AIR QUALITY EXPERT GROUP

New Opportunities for Particulate Measurements

Prepared for:

Department for Environment, Food and Rural Affairs;
Scottish Government; Welsh Government;
and Department of Agriculture, Environment and Rural Affairs in Northern Ireland

This is a report from the Air Quality Expert Group to the Department for Environment, Food and Rural Affairs; Scottish Government; Welsh Government; and Department of Agriculture, Environment and Rural Affairs in Northern Ireland, on scientific and technical opportunities for the measurement of airborne particulate matter. The information contained within this report represents a review of the understanding and evidence available at the time of writing.

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Terms of Reference

The Air Quality Expert Group (AQEG) is an expert committee of the Department for Environment, Food and Rural Affairs (Defra) and considers current knowledge on air pollution and provides advice on such things as the levels, sources and characteristics of air pollutants in the UK. AQEG reports to Defra's Chief Scientific Adviser, Defra Ministers, Scottish Ministers, the Welsh Government and the Department of Agriculture, Environment and Rural Affairs in Northern Ireland (the Government and devolved administrations). Members of the Group are drawn from those with a proven track record in the fields of air pollution research and practice.

AQEG's functions are to:

1. Provide advice to, and work collaboratively with, officials and key office holders in Defra and the devolved administrations, other delivery partners and public bodies, and EU and international technical expert groups;
2. Report to Defra's Chief Scientific Adviser (CSA): Chairs of expert committees will meet annually with the CSA, and will provide an annual summary of the work of the Committee to the Science Advisory Council (SAC) for Defra's Annual Report. In exception, matters can be escalated to Ministers;
3. Support the CSA as appropriate during emergencies;
4. Contribute to developing the air quality evidence base by analysing, interpreting and synthesising evidence;
5. Provide judgements on the quality and relevance of the evidence base;
6. Suggest priority areas for future work, and advise on Defra's implementation of the air quality evidence plan (or equivalent);
7. Give advice on current and future levels, trends, sources and characteristics of air pollutants in the UK;
8. Provide independent advice and operate in line with the Government's Principles for Scientific Advice and the Code of Practice for Scientific Advisory Committees (CoPSAC).

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Further information on AQEG can be found on the Group's website at:
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New Opportunities For Particulate Measurements.

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Executive summary

Airborne particulate measurement science is a substantial area of both academic and commercial research with new tools and techniques emerging all the time. It is necessary therefore to regularly reassess where new measurement techniques might be able to provide additional insight to support national and local management of PM in the UK. This report is not an exhaustive list of available techniques, but instead provides some descriptions of key methods and new opportunities that may be of relevance to local and national government. It is focused on proven techniques that are available for operational use in air quality and emissions measurement and does not cover emerging technologies that are currently only used within the research community.

Most measurements currently commissioned by Defra, Devolved Administrations and local authorities are associated with the measurement of the mass concentration of particles smaller than 2.5 micrometres diameter ($PM_{2.5}$) and 10 micrometres (PM_{10}). These extensive (and growing) networks of monitors provide the evidence for attainment of ambient air quality standards. These will continue to be needed in the coming decades and will provide the primary indicator of progress towards targets set in the Environment Act (2021). The challenges of measuring $PM_{2.5}$ in a consistent and reproducible manner, and at concentrations below the recently updated target annual average limit value of 10 $\mu g\ m^{-3}$ should not be underestimated; continued investment in monitoring, calibration and performance evaluation are essential.

Whilst $PM_{2.5}$ will continue to be the primary health–relevant air quality metric that is measured in the UK, evidence has strengthened around the role played by smaller particles in causing harm (sub-micrometre and nanometre scale) . Further expansion of long-term urban measurements of particle number and ultrafine particles, and more broadly the size distributions of particles in UK air, would provide an opportunity to understand more about their sources and to support future epidemiological research. The inclusion of particle number and ultrafine particles in tailpipe emission standards for transport sources (e.g. in Euro 6 and proposed Euro 7 standards for road vehicles and proposed international standards for aviation) will further increase the necessity for ambient monitoring.

Measurement of the chemical composition of particles continues to be an area of intensive academic study, but it has been used to only a limited extent by policymakers and regulators. Growth in technological capabilities, hardware, software and data analytics, have created opportunities for more detailed information of this kind to inform on individual PM sources and their changes over time. Such information can provide insight into the progress of individual sectors in managing emissions. Such measurements can assess the effectiveness of interventions to reduce primary PM and precursor emissions of secondary PM from sources including woodburning, agriculture and road transport. The composition of PM also provides rich information that can be used to test and improve the performance of models, enabling better short-term forecasts of pollution and models used for assessments of the impacts of future emission scenarios. More detailed knowledge of the types of PM emitted from individual sources, such as from tyre and brake wear, from the tailpipe, from industry and so on, can enhance the accuracy of emissions inventories, improve international reporting and in turn improve model performance.

Whilst the vast majority of existing PM monitoring is conducted long-term and *in situ* at fixed measurement sites, a range of other techniques are available that can give additional insight into source strengths and the spatial distribution of pollution. Compact sensors provide an opportunity to expand the number of indicative PM monitors within a community, including sensors carried by people, used by community groups, and sensors designed for indoor spaces. Earth observation satellites can now provide a national-scale picture of the spatial distribution of PM across the UK. These alternatives to fixed monitoring cabinets may help identify previously unidentified sources, find hotspots and support decision-making during unplanned events such as accidents or wildfires. These can provide significant complementary information that adds value to existing *in situ* measurements from networks such as the AURN. There are further opportunities to use shorter more focused periods of intensive research measurements to provide evaluation of models and emissions estimates, and to test process knowledge of the PM lifecycle.

The detailed information on the nature of PM pollution that can be derived from some techniques provides an opportunity for enhanced communication and information systems for the public. Realtime (or near real-time) information on PM pollution and its contributory sources during poor air quality events may help support behavioural adaptations and evidence the need for interventions and action. The co-location of multiple analytical techniques in one location, disaggregating PM into its many sub-components is a now proven powerful approach that delivers a scientific and evidential value that is greater than the sum of the parts. PM measurements should be viewed by government as a key enabler for effective air quality management, as well as a means for demonstrating regulatory compliance.

As measurement science advances it has been possible to gain insight into previously undetected sub-types of PM. For example, in recent years it has been demonstrated in research studies that it is possible to directly observe the presence of individual metals and the microplastic components within respirable PM. It is now possible to routinely identify different types of airborne bioaerosols in real-time. Offline laboratory techniques are giving ever increasing insight into the molecular and biological composition of PM, broadening the boundaries of what should be considered of in the context of air quality and health. For emerging classes of PM, particularly those involving microplastics and bioaerosols, there is a pressing need for investment in the development of standardised methodologies and reference materials to support future national scale monitoring. This is a key step in bridging between short-term research studies and long-term measurements. The field of PM measurement has also benefited from continual advancements in remote sensing, in particular observations from the latest generations of satellite instruments and their data processing methods. When combined with models of the atmosphere and measurements made at the surface, these new data products offer opportunities to study the spatial extent of atmospheric PM and track plumes from large pollution events.

Emission inventories report annual mass emissions of PM from different sources and include information on different particle sizes (PM_{10}, $PM_{2.5}$) using emission rates measured at source or activity-based emission factors (mass emissions per unit of activity). A comparison of emissions from different sources indicated by inventories can be hampered by the different measurement techniques that are used, e.g. for combustion point sources, residential combustion, transport sources. Different measurement techniques and approaches do not

always determine equivalent PM (sometimes for technical or standardisation reasons) and may not necessarily represent emissions in real-world conditions. Consequently, reported emissions for some sectors may not be equivalent to emissions reported for other sectors and may further differ from the definition of PM measured in ambient air.

There are established measurement techniques for large industry sources but their suitability to cope with ever-reducing emission concentrations is becoming an issue both for periodic sampling methods and monitoring systems used for compliance assessment. There may therefore be a requirement for renewed investment in new methods of detection to support industrial PM emissions control. The measurement of real-world exhaust emissions of PM from the latest generation of road vehicles is also becoming increasingly challenging as emissions have reduced to meet tighter regulations. The quantification of non-exhaust PM from tyre and brake wear at source and under real-world conditions remains a particular challenge due to high variability and the need to establish a realistic test procedure. Emission limits and test procedures for brake wear emissions have recently been proposed within the Euro 7 regulations, recognising the increasingly important contribution these sources make to emissions and local PM concentrations in urban areas.

As PM datasets become more extensive and complex the role played by data infrastructure and data methods grows. Extracting maximum value from investment in measurements requires sustained efforts to support innovation in use of data techniques alongside resources for interpretation. The opportunities however are very significant. An expanded role and scope for PM monitoring in the UK is likely to increase the evidence available to demonstrate the effectiveness of air pollution policy and technical interventions, provide more dynamic opportunities to use measurements to actively control pollution and increase public engagement with air quality more generally.

Opportunities

1. The rapid pace of technological development in particle and aerosol measurements means that Defra should regularly review and reassess national monitoring activities to ensure they are using the most appropriate technologies whilst maintaining consistency in the reported data. $PM_{2.5}$ and PM_{10} mass concentration will continue to be critical health-relevant metrics of PM pollution however observational networks and monitors in the UK should adapt over time to reflect strengthening evidence arising from other metrics including particle number, ultrafine particles and black carbon, as recommended also in the 2021 WHO air quality guidelines. **Long-term measurements of an expanded range of PM metrics can provide enhanced opportunities and insight to future epidemiological studies**.

2. Over recent decades, reductions in emissions of primary PM and secondary precursors such as sulphur dioxide, have led to a significant reduction in ambient concentrations in the UK. Annual average concentrations are in many places already below the Defra 2040 target of 10 $\mu g\ m^{-3}$ and in some more remote parts of the UK below the WHO recommended target of 5 $\mu g\ m^{-3}$. The current Population Exposure Reduction Target (PERT) aims for continual improvement, but detecting small changes year-on-year is towards the limits of capabilities of current monitoring technologies; there are growing challenges in obtaining consistent

measures of $PM_{2.5}$ across monitors that use different underlying analytical principles. **Investment in new calibration technologies and data science provides an opportunity to improve data quality, in turn reducing uncertainties associated with the assessment of air quality Limit Values and Population Exposure Reduction Targets.**

3. Observations made by Defra, Devolved Administrations and local government are often directed towards providing evidence for attainment of air quality standards. Other measurements might inform the real-time control of polluting activities, such as at construction sites, or inform the public on their exposure. Only a small fraction of PM monitoring investment nationally is designed to meet other evidential needs. Information on the composition and sources of PM can provide more direct insight into the contributions made by individual emission sectors and for policy and technical interventions in these sectors to be evaluated over time. **There are substantial opportunities for Defra to re-frame PM measurements as a more central tool in air pollution management to go beyond compliance assessment to understand sources and improve information to the public.** The ability to rapidly communicate information to better control either emissions or exposure provides significant opportunities. The capability to provide near-real time information on aspects of pollution beyond simply mass concentrations should also be explored, including for public communication.

4. A critical consideration for any expansion or change to monitoring of PM is that the information to be generated by a technique, or combination of techniques, is well-matched to the intended objective. Policymakers and practitioners should consider whether the data is to test compliance, quantify sources, track changes in emissions/concentrations or some combination of these. Whilst inevitably attention often focuses on the hardware element of a measurement, data quality and processing techniques are just as important. **The evidence value of routine monitoring can be increased by designing objectives, measurement techniques and data handling in combination, and also help support serendipitous discoveries.**

5. The co-location of online, multi-parameter measurements that expand on regulated pollutants delivers significant insight into source attribution and for detecting changes in emission contributions over time. Methods for performing measurements of this type are at various stages of development, with some now widely accepted. Techniques for evaluating the contribution of currently uncertain $PM_{2.5}$ sources such as non-exhaust vehicle emissions (brake/tyre wear, road dust), cooking and wood burning are now emerging. **The comprehensively instrumented air pollution research 'supersites' in the UK provide a major opportunity to directly deliver evidence of the effectiveness of Defra's PM reduction strategies out to 2040 and are aligned with developing international best-practice.** There are increasing blurred lines between what might be thought of as 'research' and what is 'monitoring' at UK air quality supersites. Defra, UKRI, Universities and other stakeholder agencies should work closely together to ensure such nationally critical activities are suitably supported, including development and evaluation of new methodologies for quantifying the influence of different emission sources.

6. Lower power and compact methods for the measurement of PM have emerged over the last two decades. Small optical particle counters are now commonplace and often have their data transformed through algorithms into a $PM_{2.5}$ or PM_{10} metric. New standards and test

procedures are emerging for sensors of this kind. Whilst many technical issues remain outstanding, and such devices are likely to only ever provide an approximate measure of PM, their potential to generate new insight through both wearable and networked, distributed measurements are clearly very high. **PM sensors have been adopted already by community projects focusing on air quality and offer opportunities to support public awareness, engagement and action**. Small sensors allow for estimates of personal exposure to be made when carried on a person and support 'gap filling' in some observational networks. They can also be combined with models of different indoor and outdoor micro-environments and be used to study indoor sources. There is a continuing need however for government to provide technical advice to individuals, community groups and local authorities wanting to use PM sensors to ensure that their use is suitably matched to the objectives of the study.

7. It is possible to derive an estimate of the spatial distribution and abundance of PM from space-based instruments and these capabilities have expanded as new satellites have been launched and data retrieval methods improved. Thus far the use of Earth Observation (EO) data on PM has been restricted largely to the research community. **Earth Observation data on PM can become a valuable resource for government, supporting the detection of unknown sources, identifying discrepancies with inventories and aiding assessment of PM pollution from unanticipated events such as industrial accidents, dust storms, volcanic eruptions or moorland/forest fires.** The inclusion of EO data into the public communication of air pollution effects is an opportunity for Defra and others to consider.

8. Analytical instruments provide only part of a PM evidence system and suitable attention must be given to data techniques. Statistical methods such as principal component analysis and factor analysis have long supported source attribution of PM. **There are many new opportunities in air quality science emerging from the use of artificial intelligence and machine learning, including improving data quality, detecting the effects of policy interventions and predicting future concentrations.** Increased complexity and richness of PM monitoring datasets create more demanding requirements for data curation, accessibility and ease of re-use.Defra' s PM observational network should therefore incorporate a data strategy to ensure maximum value is extracted from monitoring investments.

9. New measurement techniques can provide substantial improvements to emissions inventories by providing more representative or more chemically detailed assessments of PM emissions at source. **Innovative analytical methods provide an opportunity to quantify real-world PM emissions from hard-to-measure sources such a brake and tyre wear, from trucks and buses, from solid fuel combustion, and sources not currently including in the NAEI such as cooking.** Additional PM metrics such as UFP, BC and PN are being included in some future European emission standards. Investment is needed to ensure there is national capability to apply these techniques in a UK context, with benefits for Defra not only in terms of accuracy of emissions reporting, but better model forecasts that rely on correctly representing emissions.

10. Linked to previous recommendations are the opportunities that arise for improved modelling and forecasting systems given more readily available detailed and granular

information on emissions and ambient PM. Whilst mass concentrations of PM can be reasonably well represented in models, often due to non-mechanistic adjustments, it remains a major scientific challenge to correctly simulate PM components. **Information on PM composition can provide significant opportunities for advancement of air pollution models, providing tests of performance, ground-truth of emission sources and support for model parameterisation**. Taken more broadly the ingestion of increasingly diverse PM information sources, including *in situ*, sensors and EO may provide more confidence in air quality model predictions for hazard warnings and for policy scenarios.

11. Dissemination of public information on ambient PM is limited to $PM_{2.5}$ and PM_{10} and typically only becomes prominent in the media during severe air pollution events. Currently the emphasis are warnings to vulnerable individuals. A more developed system for public information on air pollution should consider also messaging those actions that would be beneficial to reduce emissions. **Increased information on PM composition during pollution events would provide an opportunity to better communicate why an event is occurring and want actions to reduce emissions might be most beneficial.** There have been anecdotal examples in recent years of chemical speciation from research supersites gaining prominence in media reporting of pollution events. The opportunities from new data sources and instruments as tools for public communication should be considered within the Defra Air Quality Information System (AQIS) review.

12. In recent years AQEG has broadened the range of pollutants that it has provided advice on to include pollutants and emissions occurring in indoor settings and from natural sources. In part this has arisen because of new insight in the scientific literature, often as a result of new observations. In this regard new analytical technologies for PM drive the science of air quality. Two emerging areas covered in this report are techniques for detecting airborne microplastics (from a range of sources including tyre wear) and discriminating bioaerosols arising from the growth of microbes such as fungi and transmission of airborne pathogens. **Further investment would enable translation of research techniques into standardised methods appropriate for routine monitoring, and to provide better insight to Defra, DHSC and DLUHC on the scale and scope of emerging pollutants.**

13. The focus in this report is predominantly on techniques that may be deployed to enhance information as part of long-term monitoring activities. The UK has however a rich history of collaborative short-term experiments that have provided vital evidence for policy and delivered a comprehensive stock-take on the state of the UK atmosphere and airborne particulate matter. This approach to air quality science remains a key party of the evidence landscape. **Short-term intensive periods of measurement offer major opportunities for better insight into PM emissions, current and emerging types of PM pollutants and secondary atmospheric processes.** These can bring together Defra, and Local Authority monitoring with the latest PM techniques from across the research community. Defra has a vital role in articulating these science and evidence needs to other key funders such as UKRI.

Chapter 1 - Introduction

1.1 Context, scope, and definitions

Particulate matter (PM) is currently the single most significant component of air pollution in the UK that gives rise to human health impacts. A growing body of research indicates that there is no 'safe' lower limit for exposure. The World Health Organisation (WHO, 2021) identified that negative health effects of $PM_{2.5}$ could be demonstrated at annual average concentrations above 5 µg m^{-3} and recommended that countries should strive to continuously reduce concentrations. WHO identified that reaching 5 µg m^{-3} would bring significant public health and economic benefits, however achieving this would be challenging and indeed may not be feasible in some countries and locations. Many of the sources that dominated PM emissions in the past have already been abated or policies exist to further reduce emissions, for example through the phase out of coal fired power stations and progressively more stringent controls on vehicle exhaust emissions.

Further reducing PM today is complex because it is emitted directly from a wide variety of diffuse sources (including combustion, friction and cooking), as well as being formed through multiple different atmospheric chemical reactions. PM consequently is composed of a highly complex mixture of different chemical compounds. The majority of PM away from the immediate vicinity of direct sources tends to be secondary in nature, i.e. formed in the atmosphere through chemical processes. Furthermore, PM exists in a wide variety of sizes and shapes that may influence their behaviour in the atmosphere and toxicity when inhaled. For instance, the so-called 'ultrafine' particulate (UFP), i.e. particles smaller than 100 nm in diameter, are hypothesised to cause more physiological damage per unit mass because of their ability to penetrate into the bloodstream and nervous system.

Air quality regulation and management is highly dependent on ambient measurements since predicting both the amount of PM and its constituent chemical parts is very difficult using atmospheric models. There have been widespread measurements of PM_{10} in the UK since the 1990s with the smaller size metric of $PM_{2.5}$ being added to UK networks from around ~2000 onwards. These ambient measurements have formed the basis for demonstration of regulatory compliance (or otherwise) with EU Air Quality Directives. Post Brexit, those existing measurement networks will support evaluation of new air quality standards for $PM_{2.5}$ set out in the Environment Act (2021) and The Environmental Targets (Fine Particulate Matter) (England) Regulations (2023).

Most instruments used to routinely measure $PM_{2.5}$ in Defra and Local Authority networks report only the mass concentration, and this data says nothing about the chemical nature of those particles, or their origins. Additional sources of information on PM chemistry, size and shape are required to apportion PM back to originating sources, to inform on differential health impacts, validate model forecasts of PM and evaluate the effectiveness of past policies and interventions. Due to climate change and the continuous changes in pollution emissions in response to technology and legislation the properties of particles evolve over time. However, these changes only become visible when detailed measurements of the chemical and physical nature of PM are made. A notable policy and hence measurement

challenge for the future is the likely growth in significance of secondary aerosols which are subject to transboundary transport and that have complex chemical formation pathways.

There has been impressive development of new analytical technology for the study of PM in recent decades, and new sources of data are coming available that are viable for routine and long-term monitoring. The purpose of this report is to provide a briefing on current technologies for measurement of PM that include traditional *in situ* measurements of mass concentration and more detailed properties relating to chemistry, morphology and toxicology. The report reviews 'low cost' or compact sensors that may be used for enhanced air pollution networks, or for community/citizen science applications, as well as remote sensing / satellite data products and associated machine learning techniques. It also reviews approaches applicable to assessing PM pollution in indoor settings, including for biological aerosols. Techniques for quantifying the amount and characteristics of PM emissions at source are reviewed, alongside technologies that may be needed to evaluate emerging sources of concern within PM pollution including bioaerosols and microplastics.

Examples will be given illustrating the use of individual technologies and their potential applications. The scope of this report is to cover current and emerging technologies suitable for general routine and long-term applications, as might be applied by users in Defra, the Devolved Administrations or Local Authorities in support of their evidence needs. The report does not aim to review all 'research grade' technology that is available in the wider atmospheric science community.

The following definitions are used throughout this document:

PM Particulate Matter, within the Environmental Targets (Fine Particulate Matter) (England) Regulations 2023 this is defined as "*anything in the air which is not a gas*".

