m/s in the outer circle.



5.34 It is also of note that the patterns for Edinburgh St Leonard's and Grangemouth are similar, but with somewhat higher concentrations at low wind speeds in Edinburgh, which probably relate to the closer proximity of the site to traffic. Further analysis of PM_{2.5} in Grangemouth is provided in Chapter 6.

Relationship with Wind Direction and Time-of-Day

- 5.35 The 1-hour mean PM_{2.5} concentrations have also been analysed in relation to wind direction and time-of-day using OpenAir to produce polar annulus plots. The results for 2011 are shown in Figure 15 for all sites other than Glasgow Kerbside, with the annual mean concentrations being broadly represented by the light yellow colours, with the blues and greens below the mean, and oranges and reds above the mean (Note the smaller concentration range as compared with Figure 14, 0-20 µg/m³ *cf.* 0-35 µg/m³).
- 5.36 The plots show generally higher concentration with winds from the southeast sector, with a tendency for the highest concentrations during the night-time. These are features identified previously at sites across the UK (Laxen et al., 2010). There are, however, some interesting differences between the sites. The highest concentrations at Aberdeen with south-southeasterly winds are during the early morning 00:00-09:00h, which differs from the pattern at Auchencorth Moss and Edinburgh St Leonard's, where the highest concentrations are in the afternoon/evening (12:00-23:00h). This may relate to port activities, which lie in this direction, affecting PM_{2.5} concentrations at the Aberdeen site.
- 5.37 The Edinburgh St Leonard's site has some high PM_{2.5} concentrations at night-time (1900:24:00h) with winds from the northeast, not seen at the other sites. It is not clear what this night-time source might be, but it is spread over a fairly wide wind sector 20-70°, suggesting it is not a point source. There is some evidence of higher concentrations with winds from the northeast in the polar plots (Figure 14), with wind speeds from 0-6 m/s.
- 5.38 Apart from the features noted above the plots show little variation by time-of-day, which suggests local sources are not dominant.
- 5.39 The Glasgow Kerbside site is not included in Figure 15, as it would be off the scale. The results for Glasgow Kerbside and the nearby Glasgow Centre site are shown in Figure 16. The pattern at the kerbside site is consistent with the analysis of the polar plots (paragraph 5.31), which attributed the easterly bias to recirculation in the street canyon. There is also some evidence of the importance of the rush hours, when concentrations are their highest. The pattern is different to that at Glasgow Centre, where there is little definition of concentrations by time-of-day. **Road traffic is clearly an important source of PM_{2.5} in close proximity to the carriageway.**





Figure 15: Polar annulus plots of $PM_{2.5}$ concentrations at the five background and one industrial SAUN site in 2011, as a function of wind direction and time-of-day. The concentrations are shown on the colour scale in $\mu g/m^3$. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00 h.





Figure 16: Polar annulus plots of $PM_{2.5}$ concentrations at the Glasgow Centre background and Glasgow Kerbside SAUN sites in 2011, as a function of wind direction and time-of-day. The concentrations are shown on the colour scale in $\mu g/m^3$. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00 h.

Relationship with Air Mass Trajectory

- 5.40 The 1-hour mean PM_{2.5} concentrations measured at three of the monitoring sites over the period 2009-2011 have been analysed in relation to the 96-hour air mass back trajectories¹². These trajectories show the path that the air has followed for the 96 hours prior to arriving at the monitoring site. The approach has been to assign the concentration at the monitoring site at the arrival time of the trajectory to the whole of the back trajectory. Patterns can then appear related to the areas these back trajectories have passed over, to suggest those areas where emissions leading to the higher concentrations may have arisen. Local sources have the potential to affect the outcome; however, the back trajectories follow a wide range of paths, so a local source from a confined area, e.g. a point source, will have little impact on trajectory concentrations. The influence of a local source can also be identified/eliminated by looking at the results for several sites. If the patterns are very similar then the local sources will not be important, and the patterns will provide a reasonable representation of more distant sources. Furthermore, as the results for a rural site should not be strongly influenced by local sources, they will give a good indication of distant sources.
- 5.41 The trajectories used in this analysis are available for Edinburgh Airport. It is reasonable to apply these trajectories to the nearby monitoring sites of Grangemouth, Edinburgh St Leonard's and Auchencorth Moss. Figure 17 shows the individual trajectories for the site at Auchencorth Moss over the three years 2009-2011. The trajectories are plotted sequentially on top of each other, so

¹² These are 96-hour back trajectories calculated using the HYSPLIT trajectory model for every three hours derived from NCEP/NCAR Reanalysis data that are available for locations across the UK as part of the OpenAir package – further details at <u>http://www.openair-project.org/PDF/OpenAir_NewsLetter_Issue10.pdf</u>



earlier trajectories with higher concentrations will be hidden under subsequent trajectories. To overcome this and allow patterns to be identified, the trajectories are used to derive gridded mean concentrations, based on concentrations associated with all the trajectories passing through a grid square. These have then been smoothed to provide a better visual representation.

- 5.42 Smoothed trajectory plots for the full three year period 2009-2011 are shown for the three sites Auchencorth Moss (Figure 18), Edinburgh St Leonard's (Figure 19) and Grangemouth (Figure 20). When looking at these plots it is important to recognise that the colour concentration scales are different, thus Figure 19 looks very different to Figure 20, however the greens in Figure 19 represent PM_{2.5} concentrations around 13-16 µg/m³, while in Figure 20 they represent 8-12 µg/m³ (in Figure 17 they represent 7-9 µg/m³) All three trajectory plots show the same broad features, with higher PM2.5 concentrations in the Edinburgh region being associated with air brought in from northern and eastern Europe, most notably the Netherlands, Belgium, Germany and Poland (little attention should be paid to the higher concentrations in the Balkans, as few trajectories had passed over this region). These source regions are also identified as having a major influence on UK concentrations in the EMEP modelling discussed in the next section of this chapter (paragraphs 5.51 to 5.59). The lowest PM_{2.5} concentrations are associated with air from the Atlantic. There is evidence of some elevation in PM25 concentrations for air that has passed over England. These higher concentrations associated with air from Europe and England will represent secondary PM_{2.5} formed from precursor emissions from these **regions** (there may also be a small component of primary $PM_{2.5}$).
- 5.43 Previous analyses have raised questions about the quality of the FDMS results at Auchencorth Moss (paragraph 5.5). Further insight comes from a comparison of Figure 18 and Figure 19. Air arriving at Auchencorth Moss that has passed over Germany has a concentration of around 13 μg/m³, but when it arrives at Edinburgh St Leonard's or Grangemouth from the same region the concentration is around 6 μg/m³ higher at around 19 μg/m³. Similarly when air arrives from central France, the concentrations at Edinburgh St Leonard's or Grangemouth are around 7 μg/m³ higher (7 μg/m³ *cf.* 14 μg/m³) than at Auchencorth Moss. While there will be some general urban enhancement at both Edinburgh St Leonard's and Grangemouth, it would not be expected to be this great. This enhancement provides further evidence to call into question the quality of the FDMS monitoring results for Auchencorth Moss.
- 5.44 It is important to recognise that these trajectory plots show the areas that are contributing to the higher concentrations. The extent to which these areas influence the annual mean concentrations will depend on the frequency of winds from these directions. The predominant winds are from the southwest, bringing cleaner Atlantic air.





Figure 17: Trajectory analysis using Auchencorth Moss PM_{2.5} concentrations (µg/m³), 2009-2011. Note PM_{2.5} concentration scale (approx. 0-70 µg/m³)



Figure 18: Trajectory analysis using Auchencorth Moss PM_{2.5} concentrations (µg/m³), 2009-2011. Note PM_{2.5} concentration scale (approx. 0-20 µg/m³)





Figure 19: Trajectory analysis using Edinburgh PM_{2.5} concentrations (μg/m³), 2009-2011. Note PM_{2.5} concentration scale (approx. 0-30 μg/m³).



Figure 20: Trajectory analysis using Grangemouth PM_{2.5} concentrations (μg/m³), 2009-2011. Note PM_{2.5} concentration scale (approx. 0-30 μg/m³).



Analysis of Modelled Concentrations

National Modelling

- 5.45 AEA is responsible for producing maps of PM_{2.5} background concentrations across the UK, for which it uses its semi-empirical Pollution Climate Model (PCM) (Brookes et al., 2011). These maps cover a 1x1 km grid and are built up from contributions from a range of sources to each grid square. The mapped concentrations across Scotland for 2011 are shown in Figure 21. Annual mean background PM_{2.5} concentrations across Scotland range from 3 to 11 µg/m³, with the highest values in the urban areas, especially Glasgow and Edinburgh. There is also a general pattern of higher concentrations across the central belt of Scotland, and up the east coast to Aberdeen (shown in detail in Figure 22).
- 5.46 The 1x1 km background concentrations of PM_{2.5} have been apportioned into 16 components at four of the SAUN monitoring sites, with the results for 2010 made available to this study by AEA. The results for Aberdeen are shown in Figure 23, Edinburgh in Figure 24, Grangemouth in Figure 25, and Glasgow in Figure 26. When considering these results it should be borne in mind that they refer to the 1x1 km grid square in which the monitor is located, and other parts of these urban areas may have slightly different compositions. The modelled and measured total PM_{2.5} concentrations are summarised in Table 7. The comparison of the modelled concentrations in 2010 is best made against the measured concentrations across all three years, as the model does not incorporate meteorology, other than that it is based on a calibration against measurements made in 2008. It should also be borne in mind that the measurement data may be affected, to some extent, by nearby sources, and thus not fully represent concentrations across the 1x1 km grid square. Given these limitations there is reasonable agreement between the modelled and measured concentrations, other than that the model appears to over-predict concentrations at the Aberdeen site (it is possible that the measured concentrations at Aberdeen may be suffering from a negative offset, as appears to be the case for Auchencorth Moss – see paragraph 5.5 – but this possibility is not explored further.

Sito	Modelled PM _{2.5} (µg/m³)	Measured PM _{2.5} (µg/m ³)			
one		2009	2010	2011	
Aberdeen Errol Place	11.1	((6.9)) ^a	(7.1) ^b	8.5	
Edinburgh St Leonard's	11.2	8.5	9.2	12.3	
Glasgow Centre	13.6	11.7	12.4	10.1	
Grangemouth	10.8	8.6	11.0	10.8	

Table 7: Annual mean modelled (2010) and measured (2009-2011) PM_{2.5} concentrations.

^a values in double brackets have <75% data capture.

^b values in single brackets have between 75% and 90% data capture.





Figure 21: Modelled 1x1 km background concentrations of $PM_{2.5}$ in Scotland in 2011 $\mu g/m^3$.





Figure 22: Modelled 1x1 km background concentrations of annual mean $PM_{2.5}$ in Central Scotland in 2011 μ g/m³.

- 5.47 The source apportionment of background concentrations at the four monitoring sites shows:
 - Secondary PM_{2.5} is dominant at all four sites, accounting for around 30-40% of the total;



- **Road traffic**, which includes vehicle exhaust, brake and tyre wear and road abrasion, is an important contributor in all areas, but especially in Glasgow (27%) and less so in Aberdeen (10%);
- **Industry** is an important contributor in some areas, being most significant in Aberdeen (16%) and less so in Edinburgh (3%). Industry accounts for 10% of the PM_{2.5} at the Grangemouth industrial site;
- **Off-road mobile machinery** can also be important, ranging from 5% in Aberdeen to 14% in Grangemouth;
- **Dust, both urban and rural** accounts for some 7-10%. This is significant, as these dusts are likely to be difficult to control;
- Long-range transport of primary-PM_{2.5} (which covers sources more than 10 km away from the grid square) makes a minimal contribution at all sites.



