

Executive Summary

1. Particulate matter (PM) is the term used to describe condensed phase (solid or liquid) particles suspended in the atmosphere. Their potential for causing health problems is directly linked to the size of the particles. A growing body of research has pointed towards the smaller particles, in particular PM less than 2.5 μm in diameter ($\text{PM}_{2.5}$), as a metric more closely associated with adverse health effects than other metrics such as PM_{10} (particles with a diameter less than 10 μm).
2. The EU's Air Quality Directive, the Directive on Ambient Air Quality and Cleaner Air for Europe (2008/50/EC), defines standards by which air pollution can be assessed and establishes specific air quality objectives. To achieve these objectives there are a number of other legislative instruments which aim to reduce air pollution by controlling emission sources. In the Directive a new approach for $\text{PM}_{2.5}$ was introduced in recognition of the lack of evidence to indicate that there is a concentration of particulate matter below which health effects do not occur. This new approach aims to achieve a reduction in the overall exposure of the population to $\text{PM}_{2.5}$ based on the concept that greater public health benefits could be obtained from a general reduction in exposure than from a policy aimed at reducing exposure in hot spots only. The focus of legislation for $\text{PM}_{2.5}$ is on limiting long-term exposure through the use of annual standards, coupled to a reduction of $\text{PM}_{2.5}$ background concentration in urban areas across the UK over the period 2010-2020. The National Emission Ceilings Directive (2001/81/EC) and the recent revision of the UNECE Convention on Long Range Transboundary Air Pollution's Gothenburg Protocol both aim to reduce transboundary transport of a number of air pollutants which affect the concentrations of background $\text{PM}_{2.5}$ by setting limits on their emission from member states.
3. This report, prepared by the Air Quality Expert Group (AQEG) for Defra and the Devolved Administrations gives an overview of the evidence base for $\text{PM}_{2.5}$ in the UK. The report challenges the robustness of the evidence for making future policy decisions in respect of $\text{PM}_{2.5}$ in the UK context. There is an analysis of the evidence concerning key relevant aspects including $\text{PM}_{2.5}$ measurement and the composition and current concentrations of $\text{PM}_{2.5}$ across the UK, as well as source emissions and receptor modelling for $\text{PM}_{2.5}$. Finally, AQEG evaluates the methods for modelling $\text{PM}_{2.5}$ and what can be said about future concentrations. The report concludes with an assessment of the key uncertainties and gaps in the evidence base that require action.

I.1 Measurement

4. The measurement of $\text{PM}_{2.5}$ mass concentration is a demanding task, as the metric does not correspond to a definite physical or chemical component of the air but is in effect defined by the measurement method itself. The reference method for legislative purposes is declared in the Air Quality Directive to be that described in the CEN European standard EN 14907.
5. $\text{PM}_{2.5}$ contains a large proportion of semi-volatile and hygroscopic material which means that the $\text{PM}_{2.5}$ mass is subject to variation due to environmental conditions during and after sampling. The difficulties of PM measurement are reflected in the fact that the required measurement uncertainty for PM in the

Air Quality Directive is $\pm 25\%$, with a 95% level of confidence, at concentrations close to the limit value; for most other gaseous pollutants the comparable value is $\pm 15\%$.