$PM_{2.5}$ PM contained within particles smaller than 2.5 μm (aerodynamic diameter)

PM_{10} PM contained within particle smaller than 10 μm (aerodynamic diameter)

BioPM PM with a biological origin

TSP Total Suspended Particulate, PM of any size

UFP Ultrafine Particulate, particles smaller than 100 nm, normally measured as a number concentration in cm^{-3}

BC Black Carbon, a measure of light absorbing material in PM (notionally similar to EC)

EC Elemental Carbon, particulate carbon in the form of refractory graphitic carbon

OC Organic Carbon, particulate carbon in the form of organic molecules

OM Organic Matter, particulate matter in the form of organic molecules (including elements beyond carbon, such as oxygen and hydrogen)

SIA Secondary Inorganic Aerosol, inorganic PM produced through atmospheric chemistry

SOA Secondary Organic Aerosol, organic PM produced through atmospheric chemistry

POA Primary Organic Aerosol, organic PM emitted directly into the atmosphere

NEE Non-Exhaust Emissions, emissions from vehicles, but not from the tailpipe, such as brake or tyre wear, or resuspended road dust

1.2 Questions that future measurements should address

Policy-relevant questions

PM monitoring is needed to inform policy development, so that interventions are focused and achieve maximum benefit and can be effectively evaluated. Some fundamental scientific questions remain outstanding and continue to be a challenge. Addressing them could provide real beneficial impact through the policy development process. Relevant questions that monitoring could help to address include:

- What are the sources of ambient PM?
- How much does each source contribute?
- What sources / components of PM are most harmful to human health?
- How do PM concentrations vary spatially and temporally (including in microenvironments and indoors)?
- How will climate change affect the spatial and temporal patterns of ambient PM concentration (eg by influencing secondary PM formation)
- Are the sources/components of ambient PM changing?
- Have interventions intended to mitigate emissions from PM sources been successful?
- In which environments are people (most) exposed to, or more at risk of exposure to, PM?
- How much PM are individuals exposed to, and from what sources?
- What interventions would achieve most health benefit?
- Are there sources which could be tackled in a way which alleviates disparities, particularly with respect to vulnerable groups, groups holding protected characteristics or from lower socio-economic groups?
- What monitoring would be required to build an evidence base for disparity-reduction measures?

Questions for AQEG to address

Different PM measurement techniques may be better suited to some uses than others, and hence to addressing different questions. New and novel approaches are becoming available, in addition to the well-established methods, providing opportunities to enhance our understanding beyond a compliance-based measurement network. AQEG is therefore asked to consider questions such as:

i. What techniques/technologies for monitoring PM concentrations are currently available?
ii. What are the benefits and limitations of each of the techniques?
iii. What type of use are each of the available technologies best suited to?
iv. Which of the available techniques is/are best suited to addressing each question?
v. To what extent can the questions above be addressed using the available techniques?
vi. What recommendations would AQEG make for additional and/or complementary monitoring or measurement techniques that relevant funding bodies (for example, Defra, Devolved Administrations, the Environment Agency, Local Authorities, Research Councils or others) could consider to address policy-relevant questions, included those listed above?

vii. What further developments in available techniques are expected and/or needed to answer policy relevant questions and improve the impact of interventions?

Previous COMEAP recommendations

COMEAP's Statement on the differential toxicity of particulate matter according to source or constituents: 2022 - GOV.UK (www.gov.uk) suggests monitoring that is needed to support the types of epidemiological studies which would provide information regarding the types/sources of airborne PM which are particularly detrimental to health.

Monitoring needs identified by COMEAP

Monitoring strategies for airborne particles should account for the needs of different types of epidemiological study, including factors such as temporal and spatial variation of particle components, and relationships between outdoor and indoor microenvironment concentrations. One of the main limitations on research in the UK is the paucity of consistent long-term data sets of speciated chemical composition for airborne particles, and the establishment of a suitable measurement programme should be considered. In view of the uncertainty regarding the health impacts of ultrafine particles, and the recent WHO recommendations, expansion of the currently very limited monitoring of UFPs in the UK is strongly advised.

Future changes in emissions should also be reflected in decisions to alter or upgrade monitoring networks and in the health research conducted. For example, in regard to road traffic, the adoption of electric vehicles will have an impact on the constituents and size distribution of particles that are associated with vehicles. Insight into the potential consequences of these changes on health will be gained as progress is made in our understanding of the exposure to non-exhaust traffic-derived particles (for example, PM from brake, tyre and road wear) and their relative toxicity. The transition to Net Zero will also lead to further changes in the composition of airborne particles. There remains some uncertainty as to the precise directions of change, but declines in sulphate and nitrate particles seem likely, with a possible increase in biogenic secondary organic aerosol. Changes in particle composition may present an opportunity to evaluate effects of differing components upon health.

Studies in which personal monitoring of exposures includes specific particle components should be carefully considered. These have great potential but are typically limited by small sample size and hence low statistical power. There is a need to establish a greater consensus regarding the appropriate practice for research with personal monitors, in terms of monitor type, monitoring protocol, alignment with health parameters measured and data analysis methods.

Epidemiological methods – recommendations for further research

Additional large-scale epidemiological studies incorporating high quality source apportionment by receptor modelling or dispersion modelling are warranted, especially in relation to less well-studied sources of PM (see below). Studies of the effect of temporal changes in particle composition upon exposure-response coefficients in epidemiological studies may yield valuable insights. Innovative designs of epidemiological study capable of distinguishing the effects of correlated exposures are strongly encouraged. As different

particle components appear to be linked to different mechanistic pathways, the study of health outcomes other than all-cause mortality in conjunction with particle speciation may provide useful insights into effects of differing particle types.

Also, there is a need to evaluate the relationship between pollutant exposures and early markers of disease in longitudinal cohorts in order to address the contribution of air pollution to the development of non-communicable diseases.

Further clarity may be obtained from systematic reviews and meta-analyses of studies that make comparisons between specific PM sizes or constituents and health-related outcomes, for example. PM_{10} and $PM_{2.5}$, $PM_{2.5}$ and UFP metrics (such as particle number concentration) or $PM_{2.5}$ and BC/EC.

In recognition of the high relative surface area of UFPs and increasing evidence that these particles gain access to the blood and systemic organs these particles warrant continued attention. In particular more research is required on the source and composition of UFPs, exposure levels and how exposure to these may impact on different organ systems and, accordingly, population groups with different risk factors and health conditions.

More research is needed on sources of PM for which the potential effects on health are less well studied, including indoor PM, non-exhaust traffic-derived PM, micro- or nano-plastics, PM from burning wood, PM-derived from agricultural emissions, and PM from shipping and aviation. The need for research into the health-related effects of these emissions aligns with the UK Clean Air Strategy (2019). The risks associated with PM from wildfires also represents a pressing research need given the increasing prevalence of wildfire events internationally. Toxicological studies directly comparing PM from these sources to that of more established sources would be beneficial (for example, Selley et al., 2020).

Chapter 2 - Technology state of the art: *In situ* instruments

Measurements made in ambient air have been a mainstay of evidence used for regulation since the original Clean Air Act. Originally particulate matter measurements were made according to a black smoke metric. Over time this was replaced with use of measurements against the PM_{10} and $PM_{2.5}$ metrics, and these have formed the basis for modern epidemiological research and air quality compliance as part of the Automatic Urban and Rural Network (AURN). More recently, there has been some emphasis placed also on the measurement of the composition and number concentrations of particles to support evidence on emissions but also as candidates for use as additional health metrics. The WHO (2021) air quality guidelines, while stopping short of recommending ambient targets for black carbon (BC) and ultrafine particulates (UFP), did recommend an expansion of monitoring capacity for these. Other on- and off-line measurements of composition also exist.

Expanded measurements of PM, and real-time data, form a major part of the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) network of monitoring sites across Europe. At the time of writing these are proposed to be used in support of a revised European Union Air Quality Directive. These measurements are, again at the time of writing, also being replicated in the newly created Atmospheric Science and Chemistry. mEasurement NeTwork (ASCENT) in the USA. In the UK, some of these measurements have formed part of the Particle Composition Network (PCN) and Black Carbon monitoring networks, however these have tended to be limited to specific sites, such as in central London and the EMEP supersites. There have also been measurements by the research community, in particular at the NERC air quality supersites in London, Manchester and Birmingham.

Highly detailed measurement of PM can also be delivered through short-term ‘intensive’ research measurement campaigns. Here a large number of complementary measurements can be performed simultaneously to comprehensively assess the composition, sources and properties of airborne PM. This can also utilise mobile measurement facilities used by the research community. A UK-based example of this is the ClearfLo campaign that took place in and around London in 2012 (Bohnenstengel et al., 2015), which allowed for a multi-site assessment of the sources of PM, using multiple independent techniques (Visser et al., 2015; Yin et al., 2015; Young et al., 2015; Liu et al., 2014). More recently, the NERC OSCA project has taken place in London, Manchester and Birmingham with intensive measurements taking place in 2021-22, but at the time of writing analysis is ongoing.

2.1 Regulatory $PM_{2.5}$/PM_{10} Techniques

While there are many interesting and insightful aspects of PM that can be measured including particle size, shape and chemical composition, the primary measure of health-impacting airborne particular matter in UK legislation remains $PM_{2.5}$ and PM_{10}. This is section summarises the current state of the art for their measurement.

Current air quality standards of relevance to $PM2_{.5}$ and PM_{10} are set out in Table 2.1 below:

Table 2.1 UK Air Quality Objectives, Limit Values, and Targets for PM_{10} and $PM_{2.5}$ Concentrations

Pollutant	Description	Measured as
Local Air Quality Management Objectives		
PM_{10}	40 µg m^{-3} (18 µg m^{-3} in Scotland) [i]	Annual mean
	50 µg m^{-3} not to be exceeded more than 35 times/ year (7 times a year in Scotland) [i]	24-hour mean
$PM_{2.5}$	Local authorities are required to work towards reducing $PM_{2.5}$ [ii]	
	10 µg m^{-3} in Scotland [iii]	Annual mean
UK Limit Values		
PM_{10}	40 µg m^{-3} [iv]	Annual mean
	50 µg m^{-3} not to be exceeded more than 35 times/ year [iv]	24-hour mean
$PM_{2.5}$	20 µg m^{-3} [iv,v]	Annual mean
	Target of 20% reduction in concentrations at urban background between 2010 and 2020 [iv]	
Environmental Targets for England		
$PM_{2.5}$	12 µg m^{-3} by January 2028 (interim target) [vi]	Annual mean
	10 µg m^{-3} by 2040 (legally binding target) [vii]	
$PM_{2.5}$ population exposure	22% reduction in population exposure between 2018 and January 2028 (interim target) [vi]	
	35% reduction in population exposure between 2018 and 2040 (legally binding target) [vii]	

[i] Set in The Air Quality (England/Wales/Scotland/Northern Ireland) Regulations 2000 and the Air Quality (Scotland) Amendment Regulations 2002
[ii] Defined in Local Air Quality Management Technical Guidance 2022
[iii] Set in The Air Quality (Scotland) Amendment Regulations 2016
[iv] Set in The Air Quality Standards (+ Wales/Scotland/Northern Ireland) Regulations 2010
[v] Amended by The Environment (Miscellaneous Amendments) (EU Exit) Regulations 2020
[vi] Defined in the Environmental Improvement Plan 2023
[vii] Set in The Environmental Targets (Fine Particulate Matter) (England) Regulations 2023

Since PM_{10} and $PM_{2.5}$ size fractions of particles are key metrics used in air quality regulations and legislation, but they are not pure chemicals like NO_2 or O_3, their physical definition is critical, since this provides a common starting point for evaluation of instrument performance and standardisation. Specific definitions for both pollutants are set out by the International Standards Organisation:

PM_{10} – particles which pass through a size-selective inlet with a 50 % efficiency cut-off at 10 µm aerodynamic diameter. PM_{10} corresponds to the “Thoracic convention” as defined in ISO 7708:1995, Clause 6.

$PM_{2.5}$ – particles which pass through a size-selective inlet with a 50 % efficiency cut-off at 2.5 µm aerodynamic diameter. $PM_{2.5}$ corresponds to the “High-risk respirable convention” as defined in ISO 7708:1995, 7.1.

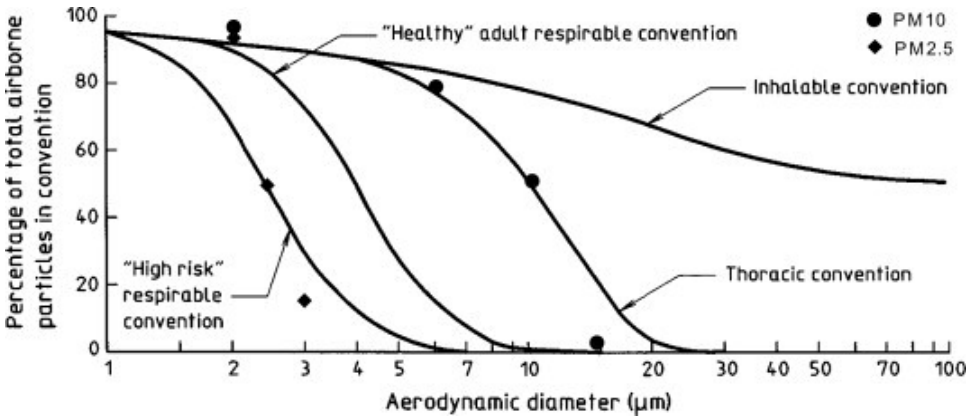

Figure 2.1 Inhalable, thoracic, respirable, and high risk respirable penetration curves defined in ISO 7708 (Cambra-Lopex et al. 2009) as percentage of total airborne particles, and PM_{10} and $PM_{2.5}$.

Regulatory measurement methods are designed to operate with a size selective inlet in order to achieve the requirements of these ISO standards.

For regulatory (compliance) reporting of PM_{10} and $PM_{2.5}$ the UK has followed the requirements of the Air Quality Directive (2008/50/EC)) through the Air Quality Standards Regulations 2010. This sets out how standard methods of measurement should be implemented by those competent authorities identified for reporting air quality data across Europe. For PM_{10} and $PM_{2.5}$ the relevant reference method is the standard gravimetric method as set out in BS EN12341: 2014. This standard is an evolution of the earlier individual standards for PM10 (BS EN12341: 1998) and for $PM_{2.5}$ (BS EN 14907:2005).

This method of sampling of particulate matter in EN 12341: 2014 is filter-based, i.e., capturing the particulate matter on filters and weighing them by means of a balance. Size fraction differentiation is achieved through a design modification to the sampling inlet of the reference instrument to align to the particulate size cut-off operating at a nominal flow rate of 2.3 m^3 h^{-1} over a sampling period of 24 hours. Filters are pre- and post-weighed under standardised temperature and relative humidity conditions to avoid artefacts associated with changing contributions of particle-bound water during the weighing process. Measurements (in µg m^{-3}) are reported against ambient conditions near the sampling inlet at the time of sampling.

Provision of gravimetric PM_{10} and $PM_{2.5}$ measurements is very expensive and time consuming and gives rise to delay in the availability of information to the public; one of the key aspects of the compliance monitoring programmes undertaken in the UK. Adoption of non-gravimetric methods is permitted for the measurement of PM_{10} and $PM_{2.5}$. Non reference methods are referred to as “Candidate” methods – and to be fully adopted into a compliance network each Candidate method must go through a programme of cross-comparison of measurements with the Reference method. Achievement of specific criteria (including measurement uncertainty thresholds) must be met between the two samplers to show that the Candidate method is effectively “equivalent” to the Reference method. The Guidance for the Demonstration of Equivalence (GDE) of Ambient Air Monitoring Methods

sets out how a programme of cross comparison should be undertaken and the criteria to be fulfilled. GDE also applies to automated continuous particulate matter monitors as set out in the Technical Specification (BS16450:2013). The Technical Specification lays down minimum performance requirements and test procedures for the selection of automatic continuous particulate matter monitors (type approval processes), which includes an evaluation against the reference method as aligned to the requirements set out in GDE. Ongoing quality assurance / quality control minimum standards are also included in the Technical Specification, alongside information for different user groups ranging from general information on the principles of automated continuous particulate matter instruments to details geared towards test house and laboratories that perform the type approval tests.

The UK compliance network for measurement of PM_{10} and $PM_{2.5}$ is the Automatic Urban and Rural Network[1], which additionally reports on gaseous pollutants as required under the Air Quality Standards Regulations 2010[2]. For PM_{10} and $PM_{2.5}$ measurements instruments which have been shown to meet the requirements of equivalence are certified in the UK by the Environment Agency's appointed certification committee under the MCERTS programme for ambient measurement methods and published accordingly[3]. Further information can be found here: Certification - MCERTS for UK Particulate Matter - Defra, UK

Further details on the specific instruments deployed in the AURN for particulate matter monitoring is provided as follows:

1. Standard Reference Method

Several manufacturers make different instruments that conform to the European Reference Method requirements set out in EN12341:2014. Those deployed in the UK are the SEQ47/50 as manufactured by Sven Leckel GMBH based in Berlin, Germany.

As mentioned above, air is drawn at a rate of 2.3 m^3 hr^{-1} through a sampling head that is designed to select either the PM_{10} or $PM_{2.5}$ size fraction as required. The air flow containing the size-selected particles is then passed through a filter for 24 hours. The instrument holds multiple filters that are exchanged automatically on a daily basis.

EN12341:2014 defines a number of permissible filter materials which in turn can be made by multiple manufacturers to multiple specifications. The UK uses Teflon coated glass fibre filters as these have been shown to have limited effects due to absorption of water (as would quartz fibre or to lesser extent glass fibre) or problems with static and overloading the filter at moderate concentrations (as would Teflon). Further, Teflon coated glass fibre is only manufactured by a single manufacturer (Pall under the brand name Emfab), which reduces the potential for variability.

Filters are weighed twice before sampling and again twice after sampling. Prior to weighing the filters are conditioned at 45 to 50 % Relative Humidity (RH) and 20 to 21 ºC. The mass of the particulate matter collected on the filter is calculated as the average mass post

[1] https://uk-air.defra.gov.uk/networks/network-info?view=aurn
[2] https://www.legislation.gov.uk/uksi/2010/1001/contents/made
[3] https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme

sampling minus the average mass prior to sampling. The concentration is calculated as the mass of the particulate matter divided by the volume as measured by the SEQ47/50.

2. Candidate Methods

a) Beta Attenuation Monitor

Beta Attenuation Monitors use beta radiation to measure particulate concentrations. The instruments currently used in UK networks are Smart Heated versions of the BAM 1020 as manufactured by Met One based in Grants Pass, Oregon, USA.

For the PM_{10} version, air is drawn at a rate of 1 m^3 hr^{-1} through a sampling head (that is designed to remove particles greater than 10 micrometres in diameter) on to the tape. This is of a different design to that deployed in the Reference Method. Following this, the air stream is heated slightly to force some of the particle bound water and any water droplets to enter the gaseous phase. The air is then passed through a glass fibre tape, to which the particles are deposited, but the gaseous phase water that was previously in the particle phase passes through. The instrument works by measuring the Beta particle attenuation through a blank of tape for 4 minutes, then following 1 minute to move the tape, sampling PM_{10} laden air for 50 minutes through the tape, before moving the tape again and measuring the Beta attenuation for a further 4 minutes. The mass of particulate matter is calculated from the change in the Beta attenuation before and after sampling. The concentration is calculated as the mass divided by the volume of air sampled.

The $PM_{2.5}$ version is very similar. After the PM_{10} inlet it has a $PM_{2.5}$ cyclone that removes particles greater than 2.5 micrometres. It performs the Beta counts for 8 minutes rather than 4 and the sampling for 42 minutes rather than 50. This is to increase the signal to noise ratio and so more accurately measure $PM_{2.5}$ concentrations.

b) FIDAS 200 Method 11

The Fidas 200 is manufactured by Palas based in Karlsruhe, Germany. The Fidas 200 utilises optical particle counting and sizing to calculate mass concentrations. Air is drawn at a flow rate of 0.3 m^3 hr^{-1} through a sampling head that is not designed to remove larger particles but that does prevent insects entering the instrument. The instrument then heats the sample stream slightly to force some of the particle bound water and any water droplets to enter the gaseous phase. Following this the instruments counts particles and puts them in to bins of different size ranges. It then uses an algorithm to calculate PM_{10} and $PM_{2.5}$ based upon the numbers of particle in each bin combined with a pre-determined particle size density distribution. Through the programme of equivalence it was shown that raw $PM_{2.5}$ data needed to be corrected by dividing by 1.06. No such correction is required for PM_{10}.

c) Additional information for end users

Details of all the instruments which have passed full equivalence testing against the Standard Reference Method (SRM) and could be used for fixed measurements (25% uncertainty criteria) are given here: (https://www.csagroup.org/en-gb/services/mcerts/mcerts-product-certification/mcerts-certified-products/mcertscertified-productscontinuous-ambient-air-monitoring-system-mcerts-for-uk-particulate-matter/). The website has links for downloading the relevant test certificates and associated test reports

for end users to assess the demonstration of instrument performance together with the operating manuals. In the UK, currently only the MetOne BAM and FIDAS 200 are used extensively in the AURN, while the TEOM FDMS has been shown to be susceptible to location influences but is often still used by Local Authorities. Partisols are used for taking filter samples.

For completeness, instruments which meet the requirements for indicative measurements (50% uncertainty criteria) are listed here: https://www.csagroup.org/en-gb/services/mcerts/mcerts-product-certification/mcerts-certified-products/mcerts-certified-products-indicative-ambient-particulate-monitors/).