Figure 23: Aberdeen Errol Place: Apportionment of background PM_{2.5} concentrations at the monitoring site in 2010.





Figure 24: Edinburgh St Leonard's: Apportionment of background PM_{2.5} concentrations at the monitoring site in 2010.



Figure 25: Grangemouth: Apportionment of background PM_{2.5} concentrations at the monitoring site in 2010.





Figure 26: Glasgow Centre: Apportionment of background PM_{2.5} concentrations at the monitoring site in 2010.

- 5.48 The national maps of PM_{2.5} concentrations also provide information on the spatial distribution of industrial point sources contributions (these will be from Part A processes) (Figure 27). There is a large area in the central belt of Scotland, between Glasgow and Edinburgh, where point sources contribute 0.1-0.3 μg/m³ to PM_{2.5} concentrations, rising to 3-4 μg/m³ in two 1x1 km grid squares over the Grangemouth industrial complex. The concentrations in the central belt are shown in more detail in the expanded map in Figure A4.1 in Appendix A4.
- 5.49 The spatial distribution of PM_{2.5} concentrations arising from more general industrial sources (these will be from Part B processes) is shown in Figure 28. The contributions are generally smaller than those of point sources and more dispersed. They also affect smaller areas but are still most prevalent in the central belt of Scotland. The concentrations in the central belt are shown in more detail in the expanded map in Figure A4.2 in Appendix A4.





Figure 27: Modelled contribution of point source emissions to annual mean background concentrations of $PM_{2.5}$ in Scotland, 2011 (μ g/m³).





Figure 28: Modelled contribution of industrial (in-square, non-point source) emissions to annual mean background concentrations of PM_{2.5} in Scotland, 2011 (μg/m³).

EMEP Modelling

5.50 Chemical-transport models simulate the transport and chemical reactions of air pollutants in the atmosphere from source to receptor, with realistic time resolution, and so can help quantify the extent to which the PM_{2.5} concentration at a location is composed of species emitted more locally or from further afield. Results from the European Monitoring and Evaluation Programme Unified



Model (EMEP UM) and EMEP4UK models have been used here. A detailed description of the models and their limitations is provided in Appendix A5.

Sensitivity of UK PM_{2.5} to UK and Non-UK Emissions

- 5.51 The main EMEP Unified Model (at 50 km horizontal spatial resolution) has been used to provide source-receptor matrices at the level of individual country within the EMEP domain. See http://www.emep.int/SR_data/index_sr.html.
- 5.52 The methodology for these PM source-receptor matrices is to reduce by 15% the anthropogenic emissions of certain primary components in each country and to calculate the resultant reduction in PM concentrations in that country and in every other country. The source-receptor information is available for 15% reductions in emissions in each of primary-PM_{2.5}, primary-PM₁₀, sulphur oxides, nitrogen oxides, ammonia and volatile organic compounds (VOC). Given that current public versions of EMEP PM models do not include modelling of secondary organic aerosol (see Appendix A5), the impact of reductions in VOC emissions on PM_{2.5} (and PM₁₀) is much smaller than the impact in reductions of primary-PM_{2.5} and PM₁₀, sulphur oxides, nitrogen oxides and ammonia. (The impact of VOC emissions on inorganic components of particles arises indirectly from their effect, through OH radical and ozone, on the tropospheric oxidation rates of the inorganic components.)
- 5.53 The source-receptor data are only available at the level of the sovereign state, e.g. the UK. It is therefore not possible to separate the impact of emissions from sources within and outwith Scotland on PM_{2.5} concentrations in Scotland specifically. However, useful insight on the transnational impact of precursor emissions on PM_{2.5} is gained from analysis at the UK level.
- 5.54 Using the data available at <u>http://www.emep.int/SR_data/index_sr.html</u>, the reduction in UK average surface PM_{2.5} concentrations arising from 15% reductions in emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia, individually, from the UK and from other geographic areas has been calculated for this report. The results for 2010 (annual averages) are given in Table 8. The simultaneous reduction in the final 'All' columns includes a 15% reduction in anthropogenic VOC emissions, but as indicated above VOC emissions have much smaller impact on the modelled PM_{2.5} components than the other precursor emissions. Figure 29 shows the percentage data in Table 8 as pie-charts.



Table 8:Reductions in 2010 UK-average surface PM2.5 resulting from 15% reductions in
emissions of each of primary-PM2.5, sulphur oxides, nitrogen oxides and
ammonia individually, and in all emissions simultaneously, from each of the
countries or regions indicated. Source of raw data: http://www.emep.int.

	Anthropogenic Precursor with Reduced Emission									
Source Region	Primary-PM _{2.5}		SOx		NOx		NH ₃		All ^a	
	μg/m³	%	μg/m³	%	μg/m³	%	μg/m³	%	μg/m³	%
UK	0.072	38.3	0.052	17.0	0.093	15.7	0.14	26.6	0.365	21.8
Ireland	0.013	6.9	0.019	6.2	0.084	14.2	0.059	11.2	0.174	10.4
Atlantic & North Sea	0.027	14.4	0.03	9.8	0.034	5.8	0.073	13.9	0.168	10.0
France & Benelux	0.037	19.7	0.091	29.8	0.197	33.3	0.134	25.5	0.498	29.7
Scandinavia inc. Baltic Sea	0.016	8.5	0.036	11.8	0.045	7.6	0.046	8.7	0.144	8.6
Germany & Central & East Europe	0.018	9.6	0.052	17.0	0.103	17.4	0.061	11.6	0.242	14.4
Southern Europe	0.002	1.1	0.012	3.9	0.011	1.9	0.004	0.8	0.03	1.8
Regions further afield	0.003	1.6	0.013	4.3	0.024	4.1	0.009	1.7	0.054	3.2
Totals	0.188	100	0.305	100	0.591	100	0.526	100	1.675	100

The simultaneous reduction scenario also includes 15% reductions in anthropogenic VOC but this precursor has much smaller impact on the modelled PM_{2.5} components than the other precursor emissions.









All

22%

10%

10%

2% 3%

14%

Total UK PM2.5 reduction 1.68 µg/m3

9%









∎UK

Ireland

Atlantic & North Sea

- France & Benelux
- Scandinavia inc. Baltic Sea
- Germany & Central & East Europe
- □ Southern Europe
- □Regions further afield
- Figure 29: Reductions in 2010 UK average surface PM_{2.5} resulting from 15% reductions in emissions of each of primary- PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia individually, and in all emissions simultaneously, from each of the countries or regions indicated. Source of raw data: <u>http://www.emep.int</u>.



- 5.55 The following observations can be made on the data in Table 8 and Figure 29 (the term 'average PM_{2.5} concentration' refers to the 'annual average surface PM_{2.5} concentration'):
 - The UK average PM_{2.5} concentration is reduced more by reductions in nitrogen oxides or ammonia emissions across the whole EMEP domain (UK PM_{2.5} reductions of 0.59 μg/m³ or 0.53 μg/m³, for 15% reductions in nitrogen oxides or ammonia respectively) than by reductions in sulphur oxides or primary-PM_{2.5} emissions across this domain (UK PM_{2.5} reductions of 0.31 μg/m³ or 0.19 μg/m³, for 15% reductions in sulphur oxides and primary-PM_{2.5}).
 - The impact on the UK average PM_{2.5} concentration of 15% reductions in all emissions simultaneously (1.68 µg/m³) is very close to the sum of the impacts on UK PM_{2.5} from 15% reductions in emissions of each of the four components separately (1.61 µg/m³). The former scenario also includes 15% reductions in anthropogenic VOC emissions. When VOC emissions are reduced, the reduction in the UK average PM_{2.5} concentration is 0.06 µg/m³ (data not shown) so adding this to the sum of the impact on the UK average PM_{2.5} concentration of separate emissions reductions gives 1.67 µg/m³. The agreement of these total values shows that, at least for these relatively small reductions in precursor emissions (15%), the impact of reductions in different emission components is essentially linear and additive.
 - In all cases, the impact on the UK average PM_{2.5} concentration of reductions in the UK (and Ireland) emissions is only minor in comparison with the impact of equivalent magnitudes of reductions of emissions elsewhere in the EMEP domain. This is particularly the case for sulphur oxides and nitrogen oxides emissions. The data indicate that UK emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia contribute only 38%, 17%, 16% and 26%, respectively, to the total impact on the UK average PM_{2.5} concentration from reductions in emissions of each of these components across the whole EMEP domain. The percentages increase to 45%, 23%, 30% and 38%, respectively, for consideration of the sum of UK and Ireland emissions, as compared with UK emissions alone. These percentages contrast with those presented in Figures 28 & 29 of the report on PM_{2.5} in the UK, prepared for SNIFFER (Laxen et al., 2010), which had been taken directly from an earlier AQEG report (AQEG, 2005); this indicated that the majority of sulphate and nitrate in the UK was derived from UK emissions. This former modelling work dates from at least 10 years ago, which suggests UK sulphate and nitrate concentrations have become more sensitive to the emissions from outside the UK during a period when UK emissions of sulphur oxides and nitrogen oxides have declined.
 - Emissions of nitrogen oxides, sulphur oxides and ammonia from France and the Benelux countries have a large impact on the UK average PM_{2.5} concentration, and contribute more than emissions of these gases from UK sources. Emissions of nitrogen oxides, sulphur oxides and ammonia from France and Benelux contribute 30%, 33% and 26% respectively, to the



overall sensitivity of the UK average $PM_{2.5}$ concentration to a 15% change in emissions. This will probably apply mostly to $PM_{2.5}$ concentrations in the more southern areas of the UK.

- Emissions of primary-PM_{2.5} and sulphur oxides from sources in the North Sea and Atlantic regions (shipping) make a relatively important contribution to the UK average PM_{2.5} concentration sensitivity to emissions reductions. This also appears to be true for ammonia emissions from these areas; there may be issues with appropriate assignment of land and marine sources for a model grid of 50 km resolution.
- For sulphur oxides and nitrogen oxides, in particular, there is clear evidence that emissions from far afield are important for the UK average PM_{2.5} concentration. The geographic region labelled Germany and Central & East Europe extends to the Baltic and Balkan states; sulphur oxides and nitrogen oxides emissions in this region contribute 17% each to the overall sensitivity of the UK average PM_{2.5} concentration to a 15% change in emissions. The region labelled Southern Europe includes Spain, Portugal, Italy and Greece. The regions further afield category includes Belarus, Russia, Ukraine, Turkey. Together these latter two regions still contribute 8% and 6% of the sensitivity of the UK average PM_{2.5} concentration to sulphur oxides and nitrogen oxides emissions reductions.
- 5.56 Overall, these observations indicate that concentrations of UK PM_{2.5} are significantly associated with emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia outside of the UK. Consequently reducing UK PM_{2.5} requires reductions in emissions of precursor gases outside of the UK at least as much as within the UK.
- 5.57 These model sensitivities are for 15% reductions in emissions. It is likely that the reduction in the PM_{2.5} components derived from these emissions will be reasonably linear with the emissions reductions, at least for further modest reductions; for example, it may be assumed that a 23% reduction in emissions will lead to around a 1.5 times reduction in the UK average PM_{2.5} concentration as compared with that simulated for 15% reduction in emissions. Linearity will certainly be the case for reductions in primary-PM_{2.5} emissions which are essentially chemically inert and therefore subject only to transport and deposition processes (although if the size distribution of primary-PM_{2.5} emissions were to change the geographical range of influence of primary-PM_{2.5} would also change.)
- 5.58 However, some non-linearities between precursor inorganic gas emissions and particle components are recognised, e.g. for sulphate (Jones and Harrison, 2011), such that reductions in sulphur dioxide emissions are associated with smaller reductions in particle sulphate.
- 5.59 The model data discussed above are for the average PM_{2.5} concentrations across the UK as a whole, and not specifically for Scotland. Since Scotland is situated further from the continental source regions it is anticipated that PM_{2.5} concentrations in Scotland will be influenced by emissions from Scotland and the rest of the UK to a greater extent, than is the case for the UK-



average picture. On the other hand, Scotland is a relatively small source region for these inorganic precursor gases, so primary-PM_{2.5} and secondary inorganic $PM_{2.5}$ concentrations in Scotland are still comprised to a significant extent of emissions from further afield. This is explored in more detail by the Scotland specific model simulations carried out for this report, as described in the next section.