6. There are three aspects to the question 'Do we have robust measurements of PM_{2.5}?';
 - a) whether the AURN (Automated Urban and Rural Network) measurements meet the reporting requirements of the Directive. The data capture (the percentage of time for which data meeting the uncertainty requirement are available) currently (2009 and 2010) falls slightly short of the Directive requirements.
 - b) whether conclusions about small changes in concentration (typically smaller than the $\pm 25\%$ uncertainty required by the Directive) can be drawn from UK data. Data obtained using the same type of instrument and the same quality assurance/quality control (QA/QC) procedures are expected to be comparable with each other such that the relative uncertainty is significantly less than 25%. Relative uncertainties should be even less when longer term averages are taken, reducing the total impact of random variations. However, operational problems with the monitoring instruments on the UK network have made relative uncertainties hard to quantify.
 - c) whether the measurements are robust enough to improve our understanding of the sources of PM_{2.5} and PM_{2.5} precursors. At the current time, uncertainties in PM_{2.5} measurement data make them far from ideal for comparison with models. The lack of robustness is due to a combination of PM_{2.5} being defined operationally, the relatively large uncertainties in the measurement of PM_{2.5} concentrations and the inherent complexities of airborne particle formation and evolution.
7. The current analysers most widely used to measure PM_{2.5} (FDMS, Filter Dynamics Measurement System, see Chapter 2 for full details) are based on new and relatively complex technology. Significant work is required to fully understand and characterise this technology. In particular, it is recommended that issues concerning long-term reliability and the handling of semi-volatile components are further investigated.

I.1.1 Measurement recommendations

8. At present the measurement of PM_{2.5} remains a challenge, with current measurements falling just below the data capture requirements of the EU Air Quality Directive. **AQEG recommends a focused working group is assembled to address the achievement of Directive requirements using current measurement methods.**
9. High quality and comprehensive measurements of the chemical components of PM_{2.5} are an important future goal. Chemically-speciated measurements have twofold benefits, in terms of the attribution of composition of PM_{2.5} for control purposes and their ability to be used for source apportionment. To date, the measurements of some components of a non-volatile nature have a lower uncertainty than the overall mass concentration, but some methods still have

significant uncertainty. The area of measurement of the chemical components of PM_{2.5} needs review and the evidence requirements need clearly stating.

AQEG recommends a comprehensive review and rationalisation of the requirements for PM chemical composition measurements in the UK.

I.2 Concentrations and composition of PM_{2.5}

10. A new measurement network was established in 2009 for PM_{2.5} using reference equivalent instrumentation. Data are now becoming available from this network to evaluate PM_{2.5} distribution and legislative compliance. However, there is a requirement to define the important rural background contribution to PM_{2.5} across the UK. The relative lack of rural background sites, there are only three, restricts our ability to quantify sources of PM_{2.5}. Additionally, there are limited measurements of PM_{2.5} composition in different types of location and in different areas of the UK.
11. Evidence from urban sites and the limited number of rural background measurement sites indicates that regional (rural) background concentrations make a considerable contribution to the overall mass of PM_{2.5} in urban areas, accounting for around 60-80% of the background concentrations in the major urban areas of southern England. The regional background concentrations are dominated by secondary PM_{2.5}, primarily as ammonium nitrate and ammonium sulphate, but also as organic particles. In the central and southern UK around 60% of the urban background mass PM_{2.5} is made up of secondary particles. Sulphate particles remain important, despite the large reductions in sulphur dioxide emissions since the 1980s.
12. High PM_{2.5} concentrations are frequently associated with air transported into the UK from continental Europe. There is evidence that PM₁₀ episodes associated with air arriving from continental Europe, especially during the spring, are composed of fine particles (PM_{2.5}) and not coarse particles (PM_{2.5-10}), with nitrate playing a particularly important role. This nitrate is largely associated with ammonium, derived from ammonia emissions.
13. Emissions of the gaseous pollutants ammonia, oxides of nitrogen and sulphur dioxide from sources in the UK and Europe contribute to the formation of secondary PM over a large area. Black carbon is a major component of the PM_{2.5} associated with road traffic emissions and domestic (oil and solid fuel) combustion. Primary emissions from road traffic, including the non-exhaust component, make a significant (about 30-50%) contribution to the urban background increment of PM_{2.5} above rural concentrations. Road traffic can make substantial contributions to PM_{2.5} concentrations at the kerbside (within 1 m of the kerb) of around a third of total concentrations, but at the roadside (a few metres from the kerb) the contributions are more limited (~1-2 µg m⁻³ out of a total concentration of ~7-8 µg m⁻³ alongside busy roads) except in street canyons.
14. There is evidence to suggest that domestic and commercial sources make a contribution to concentrations of PM_{2.5} during the evening period, which may be due to solid fuel combustion and to particles released during cooking. Based on the limited evidence available, primary particles from industrial sources do not appear to make a major contribution to urban background concentrations.