The BS 12341 and the GDE are all in the process of redevelopment in Europe but in the meantime the current equivalence framework is intended to continue. Defra will be reviewing this to ensure ongoing suitability and establish how the programme may need to be adapted to reflect the performance of the instruments and expanded uncertainty once assessed with respect to the new $PM_{2.5}$ target levels, as well as in response to the changing nature of $PM_{2.5}$ (i.e. reducing levels and changes in composition) in England over time. The work will also explore how datasets may be impacted by the introduction of new equivalence instruments onto the network.

3. Limitations

There are many complications associated with the regulatory definitions of PM measurement methods; they are essentially a "pragmatic compromise" in order to provide the basis of achieving a self-consistent set of ongoing results. In particular, ongoing PM measurements need to remain consistent with those used to develop exposure-response relationships for human health impacts. However, the result of the definitions are PM datasets that are not easy to model or to unpick scientifically:

- The 24-hour filter sampling can be subject to positive (condensation of gases onto the filter) and negative (evaporation of volatile aerosol components from the filter) sampling artefacts as temperature and humidity change over the 24-hour measurement period. As the most relevant consequence for the UK, the reference method can underestimate PM under certain circumstances due to the loss of ammonium nitrate aerosol (Hering and Cass, 1999).

- These losses are difficult to account for when comparing different measurement approaches or when comparing modelled and measured concentrations. Candidate Methods and models effectively need to demonstrate equivalence to what is a non-perfect method, either by mimicking the artefact or by developing correction procedures.

- Equivalent or indicative methods estimate mass by measuring other particle properties and make calculations based on an assumed canonical relationship. These assumptions are tuned to achieve required uncertainties under the defined test conditions but are liable to seasonal variation and changes in the pollution climate.

- The reference method mass-based measurements provide no insight into the relative contributions of the range of different sized particles delivered by the size selective inlets. Total mass could be dominated by a few larger particles or many in the smaller size fraction.

- For the analysis of source contributions to PM, particles tend to fall into two categories: particles derived from combustion or chemically formed in the atmosphere tend to be < 1 μm in the UK currently, whilst particles originating from friction processes, sea spray or resuspension tend to be > 1 μm in size. Thus, the measurement of $PM_{2.5}$ already represents a mixture of both sources. This makes source apportionment using $PM_{2.5}$ data harder and creates ambiguities when assessing the performance of models which often simulate the PM via their different source contributions.

2.2 On-line chemical composition

Measuring the chemical composition of PM in real time is of high value to air quality practitioners and policymakers and in the last 10 years technologies have matured such that this can be monitored routinely. Knowing the composition of PM in addition to the mass permits source apportionment and can provide enhanced constraint and validation of air quality models. Collecting data over long time periods supports epidemiological studies of differential PM toxicity. Because these instruments are often expensive to purchase and operate when compared to more basic air quality instruments, their use is currently limited to a few monitoring 'supersites'.

2.2.1 Aerosol mass spectrometry

The Aerosol Chemical Speciation Monitor (ACSM) is an instrument developed by Aerodyne Research, a more compact, lower-maintenance version of the earlier Aerosol Mass Spectrometer (AMS) and works on similar principles. Particles are introduced into a vacuum chamber, then impacted on a surface heated to 600 °C, upon which they instantly vaporise. The vapours are then chemically analysed using electron ionisation mass spectrometry (EI-MS).

Because of the thermal nature of the analysis, these instruments are limited to measuring the 'non-refractory' components of $PM_{2.5}$, which in practice is ammonium, sulphate, nitrate (that derives from ammonium nitrate), chloride (that derives from ammonium chloride) and organic matter. These instruments are available in PM_1 and $PM_{2.5}$ versions and also 'quadrupole' and 'time-of-flight' versions, which are equipped with different mass spectrometers, the 'time-of-flight' version being more sensitive but more expensive.

Such instruments are capable of delivering quantitative chemical mass concentrations in near real time, typically on a half hour basis for the quadrupole version. The organic data can also be further inspected for mass spectral features that may indicate the chemical origin of the material and can also be used as the basis for receptor modelling for source apportionment using algorithms such as positive matrix factorisation.

A weakness is that these instruments are not capable of measuring refractory material, in practice meaning black carbon, mineral dust, brake wear and sea salt, however when combined with a separate measure of black carbon, they are generally capable of accounting for almost all the mass of PM_1 in a polluted environment. Another problem is that they can be relatively more or less sensitive to certain types of organic aerosol. For instance, it was recently shown AMS can over-account for cooking, so corrections may have to be applied during source apportionment.

These types of instrument are moderately expensive to run, so within the UK only used at a few air quality 'supersites' rather than smaller monitoring stations. However, they are capable of delivering long-term datasets with good coverage. Because of the specific chemical compounds they are sensitive to, it is particularly useful for quantifying the secondary components of $PM_{2.5}$, both organic and inorganic. An example of the utility of this data is shown in Figure 2.2 taken from Chen et al. (2022), which is an assessment of

aerosol composition and sources across Europe based on an analysis of data from multiple sites, which illustrates the depth of variability of this across the continent.

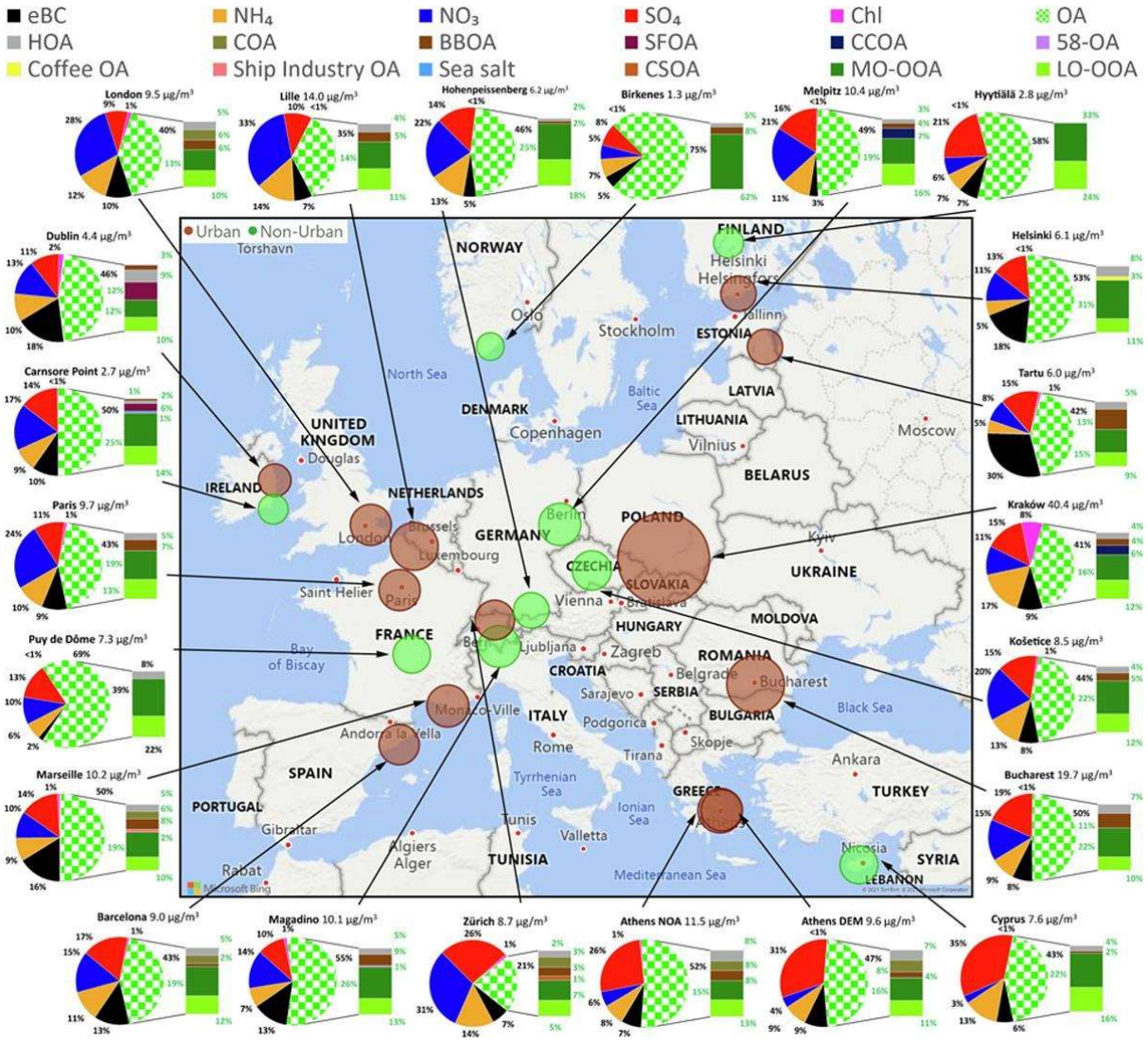

Figure 2.2: PM composition assessment across Europe based on ACSM and Aethalometer data, according to Chen et al. (2022). Marker sizes correspond to total PM mass and brown denotes urban monitoring sites, and green rural. Categories not identified by chemical formula or name include: Equivalent Black Carbon (eBC), chloride from ammonium chloride (Chl), organic aerosol (OA), hydrocarbon-like OA (HOA, likely from diesel exhausts), cooking OA (COA), biomass burning OA (BBOA), solid fuel OA (SFOA), coal combustion OA (CCOA), unknown OA with high signal at m/z=58 (58-OA), OA from coffee roasting (coffee OA, specific to Helsinki monitoring site), cigarette smoke OA (CSOA), more oxidised oxygenated OA (MO-OOA, likely secondary), less oxidised oxygenated OA (LO-OOA, likely secondary).

2.2.2 Semicontinuous XRF

The technique of semicontinuous X-Ray Fluorescence is used in instruments such as the Cooper XACT and Horiba PX-375. PM is collected on a reel-to-reel filter tape and at the end of a sampling period (e.g. an hour), the filter is advanced and passed for analysis by X-Ray Fluorescence (XRF) spectroscopy. This can quantify mass concentrations of a wide range of elements, mainly metals but also some elements such as chlorine and sulphur.

This technique is not capable of accounting for a large fraction of the PM mass budget, however it is highly useful in detecting certain types of PM that cannot be measured using the other techniques, in particular mineral dust and non-exhaust vehicle emissions (tyre wear, brake wear and resuspended road dust). The contributions of these sources can be estimated through using tracer metals as proxies or through receptor modelling. The presence of certain metals can also serve as a good indication of emissions from construction and certain industrial sectors, and events such as fireworks. Other aerosol types that can be detected are biomass burning, as this typically contains large concentrations of potassium, and 'fresh' sea salt through chlorine. An example of the type of data that can be provided is given in Figure 2.3.

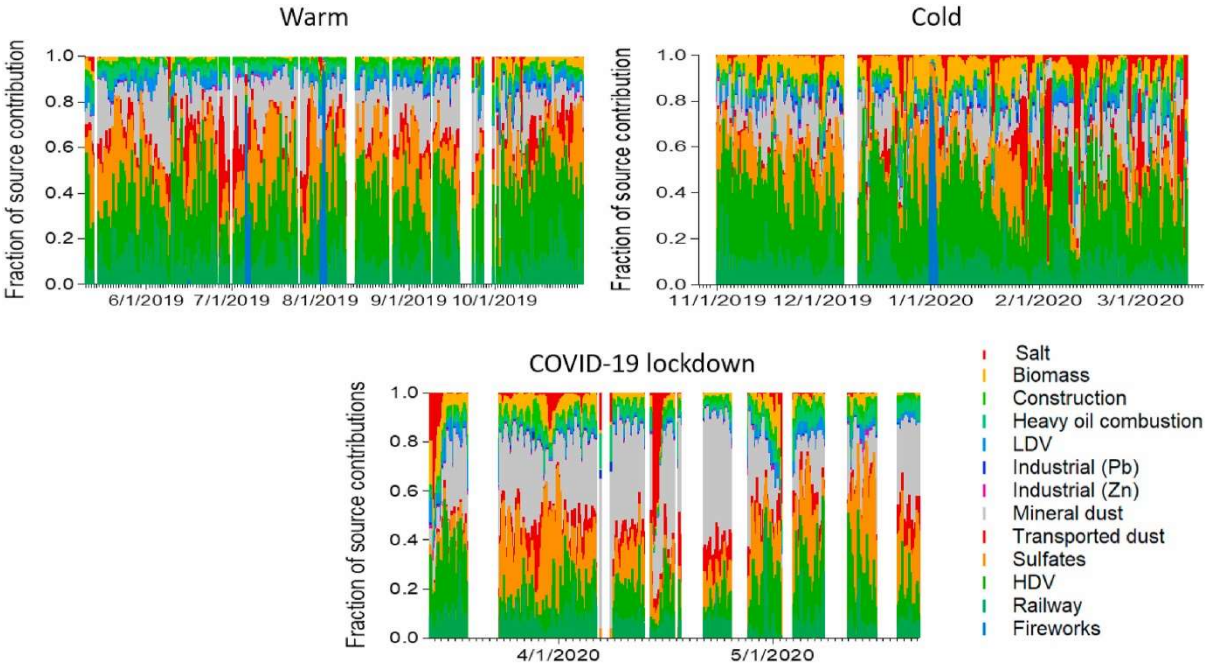

Figure 2.3: Analysis of source contributions to metals measured by an XACT, as presented by Manusakas et al. (2022). 'Warm' and 'Cold' refer to weather conditions during the sampling period. LDV and HDV refer to light and heavy-duty vehicles respectively.

2.2.3 Semicontinuous total carbon or thermal-optical

An alternative method for measuring the carbonaceous component of PM is to collect samples of PM on a filter over a period, then oxidise in a heated air flow, subsequently detecting the carbon that is liberated in the form of carbon dioxide. A more elaborate version of this is thermal-optical analysis which performs a similar programmed temperature ramp with inert and oxidising gas stages to deliver concentrations of 'elemental' and 'organic'

carbon, as with measurements performed offline on filter samples. However, it has been found that the online data is less accurate than offline.

A more straightforward approach is to quantify the total carbon using a single stage of analysis and assign the carbon to 'black' or 'organic' through comparison with a supporting black carbon measurement, e.g. an Aethalometer. Currently Magee Scientific and Sunset Labs offer instruments performing this function. This in effect provides an alternative measure of the organic content of PM compared to the ACSM, although it should be noted that strictly, total carbon analysis measures the carbon whereas the ACSM measures the mass of organic matter present, so to construct a mass budget, a ratio of organic carbon to organic matter must be assumed.

2.2.4 Online wet chemistry methods

Online wet-chemical analysis methods of PM composition provide a sub-daily (typically half-hourly or hourly) measurement of water-soluble aerosol chemical components that is a near high-resolution analogue of the European reference method. They also provide the opportunity to be coupled with gas-phase measurements that are difficult to realise with other methods, including for HNO_3, HCl and NH_3, into single instruments.

Online wet chemistry methods tend to work on the principle that gas-phase contributions are first removed from the air stream, using denuders – these may be continuous rotating wet denuders, parallel plate denuders, or denuders that need to be manually replaced on a regular basis. The solution from the continuous denuders can be analysed online to provide additional measurements of gas-phase components. The air passing through the denuder then passes a device where the particles are sampled into a liquid, usually water. The two common approaches are the Particle-Into-Liquid-Samplers (PILS) (Weber et al., 2001) and the steam-jet aerosol collector (SJAC) (Khlystov et al., 1995). In both systems particles get activated in a supersaturated environment and grow to a size where they can easily be collected by inertial impaction. The solute stream can then be continuously transferred to any online analysis system for liquid samples, or periodically filled into vials for offline chemical analysis (PILS Model 4001, Brechtel) (Sorooshian et al., 2006). The most common online coupling is to online ion chromatograph (IC) for anions (to measure NO_3^-, SO_4^{2-} and Cl^-) and/or cations (to measure NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}), as is done in the two commercial systems, i.e. the Monitor for Aerosols and Gases in ambient Air (MARGA 2060, Metrohm) and the Ambient Ion Monitor (AIM; 9000 Series, URG). This provides typically hourly water-soluble concentrations of these ions in either PM_1, $PM_{2.5}$ or PM_{10} (and potentially the gases NH_3, HNO_3, HCl, SO_2 and HONO) in air. Gas-chromatography systems can be further modified to extend the measurement to some organic compounds including organic acids (Nah et al., 2018; Stieger et al., 2019), amines (Hemmilä et al., 2018) and the biomass burning marker levoglucosan (Saarnio et al., 2013). Both PILS and SJAC have also been interfaced with analysers for total soluble carbon and nitrogen (Lin et al., 2010), and with measurements of trace metals.

The online-IC systems measure water-soluble compounds and basically mimic the European reference method for aerosol speciation measurements in which air is sampled through filters that are later extracted into water and analysed by lab-based IC. They therefore in theory deliver very comparable results with the added advantage of (a) providing

higher temporal resolution data helpful to understanding diurnal and meteorological controls, (b) minimising contamination due to manual filter handling, (c) remove the gas phase from the air stream and thus reduce gas/aerosol interferences and (d) limit the evaporation of volatile aerosol components such as ammonium nitrate during sampling. Disadvantages of the method frequently include the use of inlet systems that represent a compromise between the sampling needs for gases and particles and are thus not completely ideal for either. There are also some chemical interferences for some of the compounds, although this mainly affects the gas-phase: N_2O_5 is known to contribute to the reported HNO_3 concentration and the combination of NO_x and SO_2 can create a positive interference for HONO, whilst low pH can create a negative interference (Phillips et al., 2013; Xu et al., 2019).

In the UK, dual $PM_{2.5}$/PM_{10} SJAC-IC systems of the MARGA type have been operated at the two rural EMEP Supersite: at Auchencorth Moss, Scotland, since 2006 and at Harwell, Oxfordshire, since 2009 which was transferred to Chilbolton, Hampshire in January 2016. The URG AIM $PM_{2.5}$ instrument was operated at the London supersite at Marylebone Road (roadside) and North Kensington (urban background) from 2012 to 2021.

2.3 Optical properties, including black carbon

Measuring the optical properties of airborne particles has been a longstanding component of air pollution monitoring. The general technique continues to be of high value, for example differentiating the contribution to $PM_{2.5}$ from wood burning as separated from fossil fuel combustion.

Historically high concentrations of PM have been associated with visibility reduction. This arises through PM scattering incident light, a process that can be enhanced by high humidity. Hazes ('smog') associated with pollution are common in some parts of the world (in particular Asia) and used to be a regular phenomenon within the UK, but these are generally now less common. Nevertheless, severe pollution episodes still often produce a noticeable haze.

Instruments for measuring light scattering of PM are known as nephelometers and the technology is now mature. Instruments of this kind formed a mainstay of the IMPROVE network in the US, which was set up to monitor the impact of hazes in the US National Parks (Solomon et al. 2014). It is also a feature of some newer monitoring instruments such as the DMT PAX (which also measures black carbon photoacoustically) and the Aerodyne CAPS PM_{SSA} (which also measures optical extinction).

While not a measure of PM mass, these instruments can provide a real-time measure of visibility reduction, which in addition to causing issues for aviation and tourism has also been hypothesised to have a negative impact on peoples' mental health. They can also be used to provide supporting *in situ* data for remote sensing products (Section 3).

2.3.1 Filter-based absorption techniques

The measurement of the light absorbed by particles collected on a filter is the physical basis of one of the first practical ways to measure airborne particle pollution and the most impactful air pollution measurement technique of the 20th century (Fuller, 2018) . Today this approach is used for the measurement of black carbon particles.

Measurement of PM mass concentration by weighing material collected on a filter is very challenging today and was even more difficult with the mechanical balances available in the UK in 1910's. With a soot dominated aerosol it was, however, possible to use the darkness of material collected on a filter as a surrogate for total PM. The first Owens automatic samplers were designed as field instruments that could be deployed in large numbers and used by non-experts.

The light absorption method was not sensitive to non-light absorbing particles. This was first noted in the 1920s (Whipple et al 1929). As soot-like aerosol declined the reflectance or black smoke technique became a less useful, and eventually useless (Ball and Hulm, 1977), as a surrogate for airborne particle concentration in the developed world.

However light absorption techniques gained a second lease of life for the measurement of black carbon. The first aethalometer was created by Hanson et al (1982) as a real time black carbon instrument to investigate aerosol composition in remote locations. A light source and detector made continuous measurements of relative absorption as particles deposited on a filter. Non-linearities in light absorption and deposited filter mass of black carbon was already known from the earlier reflectance measurements (OECD, 1964) and required much post-measurement processing which is now done within modern aethalometers (Collaud-Cohen et al. 2010; Drinovec et al. 2017).

Series of papers in the 21st century used the common physical basis to link together the black carbon and reflectance black smoke techniques and enabling black carbon concentrations to be derived from reflectance black smoke measurements (Quincey, 2007, Quincey et al. 2011, Heal and Quincey, 2012; Davey et al. 2017.)

The original simplicity of the aethalometer has led to the development of wearable black carbon devices that have been widely and successfully deployed in traffic pollution exposure studies. In time, with better vehicle tailpipe emission control, black carbon will no longer be an effective tracer for traffic emissions.

Black smoke measurements in the UK

Owens automatic samplers enabled the creation of the UK's (and the world's) first national air pollution measurement network (Mosley, 2009) soon after the end of the first world war. In the days before reliable electric pumps, samplers used a water syphon (rather like a toilet cistern flush) to draw a known volume of air through a filter at times determined by a clockwork mechanism. Each station operator was supplied with a set of swatches that could be compared by eye to the blackness of the particle sample on the filter. Thus, a concentration could be read and recorded by the operator (Owens, 1918; Owens, 1926). Over time the Owens instrument was adapted to make daily samples using electric pumps and electronic reflectance meters replaced the manual comparison of dark spots and swatches.