5.60 The results from the EMEP modelling can be compared with those reported in a recent modelling study of the contribution of combustion emissions to UK PM_{2.5} exposure (Yim and Barrett, 2012). This study used the Community Multiscale Air Quality (CMAQ) modelling system to predict PM_{2.5} concentrations (both primary and secondary) across the UK arising from combustion sources in the UK and the wider European domain. The results are summarised in Table 9. There are clearly questions about the absolute values, as the total population-weighted annual mean PM_{2.5} concentration in 2007 was around a third of the value for the UK in 2010 (Laxen et al., 2010). Some of the discrepancy will be due to the CMAQ modelling not including sea salt, mineral particles and natural organic particles, but this will not be the whole reason. The relative values should though be reasonably reliable for combustion sources of PM_{2.5} (within the uncertainties of the modelling) and show road transport is a dominant source of exposure to combustion PM_{2.5}, while industry makes a limited contribution.

	Contribution			
Combustion source ^a	Concentration (µg/m³)	% of total combustion		
UK Road transport	0.745	24		
UK Other transport ^a	0.417	14		
UK Power generation	0.403	13		
UK Other	0.259	8		
UK Industry	0.132	4		
All UK	2.061 ^c	67		
All Non-UK EU combustion	0.999	33		
Total combustion	3.060	100		

Table 9:	Modelled UK population-weig	nted annual mean PM _{2.5} concentrations in 2007.
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^a The contribution includes primary and secondary contributions from these sources.

^c This component is mainly shipping and aviation.

^c This is the value provided in the paper, it is not the sum of the sources listed above. It is not clear what the missing component is.

Sensitivity of PM_{2.5} in Scotland to Primary and Precursor Emissions in Scotland

5.61 The EMEP4UK model was adapted so that emissions from the 5 km x 5 km grid squares overlying the Scotland land surface could be varied, while leaving emissions unaltered from the rest of the



British Isles inner domain and the rest of the full EMEP domain. The exact area designated as Scotland is shown in Figure A5.2 in Appendix A5. Grid squares extending into maritime regions were included to ensure that emissions directly on the coast were not excluded from the model simulations, but in the simulations described here, only land-based emissions were altered.

- 5.62 The sensitivity of Scotland's PM_{2.5} concentrations to 30% reductions in emissions from all Scottish sources of primary-PM_{2.5}, sulphur oxides, nitrogen oxides, and ammonia have been simulated, both in separate model runs, and in a model run where the Scottish emissions of all these species were reduced by 30% simultaneously. The latter run also included 30% reductions in Scottish emissions of anthropogenic VOCs and carbon monoxide but, as discussed above, these precursors have a considerably smaller impact on the modelled PM_{2.5} concentrations than the other emissions.
- 5.63 Model simulations were undertaken for 2008. All concentrations are annual averages for the surface model layer.
- 5.64 Changes in $PM_{2.5}$ are expressed relative to the baseline in which all emissions are at 100%, thus positive concentrations in the "Delta (Δ)" maps of concentration change indicate that the specified emissions reduction is associated with *reductions* in the concentrations of the specified PM_{2.5} component.
- 5.65 The combination of several emissions reductions simulations and several modelled PM_{2.5} components yields a lot of output data. The maximum effect of Scotland's emissions reductions on PM_{2.5} concentrations is illustrated by the simulation with 30% reductions in all of Scotland's anthropogenic emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously.
- 5.66 The pairs of baseline and "Delta (Δ)" maps for total PM_{2.5}, and for the separate components of primary-PM_{2.5}, sulphate, nitrate and ammonium, derived from the model sensitivity run in which all of the Scotland anthropogenic emissions are reduced by 30% simultaneously are shown in Figure 30 to Figure 34. It should be remembered that modelled PM_{2.5} comprises primary-PM_{2.5} + sea-salt(fine) + sulphate + nitrate(fine) + ammonia, which does not cover all the components of PM_{2.5} (e.g. secondary organic PM is not included) see the detailed explanation in Appendix A5.







Figure 30: Impact on modelled $PM_{2.5}$ (µg/m³) from 30% reduction in Scotland's anthropogenic emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously. Left panel is baseline PM_{2.5} (100% emissions), right panel is the reduction (Δ) in PM_{2.5}.







Figure 31: Impact on modelled primary-PM_{2.5} component (μ g/m³) from 30% reduction in Scotland's anthropogenic emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously. Left panel is baseline primary-PM_{2.5} (100% emissions), right panel is the reduction (Δ) in primary-PM_{2.5}.







Figure 32: Impact on modelled sulphate component (μ gS/m³) from 30% reduction in Scotland's anthropogenic emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously. Left panel is baseline sulphate (100% emissions), right panel is the reduction (Δ) in sulphate. Concentrations as μ g/m³ sulphate will be 3.0 times higher.







Figure 33: Impact on modelled nitrate (NO₃⁻) component (μ gN/m³) from 30% reduction in Scotland's anthropogenic emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously. Left panel is baseline nitrate (100% emissions), right panel is the reduction (Δ) in nitrate. Concentrations as μ g/m³ nitrate will be 4.43 times higher.









5.67 The "Delta (Δ)" maps of concentration change in Figure 30 to Figure 34 are shown expressed as a percentage change relative to their respective baseline maps in Figure 35. Sets of analogous maps for the separate sensitivity runs with 30% reductions in Scotland emissions of each of sulphur oxides, nitrogen oxides and ammonia individually are given in Appendix A6.











- Figure 35: Percentage reductions in modelled $PM_{2.5}$, primary- $PM_{2.5}$, sulphate, nitrate and ammonium from 30% reduction in Scotland's anthropogenic emissions of primary- $PM_{2.5}$, sulphur oxides, nitrogen oxides and ammonia (and also of VOC and CO) simultaneously, i.e. these are the Delta (Δ) maps of Figure 30 to Figure 34 expressed as a % of their respective baseline maps. Red colours indicate reductions in $PM_{2.5}$
- 5.68 It should be noted that reductions in emissions of individual gases will impact on other components of PM_{2.5} as well as on the main components illustrated in Figure A6.1 to Figure A6.3 because of



changes in the rates of formation of inorganic salts from the gas species. For example reductions in ammonia emissions impact on particulate sulphate and particulate nitrate as well as on particulate ammonium. More subtly, reductions in nitrogen oxides emissions affect sulphate formation because the ammonia that would have reacted with nitrogen oxides emissions to form particulate nitrate becomes available for reaction with sulphur oxides emissions to form particulate sulphate. Similarly, reductions in sulphur oxides indirectly affect particulate nitrate. However, these changes are smaller than the direct impacts of sulphur oxides reductions on sulphate, nitrogen oxides reductions on nitrate, and ammonia reductions on ammonium, and are all taken into account in the maps in Figure A6.1 to Figure A6.3 of impact of reductions of the individual precursor gases on total PM_{2.5}.

5.69 Examination of Figure 30 to Figure 35 yields the following observations.

- As illustrated by each of the maps of baseline PM_{2.5} and component concentrations (left-hand panels of Figure 30 to Figure 34), there is a strong trend of declining concentrations of total-PM_{2.5}, primary- PM_{2.5}, sulphate, nitrate and ammonium from south to north across the UK reflecting (1) a general decline in precursor emission intensity and increasing distance from the largest source-strength areas, and (2) that the separation from source regions is in the opposite direction to the average air flow over this region factor (2) therefore serving to accentuate factor (1). The exceptions to the declining trends in concentrations northwards across Scotland are observed for PM_{2.5} and primary-PM_{2.5} over the urban areas of the central belt and, to a less extent around the Aberdeen and Dundee-Perth urban areas.
- Reductions in Scotland's emissions of primary-PM_{2.5} and the precursor gases barely impacts on the concentrations of modelled PM_{2.5} and its primary and secondary inorganic components outside of Scotland (Figure 35). The 30% reductions in Scotland's emissions leads to a less than 3% reduction in modelled PM_{2.5} component concentrations in northern England and Northern Ireland, and a less than 1% reduction in PM_{2.5} component concentrations over the vast majority of England, Wales and Ireland (and further afield). The influence of Scotland's emissions extends somewhat out to sea to the west, east and north of Scotland, but again the reduction in PM_{2.5} concentrations and its components in these maritime areas is less than 7%. The geographical patterns again reflect that Scotland is not a major source of emissions and that air flow in this region, averaged over the year, is not in the direction from Scotland towards the rest of the British Isles and continent.
- Figure 35 shows that the reduction in the modelled PM_{2.5} concentrations from a 30% reduction in all Scotland emissions is no more than 7% over most of mainland Scotland and has a maximum of less than11% over the Clyde-Forth axis, Stirlingshire, Perth-Dundee and Aberdeen. This geographical pattern is also highlighted in the map of absolute PM_{2.5} concentration changes in the right panel of Figure 30. In the Glasgow, Edinburgh and Stirling urban areas, modelled PM_{2.5} is reduced by up to 0.4 µg/m³ from these emissions reductions.



The conclusion from this analysis is that $PM_{2.5}$ concentrations in Scotland are dominated by $PM_{2.5}$ components derived from emissions outside of Scotland.

- Figure 35 shows that the greatest proportional reduction in PM_{2.5} components resulting from a 30% reduction in all Scotland's emissions simultaneously is for the primary-PM_{2.5} and nitrate components of PM_{2.5}. The component of PM_{2.5} least affected by a reduction in Scotland's emissions is sulphate. Scotland's PM_{2.5} concentrations are therefore most sensitive to changes in primary-PM_{2.5} emissions. More important though is the absolute changes in concentrations arising from these relative changes. These are shown in the mass concentration scales of the Delta (Δ) maps in the right hand panels in Figure 31 to Figure 34. The largest changes in concentrations arise from the changes in primary-PM_{2.5} emissions (Figure 31 *cf*. Figure 32 to Figure 34, after also taking into account the multipliers on the scales of the latter to convert from μgS or μgN to total mass). This confirms that the greatest leverage on Scotland's PM_{2.5} concentrations will arise from reductions of Scotland primary-PM_{2.5} emissions, rather than those emissions in Scotland that contribute to secondary PM_{2.5}.
- The next greatest leverage of Scotland's PM_{2.5} concentrations is through reductions in nitrogen oxides emissions that give rise to the nitrate component , However, care is required in reaching this conclusion because the maps in Figure 31 to Figure 34 are derived from a sensitivity run where sulphur oxides and ammonia emissions were reduced alongside a reduction in nitrogen oxides emissions. The situation may be different if the reduction in emissions of only one species was targeted, but not the other two, because of the chemical interactions between the classes of species. This turns out to be the case when the single species emissions reduction simulations are examined (Figure A6.1 to Figure A6.3). Whilst a reduction in nitrogen oxides emissions alone leads to reductions in nitrate (Figure A6.2) it also leads to an increase in sulphate, because a proportion of the ammonia that would have reacted to create particulate nitrate reacts to create particulate sulphate instead. Therefore, overall there is only a very small net impact on PM_{2.5} concentrations in sulphur oxides and ammonia emissions.
- There are, of course, some geographical variations in the headline observations. In particular, the vast majority of sulphur oxides emissions derive from large, discreet point sources. Figure 32 and Figure A6.1 demonstrate that the major benefit on particulate sulphate of reducing Scotland's sulphur oxides emissions occurs within a few 10s km of the point sources on the Forth estuary. An additional subtlety that is hard to make out is that, as expected, the geographical extent of particulate sulphate reductions resulting from emissions reductions is slightly smaller when ammonia emissions are reduced simultaneously (Figure 32) compared with reductions of sulphur oxides emissions on their own (Figure A6.1).