I.2.1 Concentration and composition recommendations

15. Resources should be made available to ensure that the results from the PM_{2.5} monitoring network now in place are fully analysed to extract as much information as possible about the sources contributing to PM_{2.5} in different parts of the UK.
16. **The rural PM_{2.5} monitoring network should be expanded to allow a better quantification of the rural background.** This recommendation should be given urgent consideration. Additional sites will help verify modelled background concentrations and confirm the modelled spatial pattern of decreasing rural background concentrations from the south-east of England to the north-west of Scotland. They will also allow the urban increment to be determined more precisely.
17. Further work should be carried out to characterise the organic component of particles, in particular to improve our understanding of the contribution from secondary organic particles with respect to what is controllable under future policy measures.

I.3 Emissions and receptor modelling

18. The major sources of primary PM_{2.5} are combustion in the energy industries, road transport (both exhaust and non-exhaust emissions), off-road transport, residential sources and small-scale waste burning. Total PM_{2.5} emissions in the UK are predicted to decrease by 25% relative to 2009 levels by 2020, with an especially large contribution from reductions in road traffic exhaust emissions.
19. The main traffic sources of PM_{2.5} are exhaust emissions from diesel vehicles (cars, light goods vehicles and heavy goods vehicles), together with tyre wear, brake wear and road surface abrasion from all vehicles. A broadly similar picture prevails across the European Union. There are significant uncertainties attached to some of these emissions estimates and particularly to the estimates of PM_{2.5} from non-exhaust traffic sources. With reductions in exhaust emissions of PM, the non-exhaust components of traffic emissions will become much more important, emphasising the need to develop measures to control emissions from these sources.
20. With respect to PM_{2.5} secondary organic and inorganic precursor species, UK total emissions of nitrogen oxides (NO_x) have declined by 59% between 1990 and 2009 and are set to decline by a further 37% relative to 2009 levels by 2020. There is some uncertainty in inventories of road transport emissions for NO_x, with evidence to suggest that "real world" emissions of NO_x from Euro III-IV diesel vehicles are higher than previously thought and have not been declining as anticipated. While future road transport emissions of NO_x are expected to decrease, the rate of reduction may, therefore, not be as fast as current inventory projections indicate. UK emissions of sulphur dioxide (SO₂) and non-methane volatile organic compounds (NMVOCs) also fell between 1990 and 2009, by 89% and 70% respectively. Much smaller reductions of just 40% and 8% are anticipated between 2009 and 2020 for these two pollutants respectively. For ammonia (NH₃), UK emissions have fallen by only 21% between 1990 and 2009 and are not predicted to fall any further up to 2020. According to a recent estimate of past and future trends in EU-27 emissions of PM

precursors, the changes between 2005 and 2010 have been similar to those for the UK. The predicted changes in emissions between 2010 and 2020 for EU-27 are also similar to those predicted for the UK for NO_x, SO₂ and NH₃, although rather larger reductions are predicted for NMVOC emissions in EU-27 compared with the UK. Different changes in emissions would be expected where the contributions of different sources to precursor emissions vary between countries, but caution should also be taken when making comparisons because different emission factors may have been used in the inventories compiled for the UK and EU-27 countries.