In the 1952 smog, Owens instruments were key to establishing unequivocal evidence on health harm from air pollution. Following the 1956 Clean Air Act, they were deployed in very large numbers across the UK to identify problem areas and track changes. The network peaked at over 1,200 measurement sites in the 1970s (Shaddick and Zidek, 2013). Instruments based on filter reflectance formed the mainstay of PM measurement in the UK and globally until the advent of electronic beta radiation instruments in the 1970s (Liberti, 1975) and tapered oscillating microbalance instruments in the 1990s (Patashnick and Rupprecht, 1991).

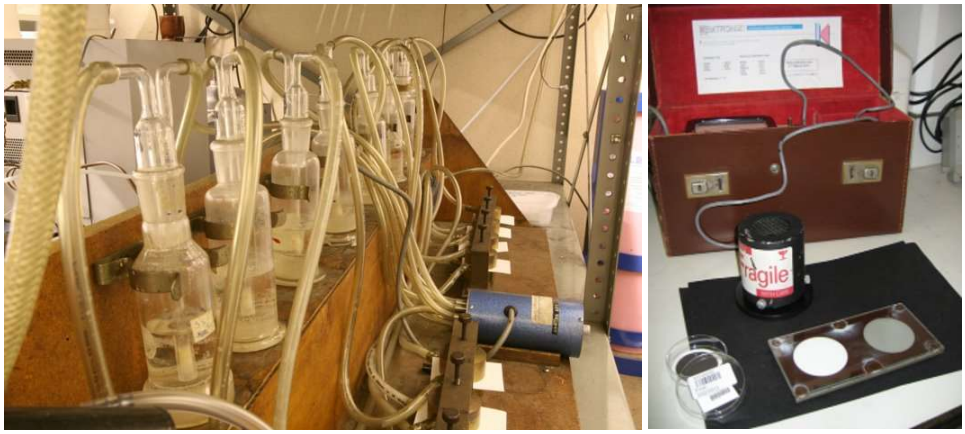

Figure 2.4. The smoke and sulphur dioxide bubbler operated at Marylebone Road in the early part of the 21st century (Left). Based on Owen's original concept, daily samples were collected on filter papers for subsequent analysis using a reflectometer (right). Photos Imperial College London and Pamela Davy.

2.4 Particle number and size measurement

The online monitoring of particle number and size has been common in the research community for several decades; its use has recently proliferated in polluted environments due to increasing interest in ultrafine particles and their possible enhanced role in causing health impacts. The protocols for calibration, measurement and validation are now becoming standardised which will likely increase the value of such measurements in health effects studies.

There has long been instrumental capability to measure both total particle number and particle number size distributions, but despite the existence of long-running AURN monitoring sites in the UK there are only limited datasets on particle number and size. Such datasets are however valuable and offer important opportunities for interpretation of processes and sources. Measurement of particle number remains the only practicable way of measuring ultrafine particle concentrations, an activity highlighted by WHO (2021) as important. Additionally, particle size distributions are a key indicator of particle sources, and are an essential component of recognising and quantifying events of new particle formation in the atmosphere. Both total particle number data and particle number concentrations within defined size ranges have been used in health effect studies in attempts to elicit those particle groups most responsible for health impacts (e.g. Samoli et al., 2016; Rivas et al., 2021). While these have not, to date been used as formal compliance metrics, standards for the measurement of particle number concentrations and sizes are included in ACTRIS and CEN specifications in Europe, which have also been adopted by the ASCENT network in the USA.

The basis for most measurements is the condensation particle counter (CPC). In this instrument, a continuous stream of air is subjected to a supersaturation of a vapour, normally either n-butanol or water. This causes the particles in the air stream to grow to sizes where they can effectively scatter a light beam, allowing individual particles to be counted as pulses of scattered light. CPC instruments measure over a wide range of sizes with a lower cut-point (D50) normally in the range of 3-10 nm. Because size distribution is usually heavily biased towards smaller sizes, around 80-90% of the particles counted are in the ultrafine range of <100 nm, as normally defined.

To make size-differentiated measurements, the CPC is preceded by an electrical charger and a differential mobility analyser (DMA), which allows separation of charged particles based on their electrical mobility (a function of their size and charge). In the DMA, the electrically charged particles pass through a grounded tube which has a charged electrode in the centre to create an electric field, and the particles migrate towards the electrodes at a rate determined by their electrical mobility. Most particles deposit to the electrodes, but the device is “tuned” by a combination of voltage and flow rate such that particles in a narrow range of sizes pass out of the tube through an opening and are passed to the CPC for counting. Normally, flow rates are fixed, and the voltage is scanned, so as to measure the entire range of particle sizes within a period of 1-2 minutes. The devices are referred to as Scanning Mobility Particle Sizers (SMPS) or Mobility Particle Size Spectrometers (MPSS) (see Wiedensohler et al. 2012).

An alternative design of electrical mobility analyser allows all particle sizes to be measured in near-real time by using a constant electric field and a series of electrometers down the length of the tube, to collect and measure all size fractions continuously, as the particles migrate to different electrometers according to their size and charge. All such mobility analysers depend upon knowing the charge distribution of the particles as a function of size, with most charged particles being singly charged, and corrections made in the instrument software for multiple charging. Multiple charging becomes a problem above about 1 μm diameter, and it is usual to incorporate an impactor to remove particles above this size. Scanning Mobility Particle Spectrometers can cover a part of the size range from 3 nm to 1000 nm, depending on the length of the DMA and the flow rate selected. Extending to smaller sizes can be achieved with a Particle Size Magnifier which operates like a CPC, but using di-ethylene glycol as the condensing reagent, and scans through different supersaturations so as to measure different particle sizes. Ultrafine particles (in terms of number or lung deposited surface area) can also be measured using devices employing diffusion charging. While this principle has until recently mainly been employed for handheld sensors, there are now versions suited for static monitoring available (e.g. AQ Guard Smart 2000). However, metrological standards do not exist for this type of measurement yet.

Extending the range to larger particle sizes can be achieved using data from instruments separating on the basis of the aerodynamic properties of particles (e.g. Aerodynamic Particle Sizer), or the optical properties using optical particle counters (e.g. FIDAS) (Beddows et al, 2010). In these cases, the data have to be merged by software which accounts for the different measurement principles of the instruments, as the diameter of a particle measured by electrical mobility is not the same as that measured from its aerodynamic or optical properties (Allan and Watson-Parris, 2022).

There are several uses of size distributions in evidencing the sources and processes affecting airborne particles. New particle formation through regional nucleation processes can lead to greatly enhanced number concentrations of ultrafine particles. This tends to occur in “events”, mostly in the afternoon which can be recognised from the growth trajectory of a new mode of particles starting from the smallest being measured and showing a curved growth trajectory on the traditional plot of dN/dlogD (colour) as a function of size (y-axis) and time (x-axis), commonly referred to as a “banana plot” (Kulmala et al., 2012).

Large numbers of consecutive particle size distributions can also be analysed by Cluster Analysis, which groups data into the most frequently measured size distributions. These can be associated with individual source types or processes in less polluted atmospheres, but are generally too complex to interpret in terms of sources in polluted atmospheres (Beddows et al., 2009). On the other hand, application of Positive Matrix Factorization (see section 6.1) can elicit size distributions associated with specific particle sources, aiding source apportionment (Gu et al., 2011). This is often less successful than apportionment based on chemical composition but can recognise sources such as new particle nucleation, which can be very hard to separate in PMF of chemical composition due to the small amount of new particle mass created.

2.5 Off-line methods

Off-line analyses enable state-of-the-science analytical tools to be applied to PM composition measurements using lab techniques that cannot yet be operated continuously in the field. Application to archives of PM filter samples can permit greater spatial coverage and longer time series of detailed chemical composition to be obtained. Offline methods often provide molecular-level information that illuminates sources and chemical process that contribute to the PM burden, including markers used in source apportionment methods. Analyses of archived PM can help retrospectively quantify historic changes, including, for example, evaluation of responses to policy.

Offline methods of PM analysis are typically applied to PM collected on filter or impaction surfaces over a specific period of time. Before the advent of online PM analytical methods, all chemical and morphological PM data was derived in this way. Offline analyses enable the deployment of techniques that can provide very detailed molecular information on PM composition, but which cannot (yet) be undertaken in real time, or to save on the cost and complexity of field deployment. PM collection with a high-volume sampler is usually required when levels of target analyte are very low and/or samples are to be split for multiple offline analyses on the same sample. Offline techniques also allow for acquisition of compositional information from longer time series of archived PM samples and/or from a greater number of sampling locations than may be possible from online measurements. Caveats include potential bias in the portion of PM that is re-sampled in the offline analysis, and susceptibility to chemical changes in the PM composition compared with ambient, but these can apply to online methods also.

The majority of compositional data still is derived from offline analyses. These include aqueous extractions followed by ion chromatography for major anions and cations or inductively-coupled plasma (ICP) optical emission spectroscopy (OES) or mass spectrometry (MS) for elemental concentrations following acid extraction. Elemental quantification can also be effected *in situ* on filter samples using energy-dispersive X-ray fluorescence and particle-induced X-ray emission (Chiari et al., 2018). Mass spectrometric detection following liquid chromatographic or gas chromatographic separation of organic extracts of PM filter samples is used to quantify many organic species, for example PAHs, PCBs, brominated flame retardants, long-chain alkanes, levoglucosan and other biomass combustion markers and benzothiazole and its derivatives as markers for tyre-wear. The application of two-dimensional gas chromatography has greatly enhanced separation capability facilitating, for example, the separation of petroleum hydrocarbons and other mixtures of isomeric compounds (Alam and Harrison, 2016).

Offline radiocarbon (carbon-14) determination is an effective tool for discriminating between fossil and contemporary sources of carbonaceous PM, particularly when undertaken on EC and water-soluble/water-insoluble fractions of OC separately and in conjunction with quantification of other source markers (Heal, 2014; Crilley et al., 2015).

Advances have been made in applying HR-ToF-AMS to airstreams of particles resuspended from filter samples. Filters are extracted in a solvent, typically ultrapure water (Bozzetti et al., 2017; Daellenbach et al., 2016; Daellenbach et al., 2017), but organic solvents have also

been used (Mihara and Mochida, 2011; Chen et al., 2016). The solutions are aerosolised, typically using HEPA-filtered compressed air, which then passes through a silicon-filled diffusion dryer to remove as much as solvent as possible, before entering the AMS inlet. The PM extraction efficiency and the nebulisation efficiency need to be quantified. This is often based on the water-soluble organic carbon component. Source apportionment methods such as positive matrix factorisation can then be applied, similar to online AMS data. The following are some example applications. Daellenbach et al. (2017) determined the relative contributions of six sources of organic aerosol to 819 filter samples of PM collected over a full year at 9 sites in central Europe. Chen et al. (2016) combined offline HR-AMS and FT-IR to differentiate different fractions of humic-like substances in the water-soluble and water-insoluble organic matter in total suspended PM in Japan. Vlachou et al. (2018) used offline AMS and carbon-14 measurements on 219 PM samples from an Alpine in valley in Switzerland to provide insight into the sources of SOA based on the type of SOC precursor and not on the volatility or the oxidation state of OC, as is typically considered.

The greatest recent technical advances in off-line compositional analysis couple ultrahigh performance chromatography and high-resolution mass spectrometry to identify and quantify an expanding suite of ultra-trace molecular species, particularly of secondary PM formation chemistry (Nozière et al., 2015; Johnston and Kerecman, 2019). Bryant et al. (2020) coupled ultra-high-performance liquid chromatography and heated electrospray ionization Orbitrap tandem mass spectrometry in offline analysis of $PM_{2.5}$ filter extracts to quantify 31 proposed molecular tracers for isoprene-derived SOA (iSOA). The results indicated that iSOA formation in urban Beijing was strongly controlled by anthropogenic emissions and results in extensive conversion to organosulfate products from heterogeneous reactions. Du et al. (2022) applied the same UHPLC-Orbitrap MS technique to offline determination of a suite of molecular markers of secondary organic aerosol collected in smog chamber experiments.

2.6 Low Cost Sensors

Recent technological developments have led to air quality sensor systems which provide a more compact and easily deployed instrument for PM measurements. These can complement existing instruments assessed as equivalent to the **reference methods. Sensor systems have the potential to support air pollution monitoring at a higher spatial density than is possible with reference methods. They also allow for new applications when coupled with spatial information, for example to provide understanding of an individual's exposure to PM, in turn delivering additional information for policy planning.**

Sensor systems share some common features with traditional reference methods despite having improved portability and lower cost. Typically, they continuously monitor PM with fast response times ranging between a few tens of seconds and a few minutes, although presently with a reduced measurement accuracy (higher uncertainty).

Sensor systems may contain one or more sensors based on different principles of operation to measure a range of pollutants of relevance to air quality monitoring. For example, these can include electrochemical sensors for NO_2, NO and O_3, non-dispersive infrared (NDIR) for CO_2, and light scattering optical particle counters for the detection of PM. The term "low

cost" refers to the initial purchase price of a single air quality monitoring system and is not an indicator of operational or ongoing costs, for example those associated calibration. The operational cost is dependent on the number of pollutant sensors employed and whether end users implement a large network of systems. Importantly the purchase price might not account for additional ongoing charges for access to the measurement data.

The implementation of larger networks of low cost sensors in projects such as in the recent "Breathe London" pilot study, and others, has enabled problematic measurement artefacts to be identified as more data is acquired from a range of locations and over a number of seasons. PM sensor outputs may be subject to regular pulsing due to electromagnetic interference during data transmission although the selection of appropriate power supplies may provide a remedy. During episodes of fog, optical particle counters (OPCs), which do not have heated inlets (in order to save power), are unable to differentiate small water droplets from particles. High humidity data flags can be used to identify such fog events, and only affect the data for relatively short periods. Heating of the sensor inlet, if possible, can reduce this cross interference but care needs to be taken not to remove volatile components from the PM being measured.

Low-cost PM sensors based on optical techniques report $PM_{2.5}$ and then employ algorithms which assume a certain particle distribution to estimate PM_{10}. Caution is required in interpreting measurements carried out at industrial sites where the particles and their distributions may be different to those normally present in ambient AQ monitoring applications (Rueda et al. 2023). End users are encouraged to collocate and calibrate PM sensors against the reference method as it is well known that reference equivalent methods may deliver different calibration factors based on measurement location and other environmental factors.

2.6.1 Latest on QA/QC/standards for sensors

Whilst the UK has performance standards for reference quality instruments and guidance for monitoring under Local Air Quality Management (LAQM) application, there is no formal code of practice for the evaluation, deployment and use of low-cost sensors and networks composed of these sensors. The European Committee for Standardization (CEN) brings together the national standardization bodies of 33 European countries and provides the platform for the development of European standards and technical documents. CEN Technical Committee 264 Working Group 42 was set up in September 2015 to develop a European Technical Specification (TS) for low cost sensors. The composition of WG42 included experts from the various National Measurement Institutes (NMIs), Operators of Air Quality Networks, sensor manufacturers, manufacturers of complete low cost monitoring systems, and the Joint Research Centre (JRC) Ispra.

The TS is not yet a CEN standard and is thus not mandatory in the EU, because the method is not mature and there is need to investigate its validity through pre-normative research studies. CEN/TS 17660-1:2021 Air quality-Performance evaluation of air quality sensor systems-Part 1: Gaseous pollutants in ambient air was published recently and Part 2 on PM is substantially in progress and is anticipated to be published in early 2024.

The proposed performance requirements for low cost sensors are given in Table 2.2:

Table 2.2: Performance requirements for low cost sensors

Pollutant	Required uncertainty of reference methods	Expanded uncertainty of indicative methods **Class 1**	Expanded uncertainty of objective estimations **Class 2**	Expanded uncertainty of informative measurements **Class 3**
SO_2, NO_2/NO_x, CO	15%	25%	75%	200%
Benzene	25%	30%	100%	200%
$PM_{2.5}/PM_{10}$	**25%**	**50%**	**75 %**	**200%**
O_3	15%	30%	100%	200%
Thresholds†	>UAT	UAT<[ ]<LAT	<LAT	200%

†UAT = Upper Assessment Threshold, LAT = Lower Assessment Threshold, [ ] = concentration

The TS subdivides sensor systems into 3 categories (Class 1, 2, and 3) and describes the methods to check if sensors can reach the Data Quality Objectives (DQO) for indicative methods, and objective estimations, as defined in the European Directive for Air Quality. Class 3 systems are identified as systems which are not for regulatory reporting but may be useful for citizen science applications including educational tools for schools. In all cases 3 systems are required to be evaluated through type-testing.

Objective estimations are a category reserved for the air quality zones with very good air quality and no large conurbations. It is usually combined with modelling. By identification of local pollution sources and information of regional air quality an estimation of concentration of a regulated pollutant is made.

Part 1 of the TS allows sensor system manufacturers to select from two possible type-testing routes. All systems will have to undergo a few preliminary laboratory tests, followed by more extensive tests in the laboratory and 2 field trials collocated with reference instruments. Alternatively, more extensive (double) field tests can be elected instead of the additional laboratory tests.

Due to the difficulty of generating atmospheres of known $PM_{2.5}$ and PM_{10} composition in the laboratory, Part 2 of the TS focusses on testing PM sensor systems in the field. There are still areas requiring further discussions due to the problem that the PM reference equivalent methods are known to respond differently depending on location, season, and composition. Compliance with the DQOs will be based on comparison with the reference method using 24 hour data. A relative humidity test against an equivalent method is being considered with the requirement of a zero baseline restricting this test to optical systems. PM_{coarse} tests in the laboratory and field tests based on hourly reference data are being considered to ensure that systems cannot be undeservedly accredited for PM_{10} measurements.

Defra and the British Standards Institute (BSI) has commissioned the National Physical Laboratory to develop a Publicly Available Specification (PAS) titled “Air quality monitors – Selection, deployment, and quality control of mountable, static air quality sensors and sensor systems in ambient air – Code of practice. The PAS is intended to provide

recommendations and guidance for end users, including local authorities, environmental health officers, citizen scientists, industrial users, sensor/sensor system manufacturers and suppliers, policy makers and those with an interest in air quality quantification and improvement. The PAS will provide recommendations for the selection, deployment, quality control and calibration of air quality monitoring systems with sensor technology as standalone units or as part of a network. It will cover sensor systems for the measurement of both gaseous and particulate pollutants and is due for publication in late 2023.

Chapter 3 - Technology state of the art: Earth observation

Comprehensive geospatial coverage of PM concentrations is highly desirable as it provides a nation-wide view of pollution and allows for the detection of unexpected or unanticipated sources. Quantifying PM concentrations at the planetary surface using satellite-based instruments is challenging. New space-based instruments combined with recent advances in atmospheric modelling, machine learning, data handling, and data inversion techniques, have led to the development of new data products that can estimate surface concentrations of PM across the whole of the UK.

Earth Observation (EO) data products have regional to global coverage and timeseries records that for some instruments span more than two decades. These data have been used to evaluate the spatial distribution and long-term changes in $PM_{2.5}$ and been paired with health risk models to quantify global and regional health burdens resulting from population exposure to $PM_{2.5}$. Use of satellite-derived $PM_{2.5}$ data products can address gaps in the geographic distribution coverage of ground-based monitoring networks. Careful consideration of the limitations and uncertainties of EO data products is required however, because $PM_{2.5}$ is not directly observed. This section presents a broad overview of the approaches adopted to derive surface concentrations of $PM_{2.5}$ from Earth observations, issues and limitations with the data, use of independent measurements to validate and correct biases in the product, and details of products available for public use.

3.1 Estimating surface concentrations of $PM_{2.5}$ from satellite observations

Satellite-derived gridded data products of surface concentrations of $PM_{2.5}$ have been widely used to determine the burden of disease from exposure to ambient air pollution on global, regional, and local scales, as these offer the benefit of addressing large data gaps in the global network of surface observations (see for example van Donkelaar et al., 2019, Halloway et al., 2021). The UK has a relatively dense network of 103 monitors measuring ground-level $PM_{2.5}$ as part of the national Automatic and Rural National Network (AURN) and 166 reference equivalence monitors managed by local authorities (Section 2.1). However, more than 90% of these national and local monitors are in urban regions. This limited spatial coverage biased toward urban areas limits the ability to assess UK air quality models, determine the health effects of UK population exposure to air pollution, determine long-term trends and spatial variability in UK-wide $PM_{2.5}$, and assess the efficacy of policies targeting $PM_{2.5}$ pollution. The map in Figure 3.1 shows annual mean $PM_{2.5}$ from a satellite-derived product providing complete coverage of the UK compared to the patchy coverage of the AURN network concentrated in urban areas.

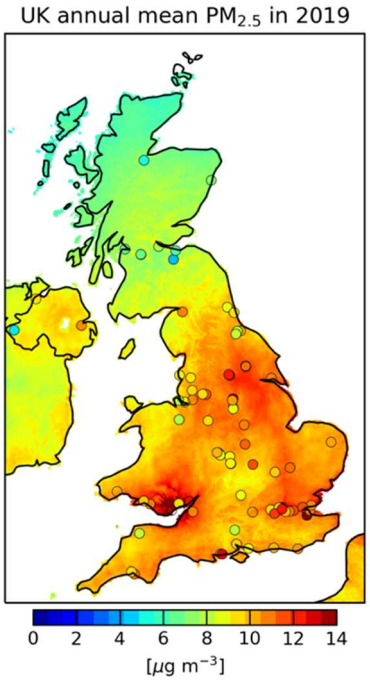

Figure 3.1: Annual mean surface concentrations of $PM_{2.5}$ in the UK in 2019. The background is the ~1 km spatial resolution satellite-derived $PM_{2.5}$ product developed by Washington University at St Louis (Table 3.2). Filled circles are 75 of the total mostly urban national monitors with > 80% temporal coverage in 2019.