- The geographical extent of reductions in PM_{2.5} components is greater for ammonium (Figure 34 and Figure A6.3) and for nitrate (Figure 33 and Figure A6.2), than for sulphate, reflecting the more diffuse sources for precursor ammonia and nitrogen oxides than sulphur oxides.
- As discussed above, it must be remembered that net reductions in the secondary inorganic components of Scotland's PM_{2.5} concentrations depend to some degree on whether emissions of the three precursor species are reduced together or in isolation. However, PM_{2.5} concentrations are not reduced by more than 11% (~0.4 µg/m³), and in most areas by considerably less, even for the most influential case where the 30% reductions are applied to all of Scotland's anthropogenic emissions, and for the most sensitive area (the Central Belt). (To a first approximation the source reduction to particle reduction relationship can be taken as linear, particularly for emissions reductions to the lower side of the 30% used in these simulations.) (Figure 30 and Figure 35).
- Overall, the primary-PM_{2.5} and secondary inorganic components of PM_{2.5} concentrations in Scotland are influenced to a greater extent by emissions reductions outside Scotland than within Scotland. However, this observation is based on model simulations with a horizontal resolution of 5 km which is still large compared with the heterogeneity of sources within urbanised areas. The relatively large grid boxes lead to a dilution or homogenisation of discrete emissions sources such as roads. Therefore close to strong primary-PM_{2.5} sources it would be anticipated that reductions in local emissions would have proportionally greater absolute and relative impacts on PM_{2.5} concentrations than simulated in this work. Higher resolution dispersion modelling would be required to address this quantitatively.
- These model scenarios have not included the influence of Scotland's VOC emissions (anthropogenic and biogenic) on the secondary organic particle component of PM_{2.5} concentrations in Scotland. The generally slower timescale of secondary organic aerosol formation compared with secondary inorganic aerosol formation would suggest that the influence of Scotland's VOC emissions on this component of Scotland PM_{2.5} concentrations would not exceed the magnitude of the influences described above for Scotland emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides and ammonia emissions on the primary-PM_{2.5} and secondary inorganic components, and would likely be smaller. However, this is currently only a hypothesis.

Sensitivity of PM_{2.5} Concentrations in Scotland to Shipping Emissions

- 5.70 Figure 36 shows the change in modelled PM_{2.5} (μg/m³) for a 30% reduction in emissions of primary-PM_{2.5}, sulphur oxides, nitrogen oxides, VOCs and carbon monoxide simultaneously, from all shipping in the EMEP4UK inner domain. All other emissions were unaltered.
- 5.71 Reductions in shipping emissions have negligible impact on PM_{2.5} concentrations in Scotland (Figure 36). A 30% reduction in all shipping emissions leads to reductions in PM_{2.5}



concentrations of less than 0.1 μ g/m³ across Scotland, including coastal regions (less than 0.05 μ g/m³ over most of Scotland), except for some larger reductions in PM_{2.5} concentrations in the immediate vicinity of the mid Forth Estuary and Aberdeen (although PM_{2.5} reductions are still less than 0.2 μ g/m³). However, owing to limitations in modelling at 5 km resolution in the vicinity of very high source regions, concentrations of PM_{2.5} immediately adjacent to these port areas (and, the corollary, the reductions in PM_{2.5} from precursor emission reductions) are likely to be greater than simulated by a 5 km x 5 km grid-square average.

5.72 Aside from the two hot-spots of sensitivity to shipping emissions, the impact of a 30% reduction in all shipping emissions results in a reduction of less than 2% in the modelled PM_{2.5} concentrations across Scotland.





30% ship emis. reduction of NO_x,SO_x,VOC,CO,PPM₂₅ 2008 Δμg m⁻³ 30% emissions reduction of NO_x,SO_x,VOC 2008 %Δμg m⁻³

Figure 36: Left panel: Change in modelled $PM_{2.5}$ concentrations (μ g m⁻³) for 30% reduction in emissions of primary- $PM_{2.5}$, sulphur oxides, nitrogen oxides, VOC and CO, simultaneously, from all shipping in the EMEP4UK inner domain (the area encompassed by the figure). All terrestrial emissions unaltered. Right panel: Change in $PM_{2.5}$ expressed relative to the baseline concentration of $PM_{2.5}$. (The baseline $PM_{2.5}$ map is shown in Figure 30 and also Figure A6.1 to Figure A6.3).



6 PM_{2.5} at Grangemouth

- 6.1 Some information on PM_{2.5} at Grangemouth has already been presented. The location of the Grangemouth monitoring station is shown in more detail in relation to the residential areas and to three major local sources of sulphur dioxide, two of which are also significant sources of PM, in Figure 37. The three major sources are:
 - Longannet, a 2.4 GW coal fired power station with a 183 m stack located 4.5 km to the northnortheast, with emissions in 2009 of 459 t/yr of PM₁₀ and 32,228 t/yr of sulphur oxides, giving a PM₁₀:SO_x emission ratio of 0.0143 (the PM_{2.5}:SO_x ratio would be smaller);
 - Ineos Refinery, located 0.6-1.2 km to the north east with emissions in 2009 of 104 t /yr of PM₁₀ and 5,657 t/yr of sulphur oxides, giving a PM₁₀:SO_x emission ratio of 0.0184 (the PM_{2.5}:SO_x ratio would be smaller);
 - BP Exploration, Kinneil Terminal located around 2 km to the east-southeast, which includes power plant and other combustion sources, with 184 t/yr of sulphur oxides emissions in 2009 and no recorded PM₁₀ emissions.
- Based on the emission ratios for $PM_{2.5}$:SO_x, a 1-hour mean concentration of 250 μ g/m³ of sulphur 6.2 dioxide (assuming all sulphur oxides are sulphur dioxide) due to Longannet power station would equate to 3.6 µg/m³ of PM₁₀ and from the Ineos Refinery 4.6 µg/m³. The PM_{2.5} concentrations would be expected to be lower than these values, probably somewhere in the range 2-4 µg/m³. The monitoring data in Table 5 show the annual mean sulphur dioxide concentration at Grangemouth to be around 3-8 µg/m³ higher than the levels recorded at Edinburgh St Leonard's and Glasgow Centre. Assuming this sulphur dioxide enhancement is due largely to the Ineos Refinery (which is most likely, given its proximity to the monitoring site and the lower discharge heights) then it equates to 0.05-0.15 μ g/m³ of PM₁₀ and somewhat less in the case of PM_{2.5}. On this basis around 1% (~0.1 µg/m³) of the PM_{2.5} measured at the Grangemouth monitoring site could be linked to the local industrial sources that are contributors to the elevated sulphur dioxide concentrations. This is somewhat less than the industrial contribution implied by the PCM model of background concentrations, which gives a value of ~0.5 µg/m³ for point sources affecting the 1x1 km grid square in which the monitor is located, with a further 1.1 µg/m³ from more general industrial sources in the surrounding area (the modelled total is 8.4 μ g/m³). The PCM model thus implies that around 20% of the PM_{2.5} in the grid square in which the monitor is located is from industrial sources.
- 6.3 The polar plot for PM_{2.5} at Grangemouth in 2011 is shown in Figure 14 and the polar annulus plot in Figure 15. The plots for Grangemouth show patterns broadly similar to those at the other sites, in particular Edinburgh St Leonard's and Auchencorth Moss (see both Figure 14 and Figure 15). This suggests that local sources are not having a major influence on PM_{2.5} concentrations at



Grangemouth. Further insight into local sources of $PM_{2.5}$ at Grangemouth can be gained by subtracting the regional background, as represented by concentrations measured at Auchencorth Moss, as shown in Figure 38 and Figure 39. These Figures also show the plots for total sulphur dioxide and nitrogen oxides concentrations. There is clear evidence of a $PM_{2.5}$ contribution to higher concentrations coming from the east-northeast at higher wind speeds ~10 m/s, which is consistent with an elevated source (Figure 38). This closely matches the pattern for sulphur dioxide concentrations (Figure 38). The timings of the higher concentrations of both sulphur dioxide and $PM_{2.5}$ in the northeast sector are also similar, being broadly between 03:00 h and 17:00 h, providing further evidence that these are from the same source(s) (Figure 39).



Figure 37: Location of SAUN monitoring site at Grangemouth and nearby major sulphur dioxide sources. Contains Ordnance Survey data © Crown copyright and database right [2012]



6.4 The relationship between local 1-hour mean PM_{2.5} concentrations and the higher sulphur dioxide concentrations (>50 μ g/m³), for wind speeds >5 m/s in the northeast sector, is shown in Figure 40. There is a clear relationship between the two and the slope of the line indicates a PM_{2.5}:SO_x ratio of 0.04, which is higher than the ratio inferred from the site emissions from the Ineos Refinery in 2009 of less than 0.02 (paragraph 6.1). This suggests that there is more PM_{2.5} being released than indicated by the reported emissions, however, the measurements are for 2011, while the emissions are for 2009, and it is possible that the relative emissions have changed over this time. Also, the measured ratio in Figure 40 will reflect emissions from a tall stack, and it is possible that there are other emission sources on the Ineos site included in the values reported to SEPA that have a different (lower ratio), which when added together account for the lower ratio in the reported total emissions. It is also possible that there are fugitive PM_{2.5} emissions that are not reported to SEPA, such that the reported values are only part of the total. Nevertheless, if the PM_{2.5}:SO_x ratio is 0.04 instead of <0.02, then the calculation in paragraph 6.2 would imply around 2% of the PM2.5 measured at the Grangemouth site could be linked to the local industrial sources that are contributors to the elevated sulphur dioxide concentrations. The PCM model would suggest a higher contribution from all industrial sources in the area of ~20% in the 1x1 km grid square with the monitoring site. To reconcile the difference it must be inferred that there are significant industrial sources of PM_{2.5} that are unrelated to sulphur dioxide. Without further work, the true contribution of industrial sources to PM_{2.5} concentrations at the Grangemouth monitoring site must remain unclear.







- 6.5 The polar plots also show an association of the PM_{2.5} with elevated nitrogen oxides concentrations. This is most evident when the wind speeds are low and the winds are from the west to southwest, suggesting a very local source (Figure 38). This local source is likely to be the traffic and domestic combustion sources located in the residential areas predominantly to the west of the monitor. Also evident is a source to the southeast associated with a wide range of wind speeds, which is most evident for nitrogen oxides (Figure 38). This source is most significant between 08:00 to 12:00h, and is also contributing some sulphur dioxide (Figure 39). It is unclear what this source is, although it is probably combustion related.
- 6.6 There is no evidence of a sulphur dioxide contribution from Longannet power station in the polar plot and only slight evidence in the polar annulus plot. This is probably related to the tall stack at the power station. **The Longannet power station is therefore making a negligible contribution**



to $PM_{2.5}$ concentrations at Grangemouth. (It will though contribute to $PM_{2.5}$ more widely through conversion of its nitrogen oxides and sulphur dioxide emissions to secondary nitrates and sulphates – see chapter 5.)