21. Emissions from shipping are not well quantified. Emissions of SO₂ from shipping in Europe are predicted to decrease by just 3% in the next decade, although SO₂ emissions in Sulphur Emission Control Areas around the UK coast are expected to fall significantly. NO_x emissions from shipping in Europe are predicted to increase by 16% over the next decade.
22. Comparisons between the results of receptor and dispersion models have highlighted significant differences in relation to industrial/commercial/residential emissions of primary particles and the model predictions of secondary organic aerosol particles. Receptor modelling results highlight the weaknesses in current knowledge of a number of sources including wood smoke and cooking aerosol, and also suggest that the UK National Atmospheric Emissions Inventory (NAEI) emission factors for gas combustion may be rather high.
23. Use of carbon-14 as a tracer allows a distinction to be drawn between carbon derived from contemporary sources, such as wood burning or emissions from vegetation, and from fossil fuel sources. Analysis of carbon-14 in airborne particulate matter collected in Birmingham indicates a major contribution to secondary organic carbon from biogenic precursors.
24. Formulation of abatement strategies is made difficult by inadequacies in knowledge of the contribution of certain sources and weaknesses in understanding precursor–secondary particle dependencies for the major secondary components.

I.3.1 Emissions and receptor modelling recommendations

25. **AQEG recommends that the enhancement of emissions inventories is essential if numerical models of atmospheric PM_{2.5} are to be improved.** The key areas for improvement are:
 - non-exhaust vehicle emissions including tyre and brake wear, road abrasion and road dust resuspension;
 - fugitive dust emissions from construction, demolition, quarrying, mineral handling, industrial and agricultural processes, and methods for quantifying them nationally and locally;
 - PM_{2.5} emissions from domestic and commercial cooking;
 - PM_{2.5} emissions from small-scale waste burning and bonfires;

- PM_{2.5} emissions from domestic wood burning, accounting for the effectiveness of control measures;
 - biogenic emissions of NMVOCs;
 - emissions of NH₃ from agriculture, with their temporal as well as spatial variability;
 - exhaust emissions from off-road machinery used in construction and industry;
 - emissions of SO₂ and NO_x from shipping, in particular their spatial distribution around ports and harbours, the temporal variability and future emissions; and
 - exhaust emissions of PM_{2.5} from diesel vehicles under real world driving conditions and the factors and technologies affecting them.
26. Inventories should be developed to provide a quantification of the spatial and temporal variability in emissions of primary PM_{2.5} and its precursors from all contributing sources, including those not covered in national inventories, or provide the means for calculating them in air quality models. Developments should include spatially-gridded inventories with high resolution temporal profiles for different source sectors.
27. Further urgent research on the emissions and atmospheric chemistry of biogenic volatile organic compounds (VOCs) in the context of secondary organic aerosol formation in the UK is required, as this may have significant impact on the options for mitigation measures. Examples of critical areas have been recently evaluated.¹

I.4 Modelling and the future

28. Models are an important tool for understanding the links between emissions and observations data and making predictions of ambient concentrations in a self-consistent framework. **Modelling of PM_{2.5} remains a substantial challenge** owing to uncertainties in and lack of measured data, uncertainties/lack of understanding of some aspects of the dynamic, physical and chemical processes which need to be described within the models, and uncertainties in the emission data and their projections.
29. Several PM models covering urban to regional scales are used to predict UK air quality. They are based on a range of modelling systems (e.g. Eulerian, Lagrangian and Gaussian plume). Models are useful for quantifying the different contributions to PM, e.g. local urban emissions, and the contribution made by the long-range transport of pollutants.
30. Modelling results have shown that PM_{2.5} concentrations exhibit localised peaks in urban areas, owing to local sources of primary PM_{2.5}, superimposed on a regional background. These local sources are generally well represented by models, except when close to roads with complex street geometries. An

¹ EPRI and A&WMA Workshop on *Future Air Quality Model Development Needs*, 12-13 September 2011, Washington, D.C., USA.

important limiting factor in estimating concentrations and human exposure in urban areas is likely to be uncertainty in the emissions, including missing sources.