Space-based sensors measure the abundance of pollutants in a vertical column of air from the surface of the Earth to the top of the atmosphere. Further steps are needed to convert these total columns to ground-level concentrations of air pollutants. Earth observation of aerosol abundance are via aerosol optical depth or AOD; a unitless measure of the extinction of light as it passes through the atmosphere due to its reflection or absorption by aerosols. In instances where a large portion of $PM_{2.5}$ in the column of air is concentrated close to the Earth's surface, the AOD serves as a proxy for trends and variability in $PM_{2.5}$ (Vohra et al., 2021). As a result, surface concentrations of $PM_{2.5}$ can be derived using Earth observations of AOD.

The earliest demonstration of deriving $PM_{2.5}$ from Earth observations of AOD used a generalized linear regression model to relate AOD and $PM_{2.5}$. The resultant product was annual mean $PM_{2.5}$ at ~100 km resolution over a portion of the eastern US (Liu et al., 2005). Derivation methods have since evolved to relate AOD and surface $PM_{2.5}$ using chemical transport models that simulate both AOD and $PM_{2.5}$ (van Donkelaar et al., 2006), simple linear regression models (Wang et al., 2003), multiple regression models or generalised additive models (Al-Hamdan et al., 2009, Ma et al., 2014) and machine learning techniques dominated by random forest (Section 5.2) (Meng et al., 2018, Stafoggia et al., 2019, Scheider et al., 2020). Further advances have included the use of multiple satellite observations of AOD, as well as data gap filling techniques and gridding approaches, such as oversampling and statistical downscaling leading to products with global coverage at increasingly finer scales. In some instances, daily means at 100 m resolution (van Donkelaar et al., 2021).

A prominent AOD instrument (or optical sensor) used to derive $PM_{2.5}$ is the passive Moderate Resolution Imaging Spectroradiometer (MODIS) (Table 3.1). There are two of these on separate satellites that have been providing observations for more than two decades. MODIS is a popular choice as it is at relatively high spatial resolution for regional and global air quality applications (<10 km), has a long and consistent temporal record (>20 years), achieves global coverage every day, is available free of charge for public use, is in a data format that is relatively easy to use, includes extensive and comprehensive user documentation, and undergoes substantial validation and retrieval improvements (https://modis.gsfc.nasa.gov, last accessed 17 November 2022). Many other sensors with a range of spatial and temporal resolutions have been used to a lesser extent to derive $PM_{2.5}$ (Table 3.1).

Table 3.1. Details of satellite aerosol optical depth (AOD) observations with coverage over the UK used to derive $PM_{2.5}$ [a]

Instrument	Satellite	Launch year	Spatial Resolution [km]	Global coverage [b]	Overpass time
MODIS	Aqua	2002	1/3/10	Daily	13h30
MODIS	Terra	2000	1/3/10	Daily	10h30
MISR	Terra	2000	4.4/50 [c]	7-9 days	10h30
SeaWIFS	OrbView-2	1997 [d]	13.5	Daily	12h00
AATSR	Envisat	2002 [e]	1	3 days	10h00
MERIS	Envisat	2002 [e]	1.2	3 days	10h00
VIIRS	Multiple [f]	Multiple [f]	0.375	Daily	13h30
3MI [g]	Metop-SG-A1 to -A3	2024	4	Daily	09h30

[a] AOD product information obtained from the Observing Systems Capability Analysis and Review Tool (OSCAR) (https://space.oscar.wmo.int/satellites/, last accessed 2 November 2022) [b] Time takes to achieve daytime global coverage. [c] 4.4 km is the resolution of the recently described version 23 AOD product (Garay et al., 2020). [d] Ceased collecting data in 2010. [e] Ceased collecting data in 2012. [f] Includes SNPP in 2011, NOAA-20 in 2017, JPSS-2 to -4 in 2022-2039. [g] Three consecutive launches expected from 2024 to 2045.

3.2 Limitations of satellite-derived $PM_{2.5}$ data products

Clouds lead to large loss of AOD data, in particular in winter in the UK when $PM_{2.5}$ pollution is at its worst (Defra AQEG, 2012). The current constellation of instruments measuring AOD (Table 3.1) also only provide one observation per day, though this can be partially addressed by combining AOD data from multiple instruments with different (morning, midday) overpass times, as in Hammer et al. (2021) and van Donkelaar et al. (2021). The chemical transport models that are used to convert AOD to $PM_{2.5}$ are prone to errors, as the relationship between AOD and $PM_{2.5}$ depends on multiple factors, such as total aerosol mass, the vertical distribution of aerosols, and aerosol optical properties that lead to systematic errors in the final product (Ford et al., 2015).

Uncertainties and biases in satellite-derived $PM_{2.5}$ have been addressed by making use of a wide range of observations. This includes correcting biases in the satellite observations of AOD using ground-based observations of AOD from networks such as the NASA Aerosol Robotic Network (AERONET) of sunphotometers (https://aeronet.gsfc.nasa.gov, last accessed 17 November 2022), using space-based observations of aerosol profiles from the active Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) sensor to correct misrepresentation of the vertical distribution of aerosols in chemical transport models (van Donkelaar et al., 2016), and correcting biases in the satellite-derived $PM_{2.5}$ data using surface in situ observations of $PM_{2.5}$ such as is detailed in Section 2.1 (van Donkelaar et al., 2021).

Future satellite instruments that will address some of the issues described above include the geostationary Sentinel-4 mission. The instrument will be in geostationary orbit and so will look over a limited domain over Europe but provide measurements of AOD roughly every daylight hour rather than once per day. None of the satellites used to estimate $PM_{2.5}$ (Table 3.1) were designed at the outset for air quality and health applications, whereas air quality and health are the primary focus of the future low-Earth orbiting Multi-Angle Imager for Aerosols (MAIA) instrument. The design of the instrument and method of acquiring observations will increase accuracy and reliability of health-related aerosol products over target cities around the world, though none of these target cities is in the UK (https://maia.jpl.nasa.gov/, last accessed 16 November 2022).

3.3 Applications and Data Products Available

Satellite-derived $PM_{2.5}$ estimates have been most extensively used to raise awareness of ambient air pollution (Martin et al., 2019, Southerland et al. 2022, van Donkelaar et al., 2019, Hammer et al. 2021), to quantify increased incidence of mortality and morbidity associated with exposure to $PM_{2.5}$ (Tapia et al., 2020, Heft-Neal et al., 2018), and to assess the efficacy of air quality policies (Knibbs et al., 2019; Ciarelli et al., 2019; Schneider et al., 2020). More recently, high-resolution satellite-derived $PM_{2.5}$ data have been used to interpret the effects of COVID-19 lockdown measures on $PM_{2.5}$ pollution (Hammer et al., 2021) and demonstrate that low-income, minority, and marginalized populations are unjustly exposed to elevated levels of $PM_{2.5}$ pollution (Castillo et al., 2021, Nowell et al., 2022).

A wide range of $PM_{2.5}$ products are made available for public use. Those available for coverage over the UK are summarized in Table 3.2. These include regional and global datasets of annual and monthly averages at ~10 km and ~1 km spatial resolutions extending from 1998 to 2019. There are also daily satellite-derived $PM_{2.5}$ data at 1 km spatial resolution over the UK for 2008-2018 (Schneider et al., 2020) and Greater London for 2002–2012 (Belloconi et al., 2016), but these products are not publicly available.

Satellite-derived datasets of individual components of $PM_{2.5}$ (nitrate, sulfate, ammonium, organic aerosol, black carbon, dust) have also been developed, but these rely on models to estimate the relative contribution of individual components to total $PM_{2.5}$, so are additionally subject to model errors in simulating $PM_{2.5}$ composition (van Donkelaar et al., 2019).

Table 3.2. Publicly available satellite-derived $PM_{2.5}$ data products with coverage over the UK

Product	Coverage	Resolution		Time period	Format	Online link to data
		Spatial	Temporal			
WUSTL [a]	Europe [b]	~1 km	Annual, monthly	2001-2018	NetCDF, ASCII	https://sites.wustl.edu/acag/datasets/surface-pm2-5/
WUSTL [a]	Global [c] Europe [b]	~1 km	Annual, monthly	1998-2019	NetCDF	https://wustl.box.com/v/ACAG-V5GL01-GWRPM25
WUSTL [a]	Global [c] Europe [b]	~10 km	Annual, monthly	1998-2019	NetCDF	https://wustl.box.com/v/ACAG-V5GL01-GWRPM25c
DIMAQ [d]	Global	~10 km	Annual	2014 [e] 2016 [f]	CSV	https://www.who.int/data/gho/data/themes/air-pollution/who-modelled-estimates-of-air-pollution-from-particulate-matter

[a] Washington University in St Louis (WUSTL) uses MODIS, MISR and SeaWIFS AOD (Table 4.1). [b] WUSTL version 4 (Hammer et al., 2020). [c] WUSTL version 5 (van Donkelaar et al., 2021). [d] Data Integration Model for Air Quality uses an earlier version of the WUSTL product (van Donkelaar et al., 2016) as input. [e] 2014 data detailed in Shaddick et al. (2018). [f] 2016 data detailed in Shaddick et al. (2017).

3.4 Using Earth observation to diagnose particulate pollution from natural hazards

Natural hazards like wildfires, dust storms and volcanoes are an important source of $PM_{2.5}$ pollution. These hazards are also exacerbated by humans and anthropogenic induced-climate change, such as seasonal shifts in snow melt, changes in land use and land cover, and extreme droughts that alter the frequency, severity and timing of dust storms and wildfires. Earth observations, combined with other tools, have been used to calculate $PM_{2.5}$ abundances and precursor emissions, estimate vertical injection heights of pollutants, and track long-range transport patterns of plumes originating from these hazards. This is because satellites observe multiple gaseous precursors of secondary $PM_{2.5}$ like nitrogen dioxide (NO_2), sulfur dioxide (SO_2), and ammonia (NH_3), the height and quantity of aerosols suspended in the atmosphere in the form of aerosol optical depth (AOD) (Section 3.1, Table 3.1), the timing and location of these events in near real-time, and, for fires and volcanoes, their severity and extent using heat signatures or thermal anomalies. As a result, Earth observation datasets are used to improve the skill of forecasting and early warning systems, such as the Copernicus Atmosphere Monitoring Service (https://atmosphere.copernicus.eu/global-forecast-plots), the European Forest Fire Information System (https://effis.jrc.ec.europa.eu/), European Volcano Early Warning System (http://www.evevolcanoearlywarning.eu/), and the Sand and Dust Storm Warning Advisory and Assessment System (https://public.wmo.int/en/our-mandate/focus-areas/environment/SDS/warnings)

Many research and operational products required for modelling and assessing the environmental impact of natural hazards have been derived with satellite observations. These include emissions of primary $PM_{2.5}$ and its precursors from wildfires using Earth observations of fire counts, burned area, or radiative power in combination with carbon balance models and emission factors (the amount of air pollutant emitted per kg fuel or carbon burned). Inventories widely used in Earth System and Chemical Transport Models include the CAMS Global Fire Assimilation System (https://atmosphere.copernicus.eu/global-fire-emissions), the Quick Fire Emissions Dataset (https://gmao.gsfc.nasa.gov/research/science_snapshots/global_fire_emissions.php), the Global Fire Emissions Dataset (https://www.globalfiredata.org/), and the Fire INventory for NCAR (https://www2.acom.ucar.edu/modeling/finn-fire-inventory-ncar). These also include emissions from fires set intentionally for land management or agricultural practices.

Earth observations are also used to trace the injection heights and long-range transport of plumes from wildfires, dust storms and volcanoes. Derivation of the vertical extent or injection height of plumes has evolved from manual inspection for the publicly available NASA injection plume height (https://misr.jpl.nasa.gov/get-data/misr-plume-height-project-2/) to recent automation using machine learning techniques (Section 5.2) (https://atmos.eoc.dlr.de/so2-lh/). The long-range transport of volcanic plumes has been tracked since the 1990s using UV/visible instrument measurements of atmospheric column abundances of SO_2 with spatial resolutions of ~40-100s km in the 1990s to <5 km today (as demonstrated in Figure 3.2). Dust storms are tracked along distances extending from their origin in the Sahara Desert to as far as northern Europe and the Amazon using satellite

observations of AOD in combination with forward and back-trajectory models and local ground-based measurements.

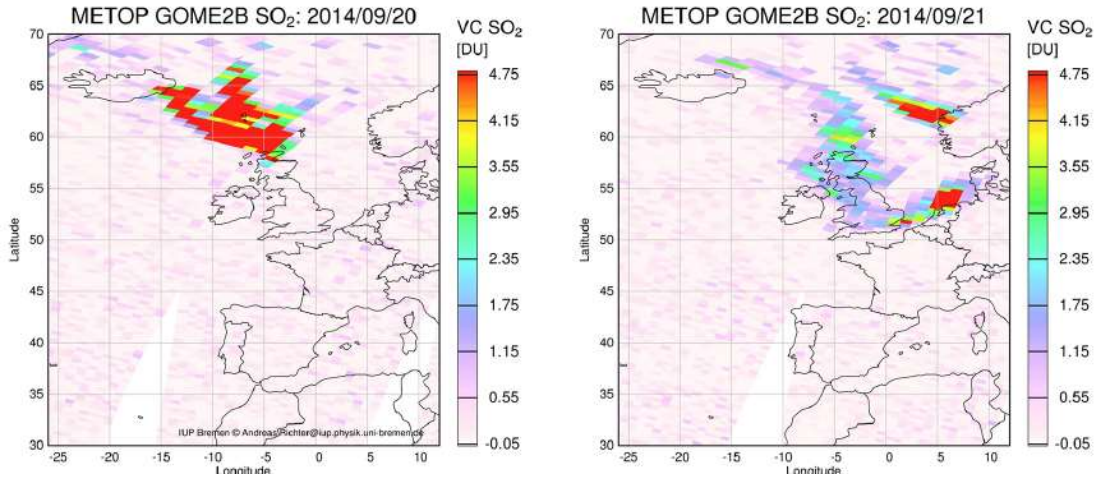

Figure 3.2. Satellite observations track a plume from the eruption of the Holuhraun volcano in Iceland in 2014 as it migrates over the UK. Images show Global Ozone Monitoring Instrument 2nd generation (GOME-2) swaths of sulfur dioxide (SO_2) column concentrations in Dobson Units (1 DU = 2.69 x 10^{16} molecules cm^{-2}). Image from Twigg et al. (2016).

Chapter 4 - Measurement techniques for quantifying PM emissions

4.1 Introduction and Context

Whilst ambient concentrations influence human exposure to PM and other air pollutants, it is the emissions which most policies target to lower concentrations, often through regulation and controls. Emission estimates also usually form the starting point for model predictions of air quality as well as for estimating the likely air quality improvements that can be expected for certain emissions control measures. Summarised here are the methods used to quantify sources and represent them in inventories and models.

Typically emissions of pollutants to air are estimated in bottom-up emission inventories on the basis of emission factors, often measured under laboratory conditions, coupled with activity figures (i.e. vehicle kilometres driven). In the UK, the official emissions are compiled via the UK National Atmospheric Emissions Inventory (NAEI; https://naei.beis.gov.uk/), although local inventories (e.g. London Atmospheric Emissions Inventory; LAEI; https://data.london.gov.uk/dataset/london-atmospheric-emissions-inventory--laei--2019) and research approaches (e.g. DUKEMS; http://www.uk-ems.org.uk/) also exist. The emission factors are based on measurements e.g. under controlled laboratory conditions or point sources.

There are several different measurement techniques that can be used for measuring PM emissions, for example, some methodologies include the use of a dilution chamber, and others do not. This gives rise to a complication, because not all measurement techniques are equivalent in their definition of PM. Emissions measurement techniques primarily fall into two categories - those which include the condensable PM component in the resulting concentration measurement, and those which do not (measuring filterable PM only). The difference between methods that include/exclude the condensable PM component can be substantial. Solid fuel use in residential appliances is an example of a source where inclusion of the condensable component can increase the estimated PM emissions by a factor of five or more. So, it is important to know the exact definition of the PM that results from a given measurement technique.

Historically, emission measurements made from large stationary sources, such as power stations and large industrial installations, have primarily used methods that exclude the condensable component. In contrast, the majority of methods used for measuring PM emissions from road transport sources include the condensable component. For emissions from residential combustion sources, the picture is rather mixed. This has led to inconsistencies between the emission factors that are used in emission inventories for different source categories. Furthermore, it has not been possible to determine whether historical measurement studies, sometimes undertaken many decades ago, give emissions data that includes or excludes the condensable component of PM. So in addition to inconsistencies within emissions inventories, there can be a lack of transparency regarding what national emission estimates actually represent.

To address this issue, the guidance in the EMEP/EEA Air Pollutant Emissions Inventory Guidebook is being standardised so that the condensable component of PM is:

- Included in emission estimates for residential combustion, road transport, and other mobile sources.
- Excluded for large point sources.

Whilst this doesn't address some of the inconsistencies within emissions inventories, for the users of the emissions inventory data it does provide improved transparency regarding the definitions of PM that are being reported in official national emissions inventories.

4.2 Point sources

Emissions from regulated point sources are subject to strict controls. Point sources including from industry and energy generation remain potentially significant sources of PM emission. Monitoring and testing of these emissions is important to ensure compliance with permits and regulations and to identify and quantify emerging any new sources or types of air pollution.

4.2.1 PM from stationary sources

Stationary sources range in scope from small (residential) appliances to large industrial processes. The nature and characteristics of PM emissions from these sources are dependent on the characteristics of the source and can differ significantly. PM emissions include material which can be solid or liquid at the point of release and also PM formed after release due to later physical and chemical processes. For example, a mineral grinding process may produce only solid material with a narrow range of chemical composition, but residential solid fuel combustion can give rise to PM which comprises a range of chemical components some of which are solid and others which give rise to a significant 'condensable' PM fraction.

4.2.2 PM Measurement methods

In general, the methods applied for emission monitoring from stationary sources have evolved to meet local and regional regulatory requirements and it is important to note that these may not align with emission monitoring in other regions, with air quality monitoring or, with the requirements of air quality models. Measurement methods include International, European and national Standards which exist for PM and PM size fractions but measurement approaches differ for different types of sources. Emission monitoring methods for regulatory compliance are not focussed on $PM_{2.5}$ or other specific particle size fractions, they are mainly concerned with 'total' PM.

4.2.3 PM measurements in industry

For industrial processes, regulatory authorities publish guidance on emission monitoring for assessing compliance with permit requirements. For example, guidance for UK 'Part A'

activities for acceptable periodic emission monitoring techniques[4] indicates that periodic PM measurements are to BS EN 13284-1. The BS EN 13284-1 Standard[5] provides an extractive, gravimetric isokinetic sampling approach for measurement of 'dust' with collection of PM material on a filter at about 160°C and from deposits recovered from surfaces of the sampling equipment between the sample inlet and the filter.

BS EN 13284-1 was developed for application at large combustion and waste incineration plant which have emission limits specified for 'dust'. Note that the EU Directives[6] which have set maximum dust ELVs for such plant do not define 'dust' – it is defined by the measurement method.

Similar 'filterable PM' approaches are adopted in other parts of the world; there is recognition in some regions that a 'condensable' fraction can be important and a filterable PM measurement approach can be extended to include determination of the condensable component. For example, in the USA, US Environmental Protection Agency (USEPA) filterable methods can be extended to include condensable PM (USEPA Method 202)[7].

In the UK, the regulatory method for condensable PM is BS ISO 25597:2013 which employs a dilution technique to measure total (filterable and condensable) PM_{10} and $PM_{2.5}$. However, in the UK (and Europe) it is uncommon for measurement of a condensable PM fraction at an industrial stack but it can be a consideration for some activities (stationary engines, high temperature processes and processes with emission of semi-volatile components).

Continuous PM measurement systems are frequently employed on industrial stacks and vents and these apply a range of measurement techniques (Nicklin and Darabkhani, 2021). Continuous monitoring systems provide quantitative or indicative emission data. Quantitative continuous systems are calibrated by comparison with multiple reference measurements (typically using EN 13284-1 in UK and Europe). Consequently, calibration of the continuous monitor depends on what is measured by the reference method.

However, as emission controls on industrial plant have improved (resulting in lower emission concentrations), concerns have been raised (Coleman et al., 2019; Antonsson et al., 2021) about the suitability of current measurement methods (including the EN13284-1 Standard) for assessing compliance with emission limit values set to align with Best Available Techniques (BAT).

One area of emerging interest is in the centralised gathering of continuous monitoring data for PM (and other pollutants) from large sources in (near) real-time. Regulatory authorities in South Korea, People's Republic of China and India have been using remote access to continuous monitoring data on power plant and other large sources for several years to aid

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[4] Information here https://www.gov.uk/government/publications/monitoring-stack-emissions-techniques-and-standards-for-periodic-monitoring/monitoring-stack-emissions-techniques-and-standards-for-periodic-monitoring

[5] BS EN 13284-1:2017 - Stationary source emissions. Determination of low range mass concentration of dust - Manual gravimetric method.