Figure 39: Polar annulus plots of concentrations of $PM_{2.5}$ minus background and total sulphur dioxide and nitrogen oxides at Grangemouth in 2011, as a function of wind direction and time-of-day. The concentrations are shown on the colour scale in μ g/m³. Inside of circle is 00:00-01:00 h running through the day to 23:00-24:00 h.





Figure 40: Local 1-hour mean PM_{2.5} vs. Total 1-hour mean sulphur dioxide at Grangemouth in 2011. Data for wind speeds ≥5 m/s and wind directions 45°-90°. The local PM_{2.5} is Grangemouth minus Auchencorth Moss. The sulphur dioxide concentrations are those ≥50 µg/m³.



7 Health Implications of PM_{2.5}

- 7.1 The health effects associated with PM_{2.5} are set out in the SNIFFER report (Laxen et al., 2010). They are not reviewed here again, although it is noted that there have been no major developments in the understanding of these effects since that report. A series of criteria have been developed by the European Commission (EC) and the UK governments to help regulate exposure to PM_{2.5} (Table 1). These are designed to help protect the public from the adverse health effects of PM_{2.5}, although it is not possible to eliminate adverse health effects, as there is no known threshold below which there are no effects.
- 7.2 The key criteria for Scotland are:
 - the EU Limit Value of 25 µg/m³ as an annual mean not to be exceeded anywhere where people are exposed. This is key because Member States have a mandatory obligation to comply with the Limit Values;
 - the annual mean objective of 12 µg/m³ as an annual mean not to be exceeded anywhere where people are exposed. This is to be met by 2020. This objective is key because it is probably the most stringent criterion within Scotland (it is twice as stringent as the equivalent UK objective). However, there is no mandatory requirement for the Scottish Government to achieve this objective, and it is not included in Regulations, so SEPA and local authorities have no obligations to assess against this objective or work towards meeting it; and
 - the objective of reducing annual mean concentrations at urban background locations by 15% between 2010 and 2010. This will be a policy driver to reduce emissions of primary-PM_{2.5} and emissions of precursor gases. It is the same as or possibly more stringent than the EU exposure-reduction target, which will either be 10% or 15%¹³. The EU exposure-reduction target applies across the whole of the UK, and is unlikely to be applied to Scotland separately¹⁴.
- 7.3 Analysis of the available data, as set out in Table 5 shows that **the EU Limit Value of 25 μg/m³ is currently being met across all sites in Scotland**, although only by a small margin at the Glasgow Kerbside site. The annual mean objective level that has been adopted in Scotland (12 μg/m³) has been exceeded in some years (marginally) at urban background sites, but exceeded by a large amount at the Glasgow Kerbside site. However, the annual mean objective does not apply at this (kerbside) location according to current guidance (Defra, 2009). The annual mean objective does, however, apply at the façade of residential properties (and schools, hospitals and care

¹³ The target will be determined once fully ratified data are available for the UK urban background sites that form the basis for the exposure reduction. The target depends on whether the UK average is below or above 13 μ g/m³.

¹⁴ The exposure reduction target, or objective, requires a stable average concentration at the beginning and end of the reduction period. This requires a substantial number of monitoring sites. The current four urban background sites in Scotland are not likely to be sufficient to meet the requirement.



homes) and if these facades are close to busy roads, then $PM_{2.5}$ concentrations are likely to exceed the objective of 12 µg/m³ (which is to be met by 2020), especially in urban areas, where concentrations are already elevated, unless emissions are reduced.

7.4 Further insight into the potential health significance of PM_{2.5} concentrations in Scotland can be obtained by examining the measured concentrations in relation to the Daily Air Quality Index. The results for the five SAUN sites and one SARN site with PM_{2.5} data are set out in Table 10 and summarised in Figure 41. It is clear that for the majority of the time 24-hour mean PM_{2.5} concentrations are in the Low Band (Index 1-3). Nevertheless, there are occasions when 24-hour mean PM_{2.5} concentrations in urban areas, especially at the kerbside, rise into the High and Very High Bands (Index 7-10). PM_{2.5} concentrations at the Glasgow Kerbside site are in the High and very high bands for almost 2% of the time.



Figure 41: Percentage of rolling 24-hour mean PM_{2.5} values with a given Daily Air Quality Index at five SAUN sites and one SARN site in Scotland, 2009-2011.



Index		Aberdeen Errol Place			Edinburgh St Leonard's			Glasgow Centre			Glasgow Kerbside			Grangemouth			Auchencorth Moss		
		2009	2010	2011	2009	2010	2011	2009	2010	2011	2009	2010	2011	2009	2010	2011	2009	2010	2011
Very High	10								99	21	13	63	23			17 ^a			
High	9							14	15	2	5	27	2			3			
	8							3	31	1	3	50	32			2			
	7							10	29	1	1	85	86			3			
Moderate	6				15			35	60	2	45	147	150	23	36	3			
	5				6		2	35	77	5	45	176	151	11	15	16			
	4		4		50	15	62	120	112	119	87	617	408	44	53	87	4		
Low	3	72	31	282	142	260	396	506	482	317	1034	1925	1874	211	463	340	89	10	88
	2	594	877	1113	1163	1553	2049	2061	1828	1518	1928	4422	4227	1288	1824	1721	303	378	654
	1	4590	6099	6531	6914	6357	6065	5830	5949	6242	920	982	892	6775	5814	5883	8188	5729	8001
^a These very high values are related to Bonfire Night – see text in paragraph 5.15																			

Table 10: Daily Air Quality Index for PM_{2.5}.

These very high values are related to Bonfire Night – see text in paragraph 5.15.



7.5 The health significance of the high concentrations that arise from time to time can be gleaned from the advice issued by Defra to accompany the Daily Air Quality Index, as set out in Table 11.

Band	Index	Accompanying Health Message for At-Risk Groups and the General Population							
		At-Risk Individuals	General Population						
Very High	10	Adults and children with lung problems, adults with heart problems, and older people, should avoid strenuous physical activity. People with asthma may find they need to use their reliever inhaler more often	Reduce physical exertion, particularly outdoors, especially if you experience symptoms such as cough or sore throat.						
	9	Adults and children with lung problems, and adults with heart problems, should reduce	Anyone experiencing discomfort such as sore eyes, cough or sore throat						
High	8	strenuous physical exertion, particularly outdoors. People with asthma may find they need to use their reliever inhaler more often.	should consider reducing activity, particularly outdoors.						
	7	Older people should also reduce physical exertion							
	6	Adults and children with lung problems, and adults with heart	Enjoy your usual outdoor activities.						
Moderate	5	problems, who experience symptoms, should consider							
	4	reducing strenuous physical activity, particularly outdoors							
	3	Enjoy your usual outdoor activities	Enjoy your usual outdoor						
Low	2								
	1								

Table 11: Health Advice to Accompany the Daily Air Quality Index for PM_{2.5}^a

Letter from Dr Clare Bayley of Defra, dated 1 December 2011, entitled Notification of Changes to the Air Quality Index.

7.6 Short-term effects arising from exposure to PM_{2.5} include hospital admissions and deaths of individuals with pre-existing heart and lung conditions, but in overall terms are not as significant as the effects of long-term exposure (Laxen et al., 2010). It is thus more important to focus on the exposure-reduction target. The aim of this target is to reduce overall exposure to PM_{2.5} of the population on an annual mean basis, especially in urban areas.

а



8 Control of Emissions from Regulated Processes

Introduction

- 8.1 The Air Quality Strategy (AQS) for England, Scotland, Wales and Northern Ireland sets the framework for air quality management in the UK (Defra, 2007). Air quality objectives for a number of pollutants form a cornerstone of the strategy. These objectives take account of EU air quality limit values and in some cases are more stringent. There are also differences between the devolved administrations, with the Scottish Government setting separate and more stringent objectives for PM₁₀ and PM_{2.5} (see Chapter 2). Local authorities and the environment agencies (the Scottish Environment Protection Agency, the Environment Agency and the Northern Ireland Environment Agency) play an important role in implementing the Strategy. The environment agencies must *"have regard"* to the UK Air Quality Strategy (AQS) in exercising their pollution control functions. The basis for the control of industrial sources is that all Part A and Part B installations must obtain a permit to operate. These permits include measures to limit emissions to air. Further information on the legislation used to control exposure to PM_{2.5}, including the controls on emissions from the industrial sector, is provided in the SNIFFER report (Laxen et al., 2010).
- 8.2 The exposure-reduction targets for PM_{2.5} set in UK and EU legislation are currently not backed up with any requirements to limit emissions from industrial operations to help meet these targets. It is therefore relevant to examine the basis for control of industrial emissions and to see how it might be possible to extend controls to help meet the exposure reduction targets.

Basis for Control of Industrial Emissions

- 8.3 The application of Best Available Techniques (BAT) is the key instrument available to the environment agencies to limit emissions from industrial installations. It is up to the regulator to determine what BAT is for each installation but there is a strong desire to achieve consistency across the EU. To help achieve this, the European Commission has published 29 BAT Reference documents (BREFs). The BREFs are prepared by technical authors utilising information supplied and considered by national technical experts in a working group. The information is based on what is actually being achieved at existing installations. The definition of BAT requires the techniques identified to be technically and economically viable in the sector as a whole.
- 8.4 Some of the BREFs include the levels of release associated with the use of a particular technique, referred to as BAT associated emission levels (BAT-AELs). There are about 1500 BAT-AELs in the current range of BREFs. The regulator utilises the information on BAT-AELs to determine what Emission Limit Values (ELVs) to set in the permit, taking account of the design, location and operation of the installation.



8.5 Under this system, once an installation is operating to BAT it is generally not possible to require any further reductions in emissions. There are though some exceptions which are discussed below.

Drivers to go Beyond BAT

- 8.6 A range of options are available to allow regulators to go beyond current BAT:
 - The National Emissions Ceilings Directive (2001/81/EC) (NECD);
 - The Large Combustion Plant Directive (2001/80/EC);
 - The Industrial Emissions Directive (2010/75/EU) (IED). This will incorporate the Integrated Pollution Prevention and Control Directive (2008/1/EC) (IPPC Directive);
 - The Ambient Air Quality Directive (2008/50/EC);
 - The Habitats Directive (79/409/EEC); and
 - The Scottish Planning System.

Other opportunities could include:

- Permit Reviews;
- Tightening BAT through the BREF process; and
- Revised regulations/guidance requiring the environment agencies to take measures to help meet the exposure-reduction target.

These are all dealt with in turn below.

National Emission Ceilings Directive

8.7 The NECD sets national emission ceilings applied at Member State level, to be achieved by 2010. The ceilings for the UK were sulphur dioxide (585 kt), nitrogen oxides (1167 kt), volatile organic compounds (1200 kt) and ammonia (297 kt). These emission ceilings have been met for all pollutants other than nitrogen oxides. The NECD is currently being reviewed, with the outcome due in 2013, and new ceilings are expected to be established, which are likely to include PM. The NECD has helped drive down total emissions from the UK without direct reference to BAT.

Large Combustion Plant Directive

8.8 Large combustion plant are significant sources of sulphur dioxide, nitrogen oxides and particulate matter and their emissions are largely controlled through the Large Combustion Plant Directive (2001/80/EC). Operators in the UK had to decide by February 2006 whether each large combustion plant under their control should be regulated under the emission limit values in the Directive, or the National Emissions Reduction Plan (NERP), from 1 January 2008. Alternatively



operators could have opted their plant out from the requirements of the directive with a 20,000 hour limited life and closure by 31 December 2015.

- 8.9 European BAT Reference (BREF) documents contain technical information on the performance of various abatement techniques. Although the BREF for large combustion plant was prepared by a Technical Working Group of European environmental regulators and industry there was not a united view and the BREF contains options. Thus BAT will comprise a combination of controls, including:
 - emission limit values (ELV) for sulphur dioxide, nitrogen oxides and particulate matter;
 - sector 'cap and trade' limits or NERP which limit mass emissions; and
 - controls to protect local air quality.