31. The largest contribution to PM_{2.5} concentrations overall is the secondary inorganic aerosol (SIA). The relatively slow formation of SIA (hours to days) means that concentrations tend to be smoothly distributed over large areas. Models show the nitrate component of SIA over the UK is now larger than that of sulphate, and is more variable in space and time, depending on the variability in ammonia emissions and concentrations owing to major reductions in sulphur dioxide (SO₂) emissions in the UK and elsewhere in Europe in recent decades.
32. The relatively small urban increment above regional background points to the need to consider control strategies for the regional background where secondary inorganic aerosol is by far the largest component according to models. Indeed, if nothing was being done to address the regional background the removal of the whole of the urban increment would be required to meet the targets set for PM_{2.5} exposure reduction.
33. Source apportionment from modelling shows how further reductions in SIA depend on control of emissions of SO₂, NO_x and NH₃ in other countries and from shipping, as well as from sources in the UK. **Modelling indicates a complex non-linear response of SIA concentrations to reductions in precursor emissions due to chemical interactions between pollutants, in particular the high dependence on the availability of NH₃, and the reversibility and temperature dependence of the formation of ammonium nitrate. The effectiveness of further SO₂ and NO_x reductions is uncertain given that overall emissions of NH₃ are likely to remain more constant as well as showing high spatial and temporal variation.**
34. Modelling of the secondary organic aerosol component, SOA, is uncertain both in terms of precursor emissions and chemical processes. It may be more difficult to control SOA and its precursors, of which biogenic emissions are a large component, than SIA. It is worth noting that the oxidants for the biogenic VOC precursors are all controlled by atmospheric chemistry and will respond to further reductions in anthropogenic emissions. It is not clear whether SOA levels will remain constant in the future if man-made emissions of VOCs and NO_x change significantly.
35. The semi-volatile components of organic aerosols and ammonium salts comprise a substantial fraction of PM_{2.5} and present a substantial modelling and measurement challenge. Other components, such as sea salt, rural and urban dusts, and water content, also need to be included in models in order to explain total PM_{2.5} concentrations and achieve mass closure. These components make a substantial contribution to overall PM_{2.5} concentrations.
36. **There is significant uncertainty in predicting PM_{2.5} concentrations into the future, owing to uncertainty in future European precursor emissions, particularly with respect to the secondary organic and secondary inorganic aerosol.**

I.4.1 Modelling recommendations

37. **AQEG recommends the more extensive and consistent evaluation of PM_{2.5} models in the UK**, considering for example, similar time periods and the speciated components of PM_{2.5}. Such an evaluation should also consider the temporal and spatial characteristics of the key components of PM_{2.5}. Furthermore, the evaluation would provide a more robust assessment of model performance beyond meeting the Air Quality Directive requirements for model performance. **Verification of models, particularly source attribution, remains challenging largely because of the lack of availability of chemically-speciated measurements.** There is also a need to develop methodologies for quantifying uncertainties in modelled values.
38. Further work on sea salt, rural/urban dusts and water content is required to investigate how they may be represented in modelling, including, for example, the response of the water content associated with the PM_{2.5} fraction to reductions in pollutant emissions.
39. With respect to mitigation, substantial further reductions in SO₂ emissions from 2006-2008 levels of around 50% across Europe will be required to achieve an appreciable reduction of about 1 µg m⁻³ in sulphate in PM_{2.5} in the southern UK. Similarly, a 1 µg m⁻³ reduction in nitrate would require a reduction of about 50% in European NO_x emissions from 2007 levels if SO₂ and NH₃ emissions remain constant. However, reductions of both SO₂ and NO_x emissions would necessitate a greater NO_x reduction to achieve the same improvement in nitrate, as reduced sulphate formation is accompanied by an increase in nitrate.
40. **Further consideration should be given to assessing the effectiveness of ammonia abatement as a way of reducing UK secondary inorganic PM_{2.5}**, in concert with any requirement for wider transboundary abatement.
41. Modelling of the secondary organic aerosol component of PM_{2.5} is uncertain both in terms of precursor emissions and chemical processes; further work is required in this area, in particular on the fundamentals of the mechanisms of formation and incorporation of precursors into the aerosol.