[6] Maximum emission limits for large combustion and waste incineration plant in Directive 2010/75/EU on Industrial Emissions (IED) and which superseded earlier directives on these types of plant : https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32010L0075&from=EN

[7] Information here https://www.epa.gov/sites/default/files/2017-08/documents/method_202.pdf

supervision of activities at a regional and national level. Access to such data sources can allow improved resources for, for example, source apportionment and incident monitoring.

4.2.4 PM measurements for residential solid fuel sources

Residential solid fuel appliances present a number of challenges for PM measurement – small ducts, relatively high emission concentrations, varying operation (particularly for manually-controlled, batch-fired, natural draught appliances). Different measurement approaches are applied which adopt:

- a range of appliance operating protocols for formal type-testing and for 'real world' emissions and,
- measurement techniques which measure different aspects of PM.

There is wide variation in the operating protocols applied (the range of operation tested and number of tests) and the variability of the measurement techniques for solid fuel appliances is relatively high[8].

Regulatory measurement techniques fall into one of the following groups:

- Sampling of undiluted flue gas using a filterable PM technique (heated filter)
- Full flow dilution tunnel - sampling of diluted flue gas (filterable and condensable PM)
- Electrostatic precipitator (ESP) – all flue gas is passed through an ESP (UK only)
- Sampling of undiluted flue gas using a filterable PM technique coupled with collection of condensable PM

In addition, partial flow dilution (where a sample of flue gas is diluted within the sampling equipment) and a variety of continuous and semi-continuous methods are applied by researchers to determine PM emissions, particle number and particle sizes.

Consequently, there can be a wide variation in what is reported as PM from a residential solid fuel appliance. The Ecodesign Regulation for solid fuel 'local space heaters'[9] includes four test methods for PM (aligning to the German, Norway and UK national test methods) with different emission limits in recognition of the different national testing requirements (and the absence of a common EN methodology at that time). These testing requirements represent a narrow range of appliance operation.

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[8] Assessment of particulate emissions from wood log and wood pellet heating appliances. Report by Ricardo Energy & Environment for Defra, 2017. Available here : https://uk-air.defra.gov.uk/assets/documents/reports/cat07/1801291425_170201_Defra_NAEI_appliance_testing_summary_Issue1_Final_copy.pdf

[9] Commission Regulation (EU) 2015/1188 of 28 April 2015 implementing Directive 2009/125/EC of the European Parliament and of the Council with regard to ecodesign requirements for local space heaters. Available here : https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32015R1188&from=EN

Recent work in Europe has focussed on a common heated filter test method which has been incorporated into the revised EN 16510-1 Standard for residential solid fuel appliances. The 'EN-PME' project[10] adopted a heated filter approach with sampling and measurement of (filterable) PM and total hydrocarbons at a common temperature (180°C) which may become applicable for type-testing of solid fuel appliances in future. A further EU-funded project is underway to investigate testing approaches for 'real life' emissions[11].

In conclusion, there are established measurement techniques for large industry sources but the suitability of current measurement Standards for decreasing emission concentrations is becoming an issue for periodic sampling methods and for calibration of continuous monitoring systems used for compliance assessment.

The measurement of PM from residential solid fuel appliances is undertaken for assessing regulatory compliance and wider research into environmental impacts. The test protocols and measurement techniques used for regulatory compliance provide different measures of PM and represent a narrow range of typical appliance use.

4.3 Vehicle sources

The measurement of PM and other emissions from road vehicles is challenging given the number of individual vehicles in the UK fleet and the myriad factors that affect emissions such as emissions control technology, fuel, vehicle type, road conditions and driving styles. The 'dieselgate' scandal highlighted the insight that could be gained from direct measurements of real world in-service vehicles, rather than simply relying on data obtained under controlled conditions.

Almost all diesel vehicles in the UK are fitted with Diesel Particulate Filters (DPF) that have been shown to be highly effective at reducing particle mass and number [AQEG 2021, Exhaust Emission for Road Transport report]. However, the measurement of in-use ('real-world') emissions of PM from vehicles is valuable in providing evidence of emissions control performance under a wider range of conditions. It is important, for example, to ensure that emission control systems remain effective for the operational life of a vehicle and that any degradation effects do not result in significantly increased emissions. Additionally, the ability to detect whether a vehicle has been tampered (such as removing a DPF) or has for some other reason high emissions is also a good reason to measure individual vehicles. Ideally, such measurements would be made of hundreds or many thousands of vehicles to understand emission distributions.

One promising approach to better understand in-use PM emissions is the fast-response measurement of specific particle metrics such as BC and PN. Particle mass emissions from

[10] Determination of particulate matter emissions from solid biomass fuel burning appliances and boilers –Proposal for a common European test method. Isaline Fraboulet, Institut National de l'Environnement Industriel et des Risques (INERIS). 2015. Available here : https://www.researchgate.net/publication/292977162_EN_PME_TEST_Project_Position_paper_December_2015_Determination_of_particulate_matter_emissions_from_solid_biomass_fuel_burning_appliances_and_boilers_-_Proposal_for_a_common_European_test_method

[11] Harmonizing reliable test procedures representing real-LIFE air pollution from solid fuel heating appliances 'Real-LIFE Emissions' Project website : https://sites.uef.fi/real-life-emissions/

vehicles are generally very low that it is difficult or impossible to measure mass emissions in a practical way. Recent developments as part of the EU CARES project have been to develop fast response BC and PN instruments capable of making measurements of individual vehicles (https://cares-project.eu). These measurements are made in the ambient atmosphere close to the road of passing vehicles. Simultaneous measurements are also made of CO_2 to associate BC or PN to the combustion of fuel. A camera is used to record the number plate of a passing vehicle, which can be used to query vehicle information databases to obtain information such as fuel type, Euro class, vehicle manufacturer etc. This approach is analogous to vehicle emission remote sensing, but additionally requires a plume to be sampled directly, which can be affected by atmospheric dispersion.

Two main technologies have been developed and tested for vehicle plume measurement: the photoacoustic measurement of BC, and PN measurement using a diffusion charging technique. While commercial instruments are available to make such measurements, the development through the CARES project focused on making such instruments practical for roadside use e.g. fast response, battery-powered, and potentially practical for cities to adopt such methods to understand vehicle fleet emissions in more detail. The instruments have been developed at the Technical University of Graz.

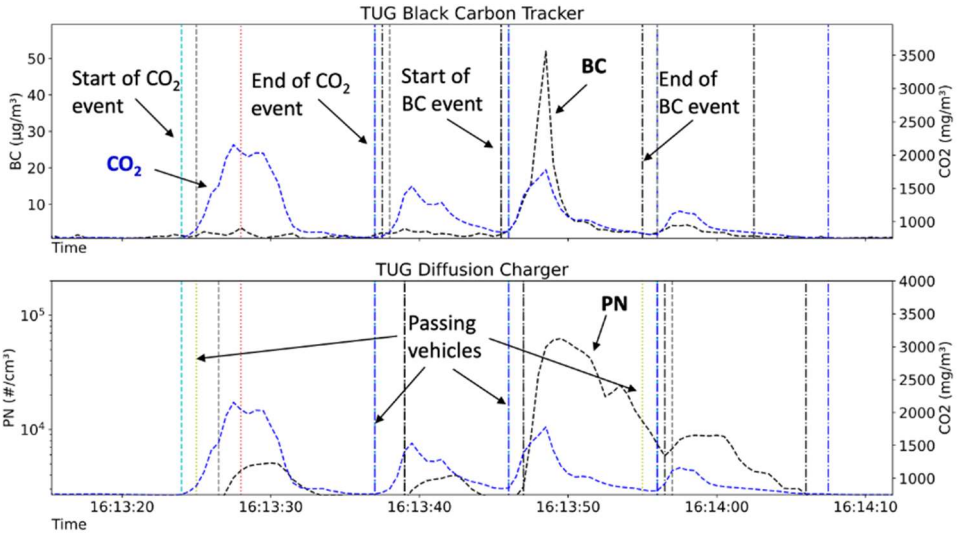


Figure 4.1 1-Hz measurement of individual vehicle plumes for CO_2, BC and PN from measurements as part of the EU CARES project.

Figure 4.1 shows an example time series of CO_2, BC and PN, highlighting individual vehicle plumes. Developments have also been made to identify discrete vehicle plumes of BC/PN and associate the concentrations to CO_2 to provide fuel-based emission factors.

4.4 Area fluxes

Tower-based micrometeorological flux measurements provide one of the few methods that provide independent assessment of emission inventories. Flux methods can detect emission changes with time, and for diffuse sources spread over scales of hectares to square kilometres. These measurements are complex to make however,

often using non-standard very fast response instrumentation and require substantial numerical post-processing.

4.4.1 Introduction to micrometeorological flux measurements

Pollution emissions can be inferred from concentrations, measured at ground level e.g. in the AQ monitoring networks, on tall towers or for some gases, sensed remotely by satellite. However, for PM this approach requires numerical atmospheric chemistry and transport models to account for the dispersion of the pollutants in response to meteorology, deposition and, in the case of reactive aerosol components, their chemical sources and sinks; and these models have their own intrinsic uncertainties.

By contrast, micrometeorological flux measurement approaches in principle allow emissions to be measured more directly. The most common and direct technique, eddy-covariance, derives the vertical flux through an imaginary horizontal plain at the measurement height well above the surface using a combination of fast-response (typically 10 Hz, depending on measurement height) ultrasonic anemometers and concentration measurements on towers (see Figure 4.2). This flux can then be linked to the surface flux (emission or deposition, or the combination of both processes) if a number of conditions are met. The flux measured with this approach is representative for an area of origin ("flux footprint") which typically extends from several 100s of square metres to a few square kilometres, growing with increasing measurement height, and the position of which changes continuously with meteorological conditions, especially wind direction and atmospheric stability.

Figure 4.2 Eddy-covariance instrument and inlet above the city of Edinburgh consisting of an ultrasonic anemometer, a fast-response hygrometer and several inlets leading to fast-response particle analysers.

4.4.2 Challenges associated with measuring particle fluxes

Eddy-covariance derives the flux as the covariance between concentration and vertical wind velocity, typically calculated from 10 Hz data over a 30-minute segment. Whilst some gases such as CO_2 can readily be measured at 10 Hz, the requirement for fast-response measurements still poses a measurement challenge for many pollutants including PM. As a

result, the measurement of PM emission fluxes has until now been a relatively niche endeavour, combining an extension of the eddy-covariance approach to particles (more often particle number than particle mass) and to source areas such as cities. Because air contains far fewer particles than it contains gas molecules, a key challenge specific to PM fluxes is to make fast measurement of particle concentration at high signal-to-noise. This is particularly true for larger particles where very few particles carry much of the mass and mass flux. This requires high sample flowrates for coarse particles which then leads to counting problems of the smaller particles. Thus, several instruments targeting different sizes may be required to cover the full size range. Nevertheless, mass fluxes of $PM_{2.5}$ and PM_{10} derived from particle number flux measurements remain difficult to measure because the uncertainty in the flux of the larger particles tends to dominate the total mass flux. In addition, to avoid disruption of the air flow at the point of measurement, particle flux measurements require inlet tubes from the anemometer to the particle instrument. For fluctuations to be maintained during transport down the tube, the flow needs to be turbulent, which can increase diffusion losses for particles < 50 nm and losses due to impaction and interception increase for particles > 1 µm.

It should be noted that other micrometeorological flux measurement approaches do exist. In particular, the aerodynamic gradient method and the modified Bowen ratio approach derive fluxes from the difference of concentrations measured at different heights and avoids the need for fast-response measurements. However, gradient approaches are even more sensitive to heterogeneities in the source and therefore less suited to measurements above complex source regions such as urban areas.

4.4.3 Examples of PM flux measurements

Most measurements of PM emissions have been made for total particle number by condensation particle counter (CPC) - defined as the size-range detected by the instrument. Whilst this achieves good measurement statistics the total particle number flux cannot easily be related to standard health metrics. Several studies have measured particle number fluxes using optical or aerodynamic particle counters but most have confined themselves to reporting these as number fluxes. The conversion introduces uncertainty due to assumptions with respect to shape, refractory index and density of the particles. Some instruments deploying such conversion have demonstrated equivalence with the reference methods, including the FIDAS now deployed in the UK AURN network (c.f. Chapter 2).

Some studies have converted their measurements to PM_1 or $PM_{2.5}$ particle mass above urban areas (Contini et al., 2012; Deventer et al., 2015; Donateo et al., 2019) and above vegetation (Donateo et al., 2006; Donateo and Contini, 2014; Pallozzi et al., 2020). One reason for not converting is that the mass flux is subject to a much larger uncertainty because it is dominated by the large particles for which counting statistics are poorer. Another reason is that unlike the (traffic-related) Aitken mode sized particles (0.01 to 0.1 µm), the accumulation mode particles (0.1 to 1 µm), and therefore also $PM_{2.5}$ mass, often show periods of apparent deposition over urban areas, contrary to expectation, especially during summer daytime. Explanations have included deposition from background aerosol, downward mixing of polluted air masses from aloft. However, composition resolved measurements are starting to provide a more robust explanation to this observation.

Flux measurements of particle chemical components are still rare but can yield additional information. A first EC for sulfate was deployed in the 1980s to measure aerosol deposition to vegetation (Wesely et al., 1985). More recently, flux measurement systems based on aerosol mass spectrometers (AMS) have been developed (Farmer et al., 2011) and also applied to urban flux measurements (Table 4.1). This measurement provides fluxes of non-refractory aerosol components within PM_1, including, in its latest version based on a time-of-flight mass spectrometer, information for factor analysis of the organic PM_1 component. A $PM_{2.5}$ inlet (lens) is now available for the AMS (Xu et al., 2017) but this has not yet been tested for flux response and is unlikely to deliver equivalence with $PM_{2.5}$ reference methods because of the omission of refractory aerosol components such as sea salt and black carbon (BC). Instead, an eddy-covariance system for BC has been developed based on a Single Particle Soot Photometer (SP2) (Emerson et al., 2018) and also applied over urban areas (Table 4.1). Although the AMS detects many ions per particle, this does not fully alleviate the problems associated with counting statistics, especially for larger particles, because these ions are not independent from each other.

Table 4.1 Overview of instruments suitable for measurements of particle flux measurements in the urban environment and existing applications.

Instrument	Entity measured		Studies
Condensation particle counter, CPC	Total number	Total particle number within a typical size range of 10 – 1000 nm	(Dorsey et al., 2002; Held and Klemm, 2006; Järvi et al., 2009; Martin et al., 2009; Contini et al., 2012)
Optical particle spectrometer, UHSAS, Grimm 1.109	Size-resolved number	Size resolved particle fluxes sized by optical diameter, e.g. 0.06 to 1 μm (UHSAS); 0.25 to 2.5 μm (Grimm 1.108)	(Deventer et al., 2011; Vogt et al., 2011a, b; Deventer et al., 2015; Donateo et al., 2019)
Electronic Low Pressure Impactor (ELPI)		Size resolved particle fluxes sized by aerodynamic diameter	(Schmidt and Klemm, 2008; Deventer et al., 2015; Deventer et al., 2018)
Aerodynamic Particle Sizer (APS, TSI)		Size resolved particle fluxes, sized by aerodynamic diameter 0.5 to 10 μm	(Nemitz et al., 2000a; Nemitz et al., 2000b)
Engine exhaust particle sizer (EEPS, TSI)		Size resolved particle fluxes, sized by electro mobility diameter 5.6 to 560 nm	(Straaten and Weber, 2021)
Aerosol Mass Spectrometer	Chemically resolved mass	Mass flux of PM_1 chemical components	(Thomas, 2007; Nemitz et al., 2008; Harrison et al., 2012; Zalakeviciute et al., 2012)
Single particle soot spectrometer (SP2)		Mass flux of BC	(Joshi, 2021; Joshi et al., 2021)

There are other existing and emerging technologies that may have the potential to report sufficiently fast aerosol concentrations at sufficient signal/noise levels and which have not yet been tested for flux measurements. This includes photoacoustic sensors for black carbon, fast nephelometers which can provide a proxy for $PM_{2.5}$ and other fast

instrumentation primarily aimed at process emission measurements (e.g. Dekati eFilter; DMS500 Particulate Analzer, Cambustion). Potentially, such instrumentation would need to be optimised for flux measurements. This may include improving the data reporting intervals, but, crucially, the internal response time to a concentration change.

4.4.4 Interpretation and artefacts

Even where (urban) EC fluxes can be measured well, these require careful interpretation. In reality in any environment with emissions, such as urban areas, deposition of particles from the background occurs alongside those emissions. The flux measurement therefore reflects the net effect of the emission and deposition components. Indeed, in less dense urban areas, particle fluxes have often been found to be bi-directional, with both net emission and deposition occurring, either in relation to flux footprints changing with wind direction, in relation to diurnal and seasonal variability or in relation to particle size. If the aim is to quantify emissions, either situations need to be chosen where the emission greatly exceeds deposition so that deposition can be ignored, or a correction needs to be applied to correct for the deposition. Usually, in modelling situations, the deposition flux is estimated as the product of concentrations and the deposition velocity. Deposition velocities vary with particle size, meteorology (mainly turbulence) and the morphology of the underlying surface and are particularly uncertain for urban matrices consisting of building, roads and green spaces.

Measured fluxes reflect the surface flux only if a number of conditions are met. Ideally, the surface strength would be homogenous over the flux footprint and beyond, concentrations should not change with time, fluxes should be stationary, turbulence should be well developed and the pollutant (in this case particles) should be stable chemically and physically. Some of these effects (non-stationary conditions and poorly developed turbulence) require individual measurement periods to be removed from the dataset. Storage errors on fluxes caused by changing concentrations can be, at least to some degree, corrected for. Others, such as advection errors associated with surface heterogeneity are difficult to estimate. This results in some uncertainty, which is likely larger for individual 30-minute flux measurements than for long-term averages.

Particles continuously evolve chemically and physically and this creates a significant problem for some particle flux measurements and their interpretation. Particles are known to change size as they take up or lose water or semi-volatile chemical compounds such as ammonium nitrate, which can lead to very strong apparent deposition fluxes of these compounds. During warm days, when the temperature near the ground is increased, this can lead to the same particles being on average smaller near the ground than aloft, creating an apparent flux which can even lead to sign reversal (Nemitz and Sutton, 2004; Nemitz et al., 2009). This affects fluxes of the evaporating compounds (e.g. nitrate and ammonium), size-segregated number fluxes and inferred mass fluxes. Aerosol evaporation near the ground therefore provides the more likely explanation for the observation of downward fluxes of accumulation mode particles even over dense urban surfaces. By contrast, mass fluxes of non-volatile compounds tend to be unaffected as well as total number fluxes as long as particles stay within the size-range of the instrument. Thus, in most cases, chemically resolved mass fluxes are easier to interpret than size-segregated particle number fluxes.

Chapter 5 - Data interpretation and experimental design

5.1 Source apportionment

The term *source apportionment* refers to quantifying the contributions of individual sources to overall observed ambient concentrations of a pollutant. This is often highly policy-relevant and can be applied to any pollutant. A very common application is to airborne particulate matter. The diverse chemical composition of PM allows the application of receptor modelling methods. This type of analysis can be used to help prioritise air pollution sources for intervention, or to inform exposure and health studies.

Source apportionment is a mature topic, but one which constantly develops and improves, providing opportunities for application to new locations or situations. Although being rather easy to use, source apportionment models require considerable skill to generate good results. That said, efforts are currently underway within the research community to develop automated tools capable of issuing real-time source apportionment data, based on PM composition from instruments such as the ACSM, XRF and Aethalometer (see Chapter 2).

There are two main categories of source apportionment model:

- Those based upon dispersion or chemistry-transport models
- Those based upon receptor models.

5.1.1 Dispersion-based models

If source strengths are known, dispersion models can predict ground-level concentrations of primary pollutants with considerable skill. For pollutants treated as unreactive in the atmosphere (which includes most common pollutants in local scale models), concentrations arising from different sources can be treated as additive, and the dispersion model will give a reliable estimate of the contributions of all specified sources within the model domain (Laupsa et al., 2009). Long-distance contributions can only be assessed by measuring concentrations at the boundaries of the model domain. These can also be estimated by use of larger scale chemistry-transport models which also take account of chemical processes causing formation or loss of pollutants and predict secondary pollutants. Use of such models has to account for non-linearities between emissions and airborne concentrations, and simple methods (e.g. the “brute force” method which simply turns sources on and off) may not always be reliable in estimating source contributions (Burr and Zhang, 2011). There is evidence that modelling effects of changes on annual averages can however be reasonable (Thunis et al. 2015).

Dispersion modelling can also be used to estimate the emission source strength directly from ambient concentration measurements by inverting the calculations. The principle of inverse dispersion modelling is mature (e.g. Rao, 2007) and has been widely used to calculate emissions from spatially-discrete sources (e.g. Thoma et al., 2009, Thoma and

Squier, 2014). EN 176828:2022 provides a method for its use in quantifying VOC emissions from industrial sources. Inverse dispersion modelling has also been used to quantify emissions from spatially disaggregated sources using networks of monitors (Carruthers et al., 2019).

Inverse methods differ in the complexity of the dispersion assumptions, with 1-, 2-, and 3-dimensional Gaussian profiles all commonly used. Methods also differ in how emissions rates are defined to match the measurements, ranging from relatively simple linear least squares functions to more complex Bayesian frameworks (e.g. Lushi and Stockie, 2010, Hosseini and Stockie, 2016). The different methods usually reflect the nature of the emission sources and the available measurements; these are not independent since while isolated downwind measurements might inform emissions estimates for a single ground-level source, more comprehensive measurements are needed to understand more spatially- and temporally-complex sources.