All these are implemented through installation-specific controls specified in each permit.

- 8.10 For plant which opted for the emission limit value option BAT based ELVs have been set with a sector cap and transferable limits for sulphur dioxide and nitrogen oxides. For plant which opted for the NERP, mass limits have been set for sulphur dioxide and nitrogen oxides based on the technology indicated in the BREF and the concentration of the resultant emission. Operator NERP limits can be transferred between operators of large combustion plant in the NERP. As a result, for plant which opted for the NERP, the mass or concentration of an emission for a specific plant may be required to operate to limits significantly less than the BAT-ELV in the Directive or BREF.
- 8.11 The environment agencies ensure that emissions from power stations of sulphur dioxide, nitrogen oxides and, to a lesser extent, particulate matter, that have an effect on local air quality, are addressed through:
 - A permit condition requiring that emissions from a power station must not cause a breach of EU air quality standards or contribute significantly to a local failure to meet national air quality objectives;
 - Annual mass limits on emissions of sulphur dioxide and nitrogen oxides;
 - An Air Quality Management Plan (AQMP);
 - Monitoring of air quality in the vicinity of power stations.
- 8.12 AQMPs have been in operation at the coal and oil-fired power stations since 2001 and aim to ensure that emissions do not result in a breach of EU air quality standards or contribute significantly to a local failure to meet national air quality objectives. The main features of the AQMP are:



- Demonstration that the anticipated generation scenario and anticipated fuel sulphur content for future years will be compliant with air quality objectives as determined by dispersion modelling;
- A continuous comparison of the number of exceedences monitored at sites close to maximum impact locations with the number anticipated for the planned compliant operational scenario and an assessment of the implications for year-end compliance;
- The development of a number of methodologies for monitoring impacts, dispersion modelling of both retrospective and future station operation, to judge compliance with air quality objectives and to manage the risk of non-compliance associated with load and fuel-sulphur options;
- An annual review which includes: an appraisal of actual impacts during the preceding year; an update on anticipated impacts for the next year using the latest information on anticipated operating pattern and fuel burn; and proposed management actions which might be required to ensure compliance.

The Industrial Emissions Directive

8.13 The requirement to secure tighter controls than those reflecting the use of BAT is maintained in the new Industrial Emissions Directive (IED) (2010/75/EU) on industrial emissions (Integrated Pollution Prevention and Control) (Recast). The Directive states (preamble item 12):

"The permit should include all the measures necessary to achieve a high level of protection of the environment as a whole and to ensure that the installation is operated in accordance with the general principles governing the basic obligations of the operator. The permit should also include emission limit values for polluting substances, or equivalent parameters or technical measures, appropriate requirements to protect the soil and groundwater and monitoring requirements. Permit conditions should be set on the basis of best available techniques."

8.14 The IED reflects the requirements in the IPPC Directive to take account of environmental quality standards. Article 18 states:

"Where an environmental quality standard requires stricter conditions than those achievable by the use of the best available techniques, additional measures shall be included in the permit, without prejudice to other measures which may be taken to comply with environmental quality standards."

- 8.15 The IED has to be transposed into UK law by 6 January 2013.
- 8.16 As noted above, the IED incorporates the requirements of the IPPC Directive 2008/1/EC which states:

"... the emission limit values and the equivalent parameters and technical measures referred to in paragraph 3 shall be based on the best available techniques, without prescribing the use of any technique or specific technology, but taking into account the technical characteristics of the



installation concerned, its geographical location and the local environmental conditions. In all circumstances, the conditions of the permit shall contain provisions on the minimisation of longdistance or transboundary pollution and ensure a high level of protection for the environment as a whole."

8.17 It also states:

"Where an environmental quality standard requires stricter conditions than those achievable by the use of the best available techniques, additional measures shall in particular be required in the permit, without prejudice to other measures which might be taken to comply with environmental quality standards."

The term *"environmental quality standard"* has been defined in the Pollution Prevention and Control (Scotland) Regulations 2000 to mean *"the set of requirements which must be fulfilled at a given time by a given environment or particular part thereof, as set out in Community legislation."*

8.18 In other words the regulator is required to set the conditions which can be achieved by the use of the BAT and to impose tighter conditions if required to meet air quality standards.

The Ambient Air Quality Directive

- 8.19 The 2008 ambient air quality directive (2008/50/EC) sets legally binding limits for concentrations in outdoor air of major air pollutants that affect public health. These cover sulphur dioxide, nitrogen dioxide, benzene, carbon monoxide, lead, PM₁₀ and PM_{2.5}. It replaced nearly all the previous EU air quality legislation.
- 8.20 Under the Air Quality Standards (Scotland) Regulations 2010 that enacts this directive the Scottish Ministers must ensure that the limit values are not exceeded, and where levels of the pollutants are below the limit values the Ministers must ensure that they are maintained below the limit values and must endeavour to maintain the best ambient air quality compatible with sustainable development.
- 8.21 The Scottish Ministers must also ensure that all necessary measures not entailing disproportionate costs are taken to ensure that concentrations of $PM_{2.5}$ do not exceed the 2010 target value of an annual mean of 25 µg/m³.
- 8.22 The Scottish Ministers have discharged these responsibilities through the development of air quality strategies and directions and guidance to local authorities and SEPA.
- 8.23 Where an installation is making a significant contribution to the exceedence of an air quality standard then the environment agencies can and may require the operator to take additional action. These actions include:



- Requiring ambient air quality monitoring around the installation and/or where the releases from the installation are likely to ground.
- Requiring reductions in activity or changes in processes when exceedences of air quality standards are occurring or are likely to occur.

These actions are justified as being consistent with the application of the BAT for the installation and protection of the environment as a whole.

8.24 Further understanding of the approach that has been adopted can be gained from the guidance produced by the Environment Agency (England and Wales) (4 August 2010) and issued to its staff which states that:

"If the emissions from an installation alone could lead to a breach of an EU AQ limit value then we must include permit conditions to prevent this. However, we have found that the usual circumstance is that where an EU AQ limit value is breached, it is mainly a result of emissions from non-Agency-regulated sources i.e. traffic. Under these circumstances we have to take a view on what level of reduction should be borne by the installation."

"We will investigate what improvements can be made if we find an installation we regulate is projected to contribute significantly to the breach of an EU AQ limit value. Previously, EU limit values applied at all locations outside the installation's boundary, whether there was relevant public exposure or not. The transposition of the new Ambient AQ Directive into UK law means, in simple terms, that limit values now only apply at locations where there is relevant public exposure. The Environment Agency must set more stringent emission limits or other controls than would be the case under BAT, or appropriate measures for waste operations, if they are needed to achieve compliance with an EU limit value where the installation is making a significant contribution."

"Similarly we may find that an installation is contributing significantly to the breach of an EU AQ target value. We will investigate what improvements can be made within BAT or appropriate measures and require the operator to implement these. But this may not completely remove the exceedence of the target value or in the timescale required by the 4th AQDD."

"We have reviewed the releases of air pollutants from the sites we regulate and have set corporate annual targets for reducing these. We are implementing BAT and appropriate measures to deliver these targets and this will contribute to the UK's obligations under the EU Ceilings and 3rd AQD Directives."

"The new EU Ambient AQ Directive contains requirements on Member States to address exposure reduction for $PM_{2.5}$. We would expect that our actions to help deliver the UK's exposure reduction targets will make a contribution to this."



The Habitats Directive

- 8.25 The Habitats Directive (92/43/EEC) requires sites which are important for either habitats or species (listed in Annexes I and II of the Habitats Directive respectively) to be designated as Special Areas of Conservation (SACs). These sites and Special Protection Areas (SPAs) classified under Council Directive 79/409/EEC on the Conservation of Wild Birds (the Birds Directive) form the Natura 2000 network.
- 8.26 The Directive requires member states to take measures to maintain or restore the relevant natural habitats and wild species to a favourable conservation status. When determining an environmental permit application that is likely to have a significant effect on a designated site under the Habitats Regulations, the national regulator must carry out an Appropriate Assessment of the implications for the site in view of that site's conservation objectives. These assessments should ascertain whether or not a permit application will have an adverse effect on the integrity of the site interest features. The assessment may therefore cause the permit application to be rejected, or to be granted subject to stringent conditions to protect the designated site. These conditions may include controls on the production or release of substances into the air. A significant pollutant of concern has been ammonia from intensive pig and poultry units. Modelling has identified potential impacts on local ammonia concentrations and the need for additional controls at some units, but only in a few cases has any impact on the designated feature been identified and additional conditions required. The Simple Calculation of Atmospheric Impact Limits (SCAIL) screening tool made available through the Air Pollution Information System (APIS) has been successfully used by regulatory agencies and farmers to help adapt farming practices and reduce ammonia emissions, both as direct ammonia gas emissions and N-rich particulate matter (PM). The control on ammonia emissions will help to contribute towards an overall reduction in background PM across the UK, particularly the secondary particulates associated with finer ammonia aerosols i.e. those that are PM_{2.5} and below.
- 8.27 The requirement to take into account the findings of an Appropriate Assessment when determining permit applications could clearly lead to a requirement to go beyond BAT to permit the process to operate.
- 8.28 The environment agencies have agreed with the Electricity Supply Industry Joint Environmental Programme that each permit application for a power station should list all European Sites and Sites of Special Scientific Interest (SSSIs) within England, Wales, Scotland and Northern Ireland where the process contribution was >1% Critical Load for acid deposition and nutrient deposition. The risks associated with the aerial and aqueous emissions from each power station, and the sensitive habitats and/or species were identified. A programme of actions and improvements was then agreed with each operator.



The Scottish Planning System

8.29 The spatial and transport planning system in the UK will play a major role over the longer term in reducing both the sources of air pollution and the exposure of individuals to poor air quality. The **environment agencies have an important role as statutory or recommended consultees in the spatial and transport planning processes.** In many cases the location of an installation or new road can have a significant effect on the air quality experienced by the local population. This is irrespective of the actual direct impact on health. SEPA is identified as a key agency under the Planning etc. (Scotland) Act 2006 and as such, it provides formal environmental advice in relation to development plans and a wide range of development proposals across Scotland. SEPA will use the information provided to identify 'showstoppers' and establish if the proposed development is capable of being consented under the licensing regime.

Tightening BAT via BREFs

8.30 BAT and associated emission limit values are derived directly and indirectly from BREFs. There is a rolling programme for the revision of BREFs. The tightest, most consistent and clearly applied controls are emission limits in the Directives, e.g. the Large Combustion Plants Directive, but as they are inflexible the EU preference is to secure hard numbers in the BREFs. The weakest approach is where a range of control techniques are identified each with their own Associated Emission Levels (AEL), which usually arises as a result of a lack of agreement on a preferred technique. There can also be site specific debates over the appropriateness of the technology proposed, its cost and timing for making changes, which can affect the final emission levels.

Permit Reviews

8.31 Opportunities for tightening up on emissions arise through the process of permit reviews which SEPA is required to do periodically. These reviews should check whether permit conditions continue to reflect appropriate standards and remain adequate in light of experience and new knowledge. They should guard against permits becoming obsolete as techniques develop. The review should take account of new information on environmental effects, best available techniques or other relevant issues.

Revised Regulations/Guidance

- 8.32 The phrase in the Regulations and Guidance to the regulators to *"have regard to the UK Air Quality Strategy"* is open to wide interpretation and could be significantly reinforced by policy guidance from sponsoring government departments. Linking the control of the source of pollution to the desired outcome in the environment could result in more proportionate regulation.
- 8.33 A further issue is that the environment agencies do not have direct responsibilities imposed on them for the delivery of EU obligations. However, if challenged it is likely that it could be argued



that, as representatives of the State, they do have responsibility for the enforcement of EU obligations.