Inverse dispersion modelling typically uses all suitable measurements to characterise emissions. It is thus unsurprising that model predictions match those measurements. Significant care is needed to ensure that the emissions source term is not used to compensate for other uncertainties.

5.1.2 Receptor modelling methods

The main utility of receptor models is in the source apportionment of particulate matter for use in air quality management. All methods depend upon the fact that each source has a characteristic chemical composition, and that the composition of the ambient particulate matter is a linear sum of the individual source contributions (Watson et al., 2008). Hence:

$$C_i = \sum^j f_{i.} g_i \qquad (1)$$

in which C_i is the mass concentration of component *i* in ambient air, f_i is the fractional contribution of component *i* in source *j*, and g_i is the fractional mass contribution of particles from source *j*. There are two main approaches to the utilisation of this principle in receptor modelling. Both methods require a substantial dataset of multi-component analyses of airborne particles.

5.1.3 Chemical Mass Balance (CMB)

The CMB method requires knowledge of the chemical composition of each contributing primary source of particles. The model uses a least squares fitting process to derive a best fit to equation 1 for each chemical component. This can, in theory, be conducted on a single sample, but in practice better and more representative results are obtained with multiple samples. The widely used USEPA version of CMB focusses on a small number of characteristic tracers for each source, and in this context the use of organic source tracers has become commonplace. These include compounds such as levoglucosan as a tracer of biomass burning, hopanes and steranes as tracers of fossil fuel combustion, alkanes and PAH to differentiate combustion sources and organic acids as tracers of cooking aerosol and secondary aerosol. The CMB method is typically applied to the organic component of airborne particles, with metallic tracers added to assist in quantifying soil dust. Secondary

inorganic components, which typically comprise a large fraction of the aerosol are added subsequently. The main weakness of CMB is the requirement for input data in the form of multi-component source profiles. This assumes that all sources affecting a site are known (although the model does report a residual if not accounting for 100% of mass), and that all source profiles are a reliable reflection of the source considered. This can be problematic, as many source data are old, some source profiles are determined by analytical methods yielding systematically different results to those used for ambient air analysis, and emission source profiles may be affected by the operational practices for a process in a given country. A case in point is the road traffic profile, which can be determined either from roadside measurements (as a fleet composite) or from laboratory studies of individual vehicles, which allows discrimination of petrol and diesel emissions. As vehicle technologies are constantly changing and the prevailing standards differ by country, selecting CMB input data for a specific location can be challenging.

5.1.4 Multivariate statistical models

These are very commonly employed in local source apportionment studies, most workers using the user-friendly USEPA version of Positive Matrix Factorization (PMF) which conveniently provides output in the form of a graphical user interface. PMF has largely superseded other factor analysis model approaches such as the much simpler Principal Component Analysis (PCA) or 'Unmix' algorithm, as it uses a weighted fitting approach and is constrained not to output negative factors (Reff et al., 2007). The main difference from CMB is that PMF (and similar models) do not require an input of source profiles. PMF solves equation 1 using a weighted approach which assigns most weight to those chemical constituents whose measurements have the least uncertainty. This is achieved by inputting an uncertainty matrix expressing the uncertainty in each analytical variable input to the model. Typically, about 12 – 20 chemical variables are input, and may be elements, ions or molecular species. It is also possible to include other variables such as trace gases or meteorological variables in the inputs to the model, and these can assist in associating factors with specific sources. As a rule of thumb, the number of samples should exceed the number of factors (explanatory variables) by at least threefold, or the model can be very unstable.

Expressed in matrix form the PMF model is:

$$X = G \bullet F + E \qquad (2a)$$

or, in index notation:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (2b)$$

where x_{ij} is the concentration of species j measured on sample i, p is the number of factors contributing to the samples, f_{kj} is the concentration of species j in factor profile k, g_{ik} is the relative contribution of factor k to sample i, and e_{ij} is error of the PMF model for the species j measured on sample i. The model seeks to find values of g_{ik} and f_{kj}, that best reproduce x_{ij}. The values of g_{ik} and f_{kj} are varied to achieve a minimum value of Q, which is defined as:

$$Q = \sum_{i=1}^{n} \quad \sum_{j=1}^{m} \quad \left(\frac{e_{ij}}{\sigma_{ij}}\right)^2 \qquad (3)$$

where σ_{IJ} is the uncertainty of the *j* th species concentration in sample *i*, *n* is the number of samples, and *m* is the number of species. This ensures that the species with the smallest uncertainty (σ_{IJ}) play the greatest role in minimising Q.

The sole inputs to PMF are the chemical composition variables and associated uncertainties, with no need to input source profiles. The program recognises associations between the input chemical species, and outputs an F-matrix, expressing the chemical profile of each of typically four to twelve source-relate factors and a G-matrix describing the temporal pattern of each factor. The Explained Variation also describes the amount of the variance in a given chemical species accounted for by each factor. From an examination of these outputs, it is usually possible to identify the source responsible for a factor; for example, a factor rich in iron, accounting for a large percentage of the variance in barium and copper, peaking in the morning and evening rush hours is likely to be due to brake wear particles. It is customary to test a number of different factor solutions with varying fitting parameters. Judgement is then needed to determine which solution best describes the sources. This is unlikely to be the solution with most factors as "factor splitting" can create spurious factors. One option, which can be exploited more fully in the closely related two dimensional Multi-linear Engine (ME2) is to specify one or more source profiles or time series as inputs to the model to "pull" the model towards these pre-determined profiles of sources known to affect the locality. This approach in effect acts as a hybrid between PMF and CMB, allowing a user-defined amount of a priori knowledge of sources to be included in the receptor model solution.

Another application of PMF and ME2 is in the analysis of mass spectral data collected by Aerosol Mass Spectrometer and Aerosol Chemical Speciation Monitor instruments (Crippa et al., 2014). In this case, the factors are mass spectral peak intensities (F-matrices) associated with different primary and secondary components of the aerosol. Factors which are commonly extracted include those associated with biomass burning, road traffic and various forms of secondary aerosol. The factor profiles associated with secondary organic aerosol are known to vary according to season at a given location, which while a reflection of the chemical complexity of this fraction, can confound the algorithm. To mitigate this, the 'rolling' version of ME2 can be used to perform the analysis on an ensemble of discrete overlaid periods and combine the results, in effect allowing the profiles to vary with time (Canonaco et al., 2021).

5.1.5 Back trajectory modelling

There is a wide range of methods used to determine likely precursor source origins based on back trajectory analysis. The main idea behind these techniques is to associate a back trajectory (typically following an air parcel back in time for 72 or 96 hours) with a concentration measured at a monitoring site i.e. a receptor. The approach requires the use of meteorological models such as Hysplit (from the US EPA), Flexpart (developed at NILU) and NAME (the UK Met Office) to track the movement of air over many hours. By considering many back trajectories e.g. air mass arrivals every hour or few hours over a long period of months to years, an understanding can be developed of the source origins of different particle measurements – for example, total $PM_{2.5}$ or compositional information such

as particulate nitrate and sulphate. These techniques are mostly appropriate for species that include a significant fraction of secondary pollutants that take hours to days to form in the atmosphere and are therefore useful for particle measurements.

An example of this type of analysis is shown in Figure 5.1 for $PM_{2.5}$ concentrations measured at the North Kensington site in London. The plot shows the most probable source origins contributing to $PM_{2.5}$ concentrations at the North Kensington site for 2020 and highlights the importance of emissions that originate in the Benelux countries and eastern parts of Europe.

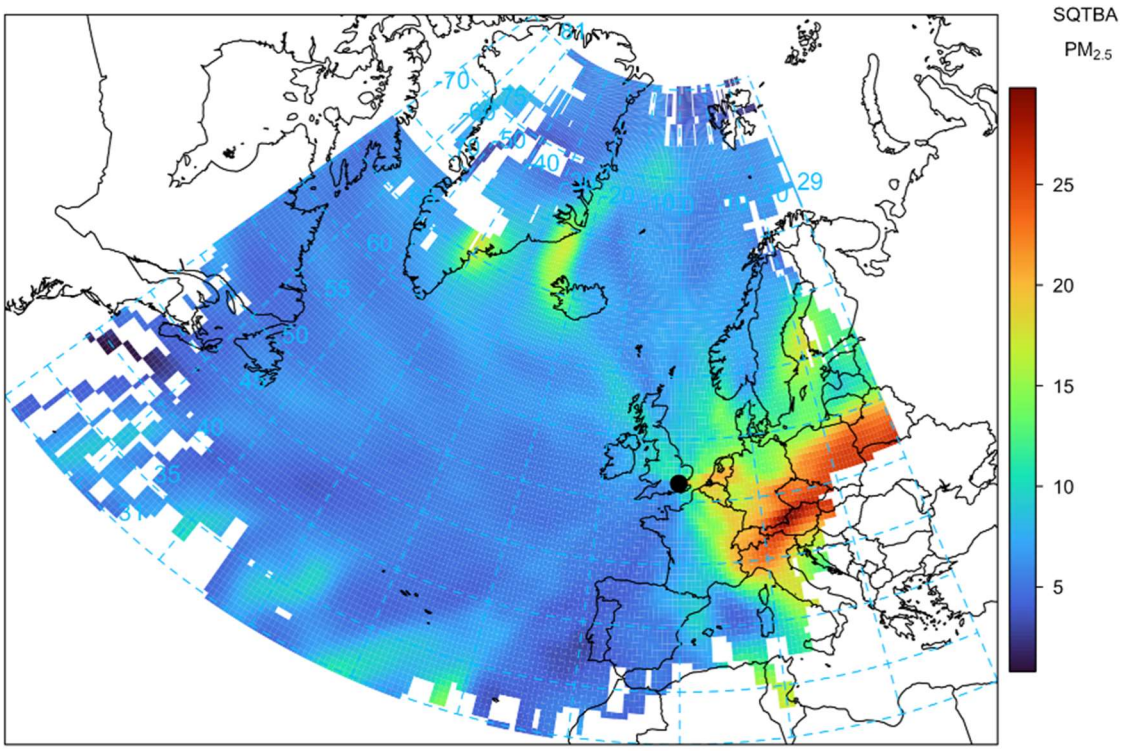


Figure 5.1. An example of source contribution analysis of $PM_{2.5}$ concentrations measured at the North Kensington site in London for 2020. 96-hour back trajectories were run at 3-hour intervals using the Hysplit model. The spatial source contributions were made using a technique called Simplified Quantitative Transport Bias Analysis (SQTBA). The calculations were made using Openair (Carslaw and Ropkins, 2012).

These types of technique (of which there are many) illustrate how particle measurements can be used to better understand precursor source origins at a regional scale. The robustness of the results shown in Figure 5.1 can be improved by pooling the back trajectories across many measurement sites that have different concentrations and are associated with different back trajectory paths, thus improving the 'triangulation' of source origins. Furthermore, these types of analysis can track source changes over time. As developments are made in PM measurement technology (for example sensor measurements, increased number of sites and particle compositional data), they offer ways in which more information on source contributions can be established.

5.1.6 Relative isotopic abundance methods

Isotopic abundances may vary between sources, and this can be exploited for source apportionment. One of the oldest applications is the use of carbon-14 to differentiate between contemporary carbon from sources such as biomass burning, and fossil carbon from coal and oil (Heal, 2014). Isotopes of lead and zinc have been used to differentiate sources such as petrol additives and industry in the former case and brake wear and metal corrosion in the latter. Isotopes of nitrogen and sulphur can also be used to differentiate sources such as agricultural and road traffic sources of ammonia, and combustion and marine sources of sulphate, but the small number of isotopes can require the use of statistical models which give results with large uncertainty ranges (Fan et al., 2019).

5.2 Machine learning

There has been an enormous growth in the use of Machine Learning (ML) and it has been used in a range of problems related to the measurement of PM and related data analysis. ML has been used extensively for low-cost sensor calibration and there is a growing use for the analysis of both compliance-type measurements and PM composition. It is clear that ML capabilities and applications will increase in the future as the techniques continue to develop and data availability grows.

Machine learning (ML) is considered to be a subset of the field of artificial intelligence and can broadly be considered as a field of study that gives computers the ability to learn without explicitly being programmed. Interest in ML has increased enormously in recent years across a wide range of disciplines including air quality. This has been driven in part by the development of numerous open-source packages provided in R, Python and now Julia. Machine learning is often associated, and sometimes confused, with ‘big data’ because for some areas of ML to be effective, in particular deep neural networks, it is generally the case that large amounts of data are required as input to help reveal underlying patterns and relationships. Fundamentally, ML is a form of statistical modelling, which can involve many potentially flexible and powerful methods that use the input data directly to reveal patterns and relationships in the data. Many of these methods, which are mentioned below, reflect recent discoveries and developments in computer and statistical science. There are broadly two principal uses of ML approaches of relevance to air quality: discovering patterns and relationships in data and for prediction.

Before considering the specific relevance to PM measurement, it is worth highlighting why ML is of interest to air pollution in general. First, the measurement of air pollutants generates a lot of data. For example, in the AURN alone there are billions of measurements, and that is only a fraction of the total data available in the UK. In that sense, the air quality field has ‘big data’ that lends itself to sophisticated ML approaches. Furthermore, even data from a single instrument can be considered suitable for ML to be applied, such as multi-species and / or high time frequency data. Second, the behaviour of pollutants in the atmosphere is inherently complex owing to the underlying physical and chemical processes involved. Concentrations of pollutants tend not to vary in simple ways with atmospheric properties such as wind speed. As such, the use of ‘conventional’ statistical models such as linear or multiple regression is limiting. ML does not seek, for example, to pre-define the relationship

between wind speed and pollutant concentration but establishes these relationships directly from the data itself. Furthermore, there are many complex interactions between variables that can be difficult to describe – for example, pollutant concentrations tend not to vary the same way with wind speed for all wind directions. ML is well-suited to model such complexities without knowing about the fundamental physics and chemistry involved.

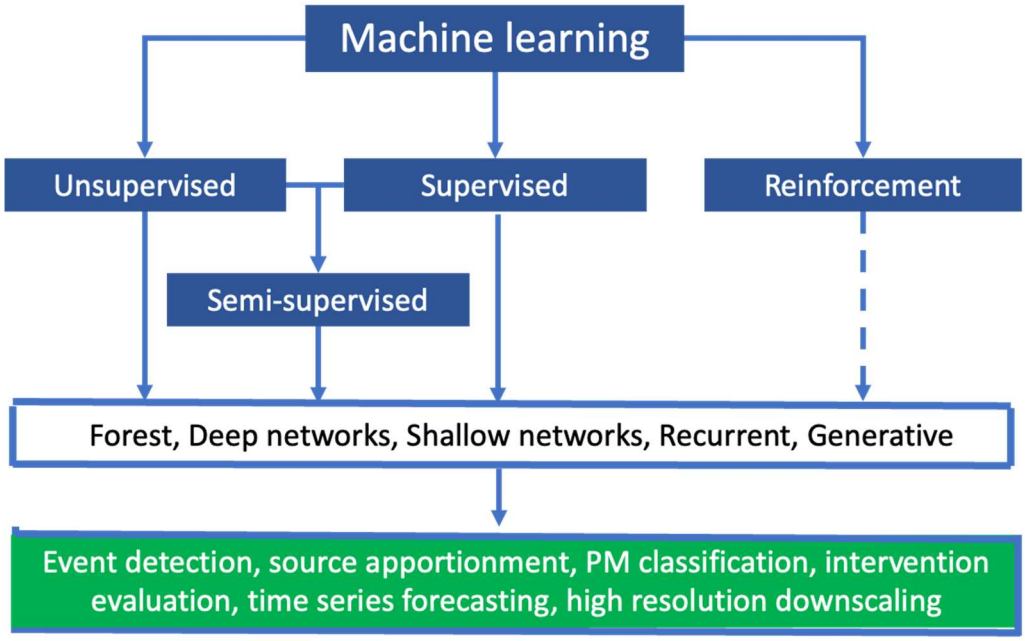


Figure 5.2 – broad classifications of machine learning approaches, their underlying numerical and computational methods and associated insight generation

ML algorithms can be broadly split into 4 distinct groups, as displayed in Figure 5.2 and briefly discussed below: unsupervised, supervised, semi-supervised and reinforcement learning.

Unsupervised learning algorithms attempt to identify distinct groupings within a dataset without any a-priori 'labels' used as part of the training process. For example, an unsupervised algorithm could take a mass spectra from an ambient measurement and extract distinct contributions to that signal for the end user to interpret. Common approaches include the traditional K-means or Hierarchical Agglomerative Clustering (HCA) to group individual observations (e.g. scattering images or mass spectra) into clusters, whilst Positive Matrix Factorisation (PMF) is used to identify common structures within collected mass spectra (e.g. wood burning signatures to the total mass spectra from an AMS. Whilst an algorithm might not be used to detect *a priori* known contributions, results can be sensitive to choice of data pre-processing and model hyperparameters.

Supervised learning algorithms can be trained to identify known signatures from labelled datasets that may have been created through controlled laboratory studies. This includes converting scattering images and/or holographic images into distinct PM types. In these examples supervised methods can be considered to provide a classification function which could be applied online or offline. Supervised methods can also be used to predict continual variables such as $PM_{2.5}$ mass as a function of time and meteorological conditions. Indeed, this includes methods to de-weather time series as discussed below. It seems likely that

deep learning time series methods may be used to predict $PM_{2.5}$ ahead of time from a network of sensors or time varying geospatial maps from distributed networks of sensors .

In some cases, it may be difficult or infeasible to collect enough data or manually construct a labelled dataset to build an accurate classifier of PM type, for example, using supervised methods. This is particularly true if relying on deep neural networks that often require large volumes of data for model parameter optimisation. **Semi-supervised learning** algorithms provide a way to mitigate some of these challenges by using unsupervised methods to learn underlying relationships between features in the data before a supervised learning approach is then used to predict known signatures/labels.

Reinforcement learning algorithms, made popular through algorithms learning how to play games to reach an optimal state, provide a potential route to optimising policy interventions through 'gamification' of PM emission through impact modelling though is very much in its infancy as a general tool and seemingly remains unused in this context of atmospheric science.

Underlying these broad categories are specific algorithm architectures including deep neural networks, forest-based methods and generative methods. For example, deep learning methods include as LSTM frameworks (Long Term Short Term memory models) that can learn from information held within a sequence of measurements (thus time-series) whilst convolutional networks, used extensively in computer vision challenges, are able to learn important dependencies between pixel locales and thus offer important abilities when dealing with instrument generated images or remote sensing datasets.

One of the most prominent uses of ML related to PM measurement is associated with small sensors. There are two principal aspects that are of interest: the use of ML to process and calibrate sensors and the potential ubiquity of measurements that provides an opportunity for ML approaches. By far the most common use to date of ML is this area has been as a way to derive reliable concentration measurements from sensors that can be strongly influenced (in complex ways) by exogenous factors such as relative humidity, ambient temperature and interferences with other pollutants. Smith et al. (2019) used a range of ML techniques to improve sensor accuracy by correcting for cross-sensitivities for measurements of NO_2, O_x (= NO_2 + O_3) and CO. The use of these techniques has increased markedly in recent years as a way to improve low-cost sensor agreement with reference measurements. A wide range of approaches is discussed in Liang and Daniels (2022), who find that regression-based algorithms are effective and reliable methods. Currently, there is no agreement on which methods are best and how they should be applied, resulting in a wide range of approaches being adopted.

There has been a growing interest and use of ML methods to conduct 'meteorological normalisation' or 'deweathering'. The basic idea of these approaches is that if a statistical model can be developed to explain concentrations of air pollutants in terms of meteorological (and other) variables, then it is possible to account for meteorology when considering trends. Such approaches help to understand the extent to which trends in air pollutants are affected by meteorology and can also provide a clearer indication of source changes. The inherent benefits of ML are well-suited for this purpose e.g. accounting for non-linearity and interactions between variables. The predictive capability of ML models is especially valuable

in this context, but so too is their ability to reveal the underlying relationships between variables. The latter point is important because it is useful to consider the patterns and relationships between variables in terms of their physical and chemical feasibility i.e. that the model captures variations in concentrations that are physically and chemically meaningful. Grange et al., (2018) used Random Forests models to explore trends in Swiss PM_{10} concentrations and to provide 'meteorologically-adjusted' trend estimates accounting for both local and synoptic scale meteorological variation.

An extension to meteorological normalisation is the application of ML models to reveal the impact of intentional or unintentional interventions that affect air pollutant concentrations. Fundamental to the effectiveness of this approach is the excellent predictive performance of ML models. It is most often the case that detecting the impact an intervention has on pollutant concentrations is frustrated by the influence of meteorology – that meteorological variation exceeds the effect of the intervention. The use of ML methods can remove or account for much of the meteorological variation to reveal underlying changes to source emissions that would not be observed or as clear in the original data. These methods have a wide range of applications, but the recent impacts of actions taken as a result of Covid-19 have provided a focus on the use of such methods for understanding the impacts of reduced activities on air quality.

There is growing concern around the energy footprint of ML as a field, particularly when it comes to training models that require huge volumes of data and the influence this has on the need for expanding data centres. It remains to be seen what the net effect might be for the exploitation of PM measurements data since, as stated above, data-driven forecasting tools that utilise single points and network data could mitigate the need for running expensive regional models. There is increasing research in the field of energy efficient ML, which includes growing use of lower energy computing hardware platform such as tensor processing units, or TPUs. These are accessible to software products developed using common environments in Python. It is also not yet clear how accessible quantum ML methods will become across applied domains in the coming years.