Conclusions

- 8.34 With regard to emissions arising from industrial installations, the EU Directives and UK Regulations achieve the greatest contribution towards the achievement and maintenance of air quality standards through the application of conditions in individual site permits based on the use of the best available techniques (BAT). Controls which go beyond the requirements of BAT can be, and are being, applied on the grounds of health and environmental protection. However, they are very site specific, and there is less legal clarity as to how far the installations responsible for the emissions can be pushed beyond the application of BAT.
- 8.35 A principal obstruction to the enforced application of measures beyond BAT with regard to the PM_{2.5} exposure-reduction target, is that **it would be very difficult to demonstrate that an individual installation was making a significant contribution to non-compliance with the target** (as the target is based on measurements averaged across the UK urban agglomerations).



9 **References**

Beevers, S., Carslaw, D., Westmoreland, E. and Mittal, H. (2009) *Air Pollution and Emissiosn Trends in London*, King's College London and the Institute for Transport Studies, Leeds, April 2009.

Brookes, D.M., Stedman, J.R., Grice, S.E., Kent, A.J., Walker, H.L., Cooke, S.L., Vincent, K.J., Lingard, J.J.N., Bush, T.J. and Abbott, J. (2011) *UK Modelling Under the Air Quality Directive (2008/50/EC) for 2010 Covering the Following Air Quality Pollutants: SO2, NOx, NO2, PM10, PM2.5, Lead, Benzene, CO and Ozone*, Rept AEAT/ENV/R/3215 Issue 1, November 2011, AEA, Harwell.

Carslaw, D. (2011) *Defra regional and transboundary model evaluation analysis - Phase 1, A report for Defra and the Devolved Administrations*, [Online], Available: <u>http://uk-air.defra.gov.uk/reports/cat20/1105091514_RegionalFinal.pdf</u>.

Carslaw, D., Beevers, S., Westmoreland, E. and Williams, M. (2011) *Trends in NOx and NO2 emissions and ambient measurements in the UK*, [Online], Available: <u>uk-air.defra.gov.uk/reports/cat05/1108251149</u> 110718 AQ0724 Final report.pdf.

Carslaw, D. and Ropkins, K. (2012) *Openair: Open-source tools for the analysis of air pollution data*, [Online], Available: <u>http://openair-project.org</u>.

Defra (2007) *The Air Quality Strategy for England, Scotland, Wales and Northern Ireland,* Defra.

Defra (2009) Review & Assessment: Technical Guidance LAQM.TG(09), Defra.

Eaton, S. and Stacey, B. (2011) QA/QC Data Ratification Report for the Automatic Urban and Rural Network, July-September 2011, and Intercalibration Report, Summer 2011, AEAT/ENV/R/3248 Issue 1.

Fagerli, H., Gauss, M., Benedictow, A., Griesfeller, J., Jonson, J.E., Nyiri, A., Schulz, M., Simpson, D., Steensen, B.M., Tsyro, S., Valdebenito, A., Wind, P., Aas, W., Hjellbrekke, A.G., Mareckova, K., Wankmüller, R., Iversen, T., Kirkevåg, A., Seland, Ø. and Vieno, M. (2011) *Transboundary acidification, eutrophication and ground level ozone in Europe in 2009.*, Oslo, Norway: EMEP Status Report 1/2011, Norwegian Meteorological Institute.

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.

Heal, M.R., Naysmith, P., Cook, G.T., Xu, S., Raventos Duran, T. and Harrison, R.M. (2011) *Application of 14C analyses to source apportionment of carbonaceous PM2.5 in the UK*, Atmospheric Environment 45, 2341-2348.

Jimenez, J., Canagaratna, M., Donahue, N., Prevot, A., Zhang, Q., Kroll, J., DeCarlo, P., Allan, J., Coe, H., Ng, N., Aiken, A., Docherty, K., Ulbrich, I., Grieshop, A., Robinson, A., Duplissy, J., Smith, J., Wilson, K., Lanz, V., Hueglin, C. et al. (2009) *Evolution of organic aerosols in the atmosphere*, Science 326, 1525-1529.



Jones, A.M. and Harrison, R.M. (2011) *Temporal trends in sulphate concentrations at European sites and relationships to sulphur dioxide*, Atmospheric Environment 45, 873-882.

Laxen, D., Moorcroft, S., Laxen, K., Boulter, P., Barlow, T., Harrison, R. and Heal, M. (2010) *PM2.5 in the UK*, Edinburgh: SNIFFER.

MacCarthy, J., Li, Y., Murrells, T.P., Okamura, S., Passant, N., Sneddon, S., Martinez, C., Thomas, J., Thistlethwaite, G. and Misselbrook, T. (2010) *Air Quality Pollutant Inventories for England, Scotland, Wales and Northern Ireland: 1990-2008*, Report: AEAT/ENV/R/3073, October 2010, AEA, Harwell.

R Development Core Team (2012) *R Project for Statistical Computing*, [Online], Available: <u>http://www.R-project.org</u>.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L.D., Fagerli, H., Hayman, G.D., Gauss, M., Jonson, J.E., Jenkin, M.E., Nyíri, A., Richter, C., Semeena, V.S., Tsyro, S., Tuovinen, J.P., Valdebenito, Á. and Wind, P. (2012) *The EMEP MSC-W chemical transport model. Part 1: Model description*, Atmospheric Chemistry and Physics Discussions 12, 3781-3874.

Tang, Y.S., Simmons, I., van Dijk, N., Di Marco, C., Nemitz, E., Daemmgen, U., Gilke, K., Djuricic, V., Vidic, S., Gliha, Z., Borovecki, D., Mitosinkova, M., Hanssen, J.E., Uggerud, T.H., Sanz, M.J., Sanz, P., Chorda, J.V., Flechard, C.R., Fauvel, Y., Ferm, M. et al. (2009) *European scale application of atmospheric reactive nitrogen measurements in a low-cost approach to infer dry deposition fluxes*, Agriculture Ecosystems & Environment, 133, 183-195.

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. (2010) *Modelling surface ozone during the 2003 heat-wave in the UK*, Atmospheric Chemistry and Physics 10, 7963-7978.

Yim, S.H.L. and Barrett, S.R.H. (2012) *Public Health Impacts of Combustion Emissions in the United Kingdom*, Environmental Science & Technology, 46, 4291-4296.

Zhang, Q., Jimenez, J.L., Canagaratna, M.R., Allan, J.D., Coe, H., Ulbrich, I., Alfarra, M.R., Takami, A., Middlebrook, A.M., Sun, Y.L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P.F., Salcedo, D., Onasch, T., Jayne, J.T., Miyoshi, T., Shimono, A., Hatakeyama, S. et al. (2007) *Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes*, Geophysical Research Letters 34, L13801.



10 Glossary

- AEL Associated Emission Level
- **AQMP** Air Quality Management Plan
- **BAT** Best Available Techniques
- BREF BAT Reference documents
- Defra Department for Environment, Food and Rural Affairs
- ELV Emission Limit Value
- **EMEP** European Monitoring and Evaluation Programme
- EU European Union
- **Exceedence** A period of time when the concentration of a pollutant is greater than the appropriate air quality objective. This applies to specified locations with relevant exposure
- IED Industrial Emissions Directive
- **IPPC** Integrated Pollution Prevention and Control

Limit Values A set of health-based concentrations not to be exceeded, established by the European Commission on behalf of the European Union

- **NECD** National Emissions Ceilings Directive
- NERP National Emissions Reduction Plan
- **µg/m³** Microgrammes per cubic metre
- **Objectives** A nationally defined set of health-based concentrations for nine pollutants, seven of which are incorporated in Regulations, setting out the extent to which the standards should be achieved by a defined date. There are also vegetation-based objectives for sulphur dioxide and nitrogen oxides
- **PM**₁₀ Small airborne particles, more specifically particulate matter less than 10 micrometres in aerodynamic diameter
- PM_{2.5} Small airborne particles less than 2.5 micrometres in aerodynamic diameter
- SARN Scottish Automatic Rural Network of air quality monitoring sites
- SAUN Scottish Automatic Urban Network of air quality monitoring sites
- SEPA Scottish Environment Protection Agency
- **Standards** A nationally defined set of concentrations for nine pollutants below which health effects do not occur or are minimal
- VOC Volatile Organic Compound



11 Appendices

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A1 Locations of Monitoring Sites

A1.1 The locations of the monitoring sites for which data are used in this study are shown in Figure A1.1.



Figure A1.1: Locations of monitoring sites in Scotland cited in this report.



A2 PM_{2.5} Data

- A2.1 1-hour mean PM_{2.5} concentrations at the five SAUN sites and one SARN site in Scotland are presented in Figure A2.1. The concentration scale has been curtailed to between 0 and 100 μg/m³ for presentation purposes maximum concentrations rise to around 300 μg/m³.
- A2.2 Figure A2.2 shows the 1-hour mean concentrations for Auchencorth Moss and Edinburgh St Leonard's sites on an expanded scale of -10 to $50 \ \mu g/m^3$.











Figure A2.2: 1-hour Mean PM_{2.5} concentrations (μg/m³) at Auchencorth Moss and Edinburgh St Leonard's, 2010-2011. Scale constrained to -10 to 50 μg/m³.



A3 Nitrate, Sulphate, Ammonium and Chloride, 2000-2010

A3.1 Figure A3.1 shows the nitrate, sulphate, ammonium and chloride concentrations measured at the AGANET sites between 2000 and 2010.



Figure A3.1: Annual mean concentrations of nitrate, sulphate, ammonium (µg/m³) at five AGANET sites.



A4 Modelled PM_{2.5} Concentrations in Central Scotland

A4.1 The following figures repeat Figure 27 and Figure 28, but expanded to the central area of Scotland.









Figure A4.2: Modelled contribution of industrial (in-square, non-point source) emissions to annual mean background concentrations of $PM_{2.5}$ in Central Scotland in 2011 (μ g/m³).



A5 EMEP Unified Model and EMEP4UK

Model Descriptions

A5.1 The EMEP Unified Model (<u>www.emep.int/OpenSource/</u>) is a grid-based chemistry-transport model comprising a collection of model pre-processors and post-processors that work together to produce a detailed representation of the physical and chemical state of the atmosphere at 50 km x 50 km horizontal resolution over a region encompassing Europe, the north Atlantic, North Africa, the Caucasus and parts of Russia (Simpson et al., 2012). The geographical coverage is illustrated in Figure A5.1.



- Figure A5.1: Geographical coverage of full EMEP Unified Model domain at 50 km horizontal resolution (red region), and of the British Isles inner domain at 5 km horizontal resolution applied in the EMEP4UK model (yellow region). (The intermediate domain at 10 km resolution is used in a model nesting procedure to go from one spatial scale to the other.
- A5.2 The EMEP4UK model is a development of the EMEP UM to enable application at 5 km resolution over a British Isles inner domain embedded within the main EMEP UM 50 km domain (Figure A5.1) (Vieno et al., 2010). The EMEP4UK model is driven by output from the Weather Research Forecast (WRF) model at the same 5 km horizontal resolution. The WRF model in turn is constrained by boundary conditions from the US National Center for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) Global Forecast System (GFS) at 1°



resolution, every 6 hours, and includes data assimilation. Both the WRF and EMEP UM (and EMEP4UK) models have 20 vertical layers, with terrain-following coordinates, and vertical resolution increasing towards the surface. The surface layer has a height of 90 m, and the vertical column extends up to 100 hPa (~16 km). The exact area designated as Scotland is shown in Figure A2. Grid squares extending into maritime regions were included to ensure that emissions right on the coast were definitely excluded in model simulations of emissions sensitivity.



Figure A5.2: Illustration of the 5 km grid squares of the EMEP4UK model designated as Scotland. Grid squares extending into maritime regions were included to ensure that emissions right on the coast were definitely excluded in model simulations of emissions sensitivity.

A5.3 Emissions in the EMEP4UK inner domain are from the 1 km x 1 km resolution UK National Atmospheric Emissions Inventory (<u>http://naei.defra.gov.uk</u>) integrated to match the 5 km x 5 km resolution of the EMEP4UK model. Emissions for the rest of the EMEP domain are at 50 km resolution as supplied to EMEP. Annual totals of anthropogenic emissions are apportioned into hourly emissions using daily, weekly and monthly factors defined for each source and species. In addition to anthropogenic emissions, biogenic emissions of isoprene and other biogenic volatile



organic compounds (biogenic-VOC) are included whose magnitudes are dependent on land-cover vegetation, air temperature and photosynthetically-active radiation.

- A5.4 Modelled chemistry includes gas-phase reactions, scavenging into the atmospheric aqueous phase and formation of inorganic components in particles (Simpson et al., 2012). Wet and dry deposition of gases and particles are included, the former linked directly to the simulated rainfall.
- A5.5 Both the EMEP UM and EMEP4UK model versions have been extensively validated and used for numerous policy applications (Fagerli et al., 2011) (Carslaw, 2011).

Scope and Limitations of EMEP UK and EMEP4UK

A5.6 The modelled PM_{2.5} concentrations comprise:

Primary $PM_{2.5}$ + sea-salt(fine) + SO_4^{2-} + NO_3^{-} (fine) + NH_4^{+}

Where:

- Primary-PM_{2.5} is the primary anthropogenic component defined using PM_{2.5} emissions supplied by NAEI (for the UK) and EMEP (for the rest of the model domain). This component comprises primary elemental and organic carbonaceous material, plus any other inventoried primary-PM_{2.5}. Whilst this include estimates of primary-PM_{2.5} from vehicle brake and tyre wear it does not include estimates for PM_{2.5} from road resuspension since this latter is estimated as a proportion of the exhaust and vehicle-wear emissions so their inclusion would lead to a double-counting of emissions. (On the other hand its omission will lead to model underestimate of primary-PM_{2.5} close to roads.)
- Sea-salt (fine) covers primary sea-salt particles in the PM_{2.5} size fraction estimated using an algorithm dependent on the wind speed over marine areas.
- The three secondary inorganic species (sulphate, nitrate and ammonium) are derived from gas and condensed-phase chemical reactions involving the gaseous precursors sulphur dioxide, nitrogen oxides (i.e. nitric oxide (NO) and nitrogen dioxide (NO₂)) and ammonia. The EMEP models assign all sulphate and ammonium to PM_{2.5} but nitrate can be present in both PM_{2.5} and PM_{coarse} (i.e. particles in the size range between PM₁₀ and PM_{2.5}). It is important to note that the formation and partitioning into particles of the secondary inorganic components also depends on other emissions in the model (VOCs, biogenic-VOCs, carbon monoxide, etc.) through the impact that the chemistry of these emissions have on the oxidative capacity of the atmosphere.
- A5.7 When comparing model output with measured total mass of PM_{2.5}, it is therefore important to note that EMEP UM and EMEP4UK modelled PM_{2.5} concentrations do not include the following:
 - <u>Secondary organic aerosol (SOA)</u>. This is semi-volatile or low-volatility carbonaceous material formed by chemical reactions of VOCs and biogenic-VOCs emitted into the atmosphere, which



condenses into new particles or onto pre-existing particles. Even for a UK urban background location (in Birmingham) the biogenic-VOC-derived component of secondary organic aerosol was shown to comprise ~18% of $PM_{2.5}$ mass on average (uncertainty range 9-29%) (Heal et al., 2011). Whilst some secondary organic aerosol will be derived from precursors from comparatively local sources, the majority will be due to a ubiquitous regional background of $PM_{2.5}$ material (Jimenez et al., 2009) (Zhang et al., 2007).

- <u>Natural wind-suspended "dust" particles and road resuspended particles.</u> The amount of the former within PM_{2.5} is not expected to be great, but the latter is likely to be important in reconciling model and measured PM_{2.5} concentrations close to roads.
- <u>Water of hydration.</u> Water soluble components, especially the inorganic salts ammonium sulphate ((NH₄)₂SO₄), ammonium nitrate (NH₄NO₃) and sodium chloride (NaCl), take up water from the atmosphere (deliquesce) which increases the masses of these components. The extent to which this contributes a negative bias of model output against measurement depends on the temperature and humidity conditions prevailing at the time the PM_{2.5} mass is measured, but it can be significant. For a study in Birmingham, a deliquescence multiplier of 1.29 was applied to the masses of both the (NH₄)₂SO₄ and NH₄NO₃ component masses (Harrison et al., 2003).
- A5.8 The most informative comparisons of EMEP4UK model output and measurements for PM_{2.5} are therefore for specific chemical components of PM_{2.5}. However, such data are very limited. The only long-running chemically-speciated PM data are from the DELTA (DEnuder for Long-Term Atmospheric sampling) denuders of the UK Acid Gases and Aerosols Monitoring Network (AGANET). These provide monthly averages of particle-bound sulphate, nitrate and ammonium concentrations (and of acid gases) at ~30 sites around the UK. Further details are at http://pollutantdeposition.defra.gov.uk/aganet.
- A5.9 The size cut-off of the DELTA sampler has been estimated to be ~4.5 μm (Tang et al., 2009), so measured concentrations lie between PM_{2.5} and PM₁₀. The EMEP and EMEP4UK models assign all sulphate and ammonium components to PM_{2.5}, so model output and measurements can be compared directly. However, modelled nitrate is assigned to both the PM_{2.5} and PM_{coarse} fractions. Therefore, in the time series shown here, both modelled PM_{2.5} and PM₁₀ nitrate are plotted, represented by the lower and upper edges, respectively, of the shaded region in the modelled nitrate time series. The measured AGANET nitrate should therefore lie within the width of the model shaded region.
- A5.10 The AGANET sites are all rural, apart from one urban background site in Edinburgh. Time series of monthly comparisons of sulphate, nitrate and ammonium for 2008 for the rural Bush Estate site (south of Edinburgh) are shown Figure A5.3, and for the Edinburgh urban background site in Figure A5.4.


- A5.11 Concordance between modelled and measured inorganic components is generally very good at the Bush Estate rural site (Figure A5.3). There is a tendency for some model negative bias for ammonium but values for sulphate and nitrate averaged over the year are comparable. The model captures monthly trends for all three components. Comparison is less favourable at the Edinburgh urban background site (Figure A5.4). Here there is more significant negative bias for ammonium and nitrate (but not, on average, for sulphate) although again the model has generally captured the monthly trends for all three components.
- A5.12 It must also be remembered that measurements are from a point locality whereas modelled values are the averages for the 5 km x 5 km x 90 m grid volume containing the measurement site. It is not possible for the model to capture spatial heterogeneity in emissions and chemistry, associated with traffic and other sources occurring at finer spatial scales. It must also be remembered that measured values, like modelled values, are subject to their own uncertainty and potential biases, particularly, in the case here, a failure to quantify correctly the division between gas and particle-phase acid components.
- A5.13 Overall, there is confidence that the model is consistent in capturing the source-receptor chemicaltransport relationships between primary emissions and primary and secondary inorganic PM_{2.5}.





Figure A5.3: Monthly-averaged sulphate, nitrate and ammonium from the EMEP4UK model and from the DELTA denuder for the Bush Estate rural AGANET site (south of Edinburgh). The lower and upper edges of the shaded region in the nitrate panel represent modelled nitrate assigned to PM_{2.5} and PM₁₀ respectively. See main text for further explanation.





Figure A5.4: Monthly-averaged sulphate, nitrate and ammonium from the EMEP4UK model and from the DELTA denuder for the Edinburgh urban background AGANET site. The lower and upper edges of the shaded region in the nitrate panel represent modelled nitrate assigned to PM_{2.5} and PM₁₀ respectively. See main text for further explanation. (Labels should all refer to Edinburgh St. Leonards, not Johnston Terrace.)



A6 EMEP4UK Results for Reductions in Single Components

- A6.1 The following Figures show the changes in annual mean concentrations of PM_{2.5} and the individual component (sulphate, nitrate or ammonium), following a 30% reduction in anthropogenic emissions of the individual precursors (sulphur oxides, nitrogen oxides or ammonia) only.
- A6.2 Figure A6.1 illustrates the reductions in the modelled sulphate component and in total PM_{2.5} from 30% reduction in Scotland's anthropogenic emissions of sulphur oxides only. Figure A6.2 illustrates the reductions in the modelled nitrate component and in total PM_{2.5} from 30% reduction in Scotland's anthropogenic emissions of nitrogen oxides only. Figure A6.3 illustrates the reductions in the modelled ammonium component and in total PM_{2.5} from 30% reduction in Scotland's anthropogenic emissions of ammonia only. The impact of 30% reduction in emissions of primary-PM_{2.5} only is not shown since this is the same as illustrated in Figures 5 & 9. (Primary PM_{2.5} is not subject to chemical transformation so there is no interaction between it and the precursor gases.)















Figure A6.1: Impact on modelled sulphate component (μgS/m³) from 30% reduction in Scotland anthropogenic emissions of sulphur oxides only. Concentrations as μg/m³ sulphate will be 3.0 times higher.

(a) Left panel is baseline sulphate, centre panel is reduction (Δ) in sulphate, right panel is reduction in sulphate as a %. (b) Left panel is baseline PM_{2.5}, centre panel is reduction (Δ) in PM_{2.5}, right panel is reduction in PM_{2.5}, as a %. Baseline maps of components are the same regardless of the emissions reduction scenario but are reproduced here for convenience of comparison.



(a)





Figure A6.2: Impact on modelled nitrate component (μgN/m³) from 30% reduction in Scotland anthropogenic emissions of nitrogen oxides only. Concentrations as μg/m³ nitrate will be 4.43 times higher.

(a) Left panel is baseline nitrate, centre panel is reduction (Δ) in nitrate, right panel is reduction in nitrate as a %. (b) Left panel is baseline PM_{2.5}, centre panel is reduction (Δ) in PM_{2.5}, right panel is reduction in PM_{2.5} as a %. Baseline maps of components are the same regardless of the emissions reduction scenario but are reproduced here for convenience of comparison. Note the different scale for the map of % change in PM_{2.5} (bottom right) compared with other % change maps in Figure A6.1 and Figure A6.3.



(a)













(a) Left panel is baseline ammonium, centre panel is reduction (Δ) in ammonium, right panel is reduction in ammonium as a %. (b) Left panel is baseline PM_{2.5}, centre panel is reduction (Δ) in PM_{2.5}, right panel is reduction in PM_{2.5} as a %. Baseline maps of components are se the same regardless of the emissions reduction scenario but are reproduced here for convenience of comparison.



A7 Emission Trends in Scotland 1990-2009

A7.1 The following three figures show trends in emissions of nitrogen oxides, sulphur dioxide and ammonia from Scotland, 1990-2009.



Figure A7.1: Trends in nitrogen oxides emissions from Scotland, 1990-2009, from NAEI (kt/yr).





Figure A7.2: Trends in sulphur dioxide emissions from Scotland, 1990-2009, from NAEI (kt/yr).



1990 1991 1992 1993 1994 1995 1996 1997 1998 1999 2000 2001 2002 2003 2004 2005 2006 2007 2008 2009