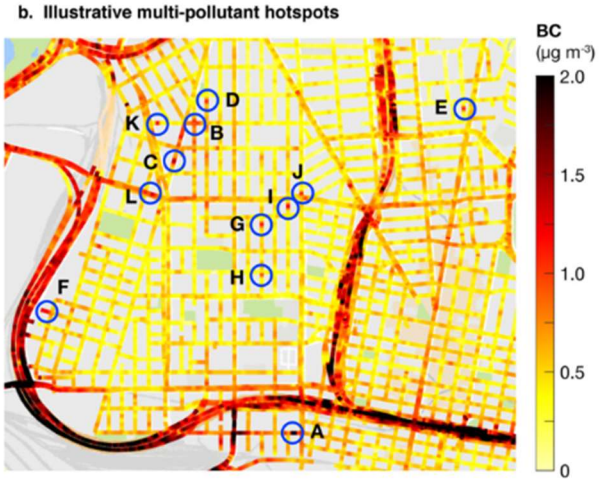
Alongside this is the ongoing debate around provenance and explainability, particularly surrounding neural network-based approaches. More traditional ML approaches, including forest-based methods, allow the scientist to interpret learned relationships and align this with current scientific understanding of relationships between features that should drive model performance. Increased adoption of e.g. Bayesian approaches across multiple disciplines is starting to address the gap in explainable deep neural nets; these approaches require a steep learning curve and software engineering for both development and model training. In exploitation of PM measurement networks for driving insights and forecasting capability, this is going to be particularly important as the emergence of digital twinning gains momentum. Whilst there may be restriction of pure ML approaches in medical domains, favouring physics informed approaches, it remains to be seen how appropriate standards are developed along with benchmarking facilities for new software products. This should include accessible, curated datasets of expert derived labelled products relevant to PM measurement technologies.

5.3 Mobile measurements, including UAVs

Mobile measurements can provide information on PM concentrations using fast response instruments (1 Hz or greater) and reveal spatial patterns in concentration, including finding hot-spots. Mobile measurements tend to be short-term in nature and represent a snapshot rather than long-term averages. The use of unmanned aerial vehicles (UAV) is less well-developed but is increasingly considered as PM sensor and drone technology develops. There are significant data and interpretation challenges with measuring PM from mobile platforms because they express simultaneous variations in concentration in space and time.

Mobile monitoring has increasingly been used to measure PM and other pollutants. The most commonly used approach is instrumented road vehicles to record concentrations of PM components along road networks. These approaches can be used to gain insights into concentration distributions beyond that possible with a static measurement network. By their nature, mobile measurements tend to be associated with short campaigns rather than providing a long-term continuous measurement. A basic requirement for mobile measurement is the use of fast response instruments (typically 1 Hz) to enable spatial variations in concentrations to be revealed. There have been several applications of mobile measurements of PM that include mapping concentrations, identifying sources and comparison with fixed site measurements.

Apte et al. (2017) instrumented a Google Street View car for a period of 1 year in San Francisco and measured BC, NO and NO_2. (See example data in Figure 5.3) A notable feature of the Apte et al. (2017) work was the comprehensiveness of the measurements, covering 750 km of road network.



c. Description of identified hotspots

ID	BC	NO	NO_2	N_{days}	Plausible sources / hypotheses
A	+	~	+	38	Truck traffic and intersection
B	+	+	+	29	Metals recycling business
C	+	+	+	41	Cement plant and automotive shop
D	+	×	~	28	Warehouses with forklifts
E	+	~	+	23	Car dealer, vehicle "smog check"
F	+	×	~	17	Near recycling business; trucks; near I-880 frontage road
G	+	+	+	26	Towing lot, residential "hangout" area
H	+	+	+	24	Residential, no clear hypothesis
I	+	+	+	23	Warehouse and parking lot
J	+	~	+	40	Major intersection, parking lots, drive-through restaurant
K	+	+	+	24	Forklift dealership
L	+	+	+	52	Trucking, tire shops

Figure 5.3 Spatial variation of BC concentrations from Apte et al. (2017) using mobile measurements together with identification of potential hotspot sources.

Such comprehensive sampling could reveal persistent concentration features spatially at a resolution of 30 m. As well as finding sharp small-scale variability in BC, NO and NO_2, areas of elevated concentrations (hot spots) could be associated with known sources such as high truck traffic and various industrial sources. In further developments to the Apte et al. (2017) study, Chambliss et al. (2020) compared the spatial distribution of a network of BC sensors

with mobile measurements using a photoacoustic method. Such studies help reveal the comprehensiveness of static networks in spatially representing concentrations of pollutants such as BC.

Padilla et al. (2021) also used an instrumented Google car to make mobile measurements in London. An interesting aspect of the London measurements was the measurement of ratios of pollutants to CO_2 (a measurement of 'emissions intensity'), which in principle allows fuel-based emission factors to be derived and quantified spatially. These types of measurement can be analysed in such a way to reveal local enhancements in concentration and emissions intensity – such as at street scales, thus providing detailed spatial information across an urban area. Such sampling has the potential to prioritise locations where further mitigation in emissions is necessary and also has a role in helping to identify locations where longer-term static measurements would be beneficial. Emission ratio / intensity measurements also provide a direct way to compare fleet average values with those derived from commonly used emission factor models such as COPERT. The Padilla et al. (2021) measurements were made before and after the implementation of the London ULEZ (Ultra Low Emission Zone) and thus provided a way of quantifying changes in emission ratios in addition to concentrations before and after the introduction of the ULEZ at a street-level scale.

An example of mobile PM measurements is shown in Figure 5.4 from London made in September 2022. The measurements were made using the University of York mobile laboratory using a PALAS AQ Guard instrument set to measure PM1 at 1 Hz. The Figure represents the mean of 48 repeats of a fixed route and highlights persistent areas where PM_1 is elevated e.g. along the Aldwych / Strand. It would be expected that PM_1 would be dominated by exhaust particles rather than non-exhaust. While further analysis of the data is ongoing, these experiments do highlight the potential of mobile measurements to identify areas of significant source strength and potentially elevated personal exposure.

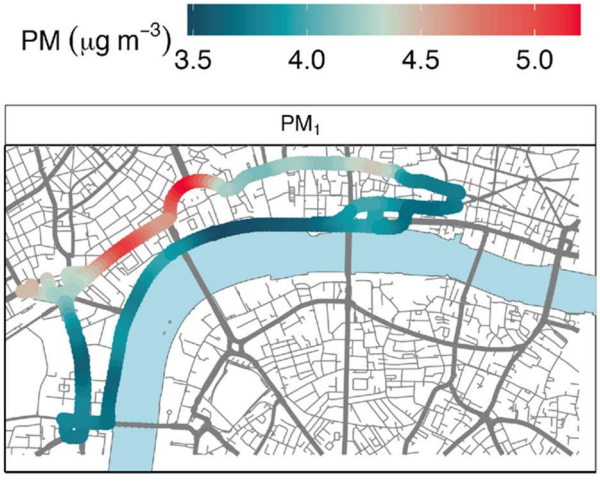


Figure 5.4 Spatial distribution of PM_1 concentrations for a small area of central London. These measurements represent the mean of 48 repeat circuits carried out in September 2022 by the university of York mobile laboratory using a AQ Guard instrument. Acknowledgement: Dr Shona Wilde, University of York.

Mobile measurements have also been used to characterise the emissions from specific source types. Mobile measurements have been used to quantify impacts of PN emissions from airport / aircraft sources at Los Angeles Airport (Hudda et al., 2014). This study is a

good example of a targeted approach to better understand a specific source and its associated spatial impact. Elevated PN concentrations extended to 16 km downwind, with a 4- to 5-fold increase to 8–10 km downwind. These types of mobile measurements have the advantage of making it possible to transect a dispersing plume (at ground level), which helps to strengthen the linkage with specific sources. Indeed, it is possible to couple plume transect measurements with a dispersion model to back-calculate emission rates, as has been carried out for pollutants such as CO_2 and CH_4 (Kumar et al. (2021).

The ongoing development of low-cost sensors and miniaturisation of PM instruments, together with that of drones (unmanned aerial vehicles, UAVs), has provided the opportunity of making airborne PM measurements. Similar to surface measurements of PM, these technologies enable spatio-temporal measurements to be made, with the potential to reveal a more detailed understanding of the spatial variation in PM concentrations (Hedworth et al., 2021). Compared with surface measurements, typically made on roads, UAVs in principle have more potential to make measurements spatially with fewer constraints e.g. measuring over a pre-defined grid. In this respect, there is more opportunity to measure in difficult to reach or dangerous locations (Cashikar et al., 2019). UAVs can provide concentration information with height, which would otherwise be challenging or limited e.g. compared with tower-based measurements at a fixed location. Given the inherent three-dimensional nature of air pollution, concentration measurements in the vertical could in principle offer valuable insights into sources, the nature of dispersion and atmospheric chemistry. For example, the ability to measure concentrations through an elevated plume from a discrete source (e.g. a large ship) to understand both the variation in concentration through a plume and the nature of the dispersion itself.

Despite the potential of UAV measurements, there are significant practical and sampling challenges associated with the measurement of PM (and other pollutants). Not least is the effect of the drone itself perturbing the air in close proximity to the sampling apparatus (Hedworth et al., 2021). If low-cost sensors are used, where measurements can be affected by atmospheric temperature, humidity and composition, these influences are likely to be greater when making measurements.

An important issue with all mobile measurements is the effect of sample size e.g. number of repeat measurements of a road or transect downwind of a source. Because mobile measurements tend to be short-term in nature, there is a question over their representativeness of longer-term averages and behaviours. In this respect, careful experimental design is required to maximise the benefit of such measurements. Mobile measurements do have the potential to provide a stronger linkage between ambient concentration measurements and personal exposure measurements through to personal exposure modelling. However, mobile measurements tend to provide information related to on-road concentrations rather than directly in environments where people are exposed, but could nevertheless provide valuable information spatially.

5.4 Personal exposure estimation

Small and portable PM sensors offer new opportunities to study human exposure to air pollution. This can be through static measurement in micro-environments where there is little data or portable instruments that can be carried by individuals to measure their exposure over a period of time. This has potential to inform human health studies (including the construction of exposure models) and identify and quantify specific environments or activities of concern.

Here we consider the term 'exposure' as the time integral of concentrations experienced by an individual. Whilst there are continued efforts to understand personal exposure in a range of indoor, outdoor and in transit microenvironments, the lack of high quality measurements limit our understanding, with AQEG's recent report (AQEG, Indoor Air Quality, 2022), spelling this out clearly for indoor environments. Relatively recently the provision of micro environmental measurements for use with models such as the London Hybrid model (Smith et al., 2016) were scarce, although Moore et al. (2016) and more recently Evangelopoulos et al. (2021) studied COPD patients who carried personal air monitors for up to 6 months, measuring exposure in a range of microenvironments. The benefits of using exposure data indoors and outdoors continues to benefit indoor models (Vu et al., 2022). There are ongoing projects such as Wellhome which aim to measure in a further 100 west London homes (part of the Clean Air Programme Strategic Priority Fund). Other transport microenvironmental exposures that have been studied include Black Carbon exposure for professional drivers (Lim et al. 2021).

A potentially important route to exposure of PM is from underground urban metro systems including the London Underground. Concentrations of PM inside these systems are many times higher than ambient concentrations and so relatively short periods of exposure can play a significant part in a person's average daily exposure. The most straightforward way to characterise exposure to $PM_{2.5}$ on the London Underground is through high quality portable measurements. Smith et al. (2020) and colleagues undertook a campaign of measurements on the London Underground to map the $PM_{2.5}$ concentrations on different lines, compare with $PM_{2.5}$ above ground, assess the $PM_{2.5}$ composition and better calibrate optical measurements.

Smith et al. found the $PM_{2.5}$ mass in the underground (mean 88 $\mu g\ m^{-3}$, median 28 $\mu g\ m^{-3}$) was 2-4 times greater than ambient urban background (mean 19 $\mu g\ m^{-3}$, median 14 $\mu g\ m^{-3}$) and road side locations (mean 22 $\mu g\ m^{-3}$, median 14 $\mu g\ m^{-3}$) and that deep lines such as Victoria had much higher median concentrations of 361 $\mu g\ m^{-3}$. The degree to which exposure to $PM_{2.5}$ concentrations of this magnitude lead to poor health is still the subject of ongoing research with PM composition being important in this regard. In the London Underground $PM_{2.5}$ composition is different from ambient PM, being comprised of 47% iron oxide, 7% elemental carbon, 11% organic carbon and 14% metallic and mineral oxides.

5.4.1 Exposure during commuting and driving

Portable PM sensors have been used to characterise air pollution exposure while travelling. These measurements have been used to help people to nudge people to different travel

choices and also to understand personal exposure for health studies. Portable black carbon instruments were used as part of as part of a public engagement exercise with the environmental campaign group Hubbub. They worked with King's College London (McAll et al. 2019) to determine air pollution exposure profiles of a diverse range of Londoners, revealing when and where individuals were most exposed, then used their personal data and stories as part of a wider communications campaign. Stories from the measurement campaign were published in The Times. Ten Londoners, each with different occupations, backgrounds and travel routines, were given micro-aethalometers (AE51) to carry with them for one week. The instruments have the advantage of robustness and ease of operation.

Black carbon (BC) was used as a surrogate for exposure to air pollution from road transport sources. Participants included a school pupil, a college student, a construction worker, an inner-city HGV driver, a Gas Safe engineer, a city cyclist, a doctor, an office worker, a runner and a retired librarian.

The results from McAll et al. (2019) were presented as a cumulative exposure expressed as concentration x time in μg m^{-3} minutes, rather than a simply mean concentrations per mode. Substantial differences were found in exposures. The participants with greatest exposures, 13,000 μg m^{-3} minutes for the long-distance HGV diver and 8,450 μg m^{-3} minutes for a construction worker, had occupational exposures as part of their job. The lowest exposures were 725 μg m^{-3} minutes for the school child. Modern mechanically ventilated buildings had lower concentrations than naturally ventilated buildings. In each case exposure was dominated by the time spent in transport modes (in some cases as part of their work) however this reflects vehicle exhaust as the dominant source of BC in London.

Oxford Street 1 and 2

The Oxford Street 1 and 2 studies set out to investigate how short-term exposure to air pollution from diesel traffic affected health. The first study (McCreanor et al. 2007) investigated the impacts on mild asthmatics and the second (Sinharay et al. 2017) investigated impacts on older people with heart disease and chronic obstructive pulmonary disease. The highly dieselised environment of Oxford Street was used as an exposure source for urban air traffic pollution. Participants were randomised and walked either along the shopping street or circuits of the Round Pond in Regent's Park as a low pollution control.

Figure 5.4 Study participants in the Oxford Street 2 study were accompanied by a researcher wearing a backpack containing portable measurement equipment. Photo - Benjamin Barratt, Imperial College London.

Both studies were enabled by the advent of portable PM measurement devices to assess real world exposures for the participants in both the high and low pollution environments. These were essential to the experiment design and enabled the measurement of contrasting real-world pollution mixtures and exercise rather than single source exposures and artificial exercise that might be simulated in a chamber study. The first study found changes in lung function following the Oxford Street walks and the second study found both respiratory and blood circulatory changes.

5.4.2 Case study on use of measurements in exposure models.

There is increasing interest in use of total personal exposure to air pollution in health research and policy development, rather than just exposure to outdoor air. In a recent call for evidence from DEFRA on the impacts of COVID the London Hybrid Exposure Model (Smith et al. 2016) was used to estimate the exposure to $PM_{2.5}$ of several population subgroups in London, indoors and outdoors, whilst travelling to work and working. The subgroups included children, professional drivers, hospital workers and tube users. It was assumed that hospital workers and professional drivers continued to work as normal, while tube users and children stayed at home. See Fig 5.5 for the estimates before the COVID lockdown, during lockdown and then during lockdown if including an extra 1 hour of cooking activity.

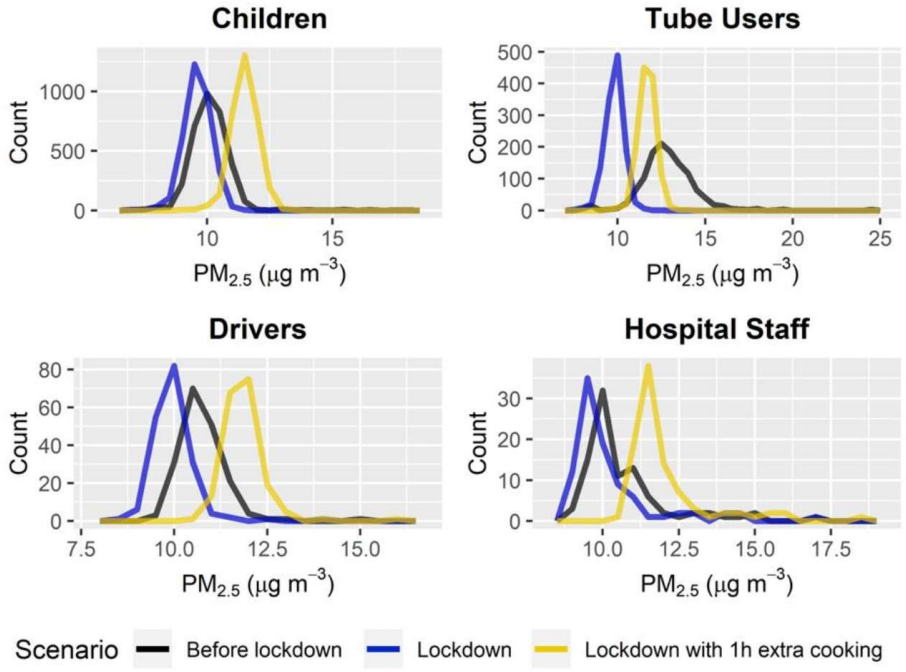


Figure 5.5 Histograms comparing modelled human exposure to $PM_{2.5}$ before and after the lockdown in different subgroups, with the "1h extra cooking" scenario also included.

Before lockdown the differences in exposure to $PM_{2.5}$ reflected the importance of the time spent indoors and the impact of transport-related exposure, with tube users and professional drivers having the highest exposure. After lockdown, and due to changes in traffic sources of air pollution and people's work activity, average subgroup exposures reduced by 5-24%. Despite these benefits the driver remained the most exposed. The sensitivity of changes to indoor activity was tested by increasing time spent at home and, importantly, additional exposure in a kitchen environment where cooking is taking place. The addition of an hour of extra cooking time demonstrates this to be an important source, increasing $PM_{2.5}$ exposure to above pre lockdown levels except in the case of the tube user.

This analysis identified several applications for small PM sensor measurements, without which personal exposure analysis would be extremely difficult. The highest, transport related environments are difficult to predict with certainty, and whilst simple mass balance models are used for this purpose the scarcity of in-vehicle exposure measurements needs to be addressed. In response, there needs to be an expansion of routine $PM_{2.5}$ exposure measurements that reflect the population's exposure within transport microenvironments, for both vulnerable groups and also professional drivers.

In the specific case of the London Underground the exposure to PM is extremely high but varies significantly between lines. The exposure contrast was made possible only through the use of a fixed site at a tube station, supporting small and mobile PM sensors on the trains themselves (Smith et al. 2019). PM measurements continue to be taken, with the aim of understanding the composition, source and health impact of this important PM exposure pathway, however, this high exposure environment would also benefit from more routine PM measurements.

Finally, the recent AQEG Indoor Air Quality report comprehensively covered the need for systematic and high quality indoor measurements to be taken in the UK. One of the most important and understudied of these is exposure to cooking sources of PM, as indicated by the LHEM model results.

5.5 Community/Citizen science approaches

Lower cost, small and portable PM sensors have opened the opportunity for citizens to be involved in air pollution measurement. Whilst many natural science disciplines have long relied on citizen observations, air pollution measurements have largely been undertaken by research professionals and public bodies. Citizen, or more inclusively community, science (Cooper et al., 2021; Ellwood et al., 2023) presents a new evidence opportunity for Defra but one that is difficult to include directly within existing regulatory frameworks.

Citizen science projects can take place across the public engagement spectrum from citizen led to researcher led. Examples are given. Common themes include data to support advocacy for solutions from local government and other agencies, often from communities that feel that they have been overlooked. Other aims include gathering information to avoid exposure and raise awareness in the local community as well as data to track changes.

5.5.1 The growth of citizen measurements of air pollution

Citizen science has a long history in observations of the natural world. For instance, ornithological research relies heavily on volunteer observers who share their data (Hakley et al. 2021). Within atmospheric science, volunteer weather observations spanning several centuries are being "rescued" from paper reports and used in climate research (Veale et al 2017).

The UK was the first country to create a national air pollution measurement network, with measurements dating back to the early 1920s. Throughout the century, the vast majority of measurements were undertaken by professional bodies such as government agencies, universities, local authorities, and industry, rather than by individuals (Fuller 2018, Mosley 2009).

The advent of low-cost and accessible sensors has opened up new possibilities for citizens to collect environmental data, to track changes, and understand their local environment and for participatory research. Critics of citizen science approaches have pointed to potential bias from the motivation of people that participate. For instance, opponents of a local industry or development may track possible pollution because they want to gather evidence of harmful effects. One mitigation is transparency around those that collect data and measurement locations. Others have pointed to poor data quality from the instruments typically used for citizen science. However, Gabeys et al.(2016) proposed the concept of "just good enough data" that can be generated to evidence and inform environmental problems (see, also Nature (2015); Hamm et al. (2020)). Other issues can relate to the *ad hoc* nature of citizen science projects and the challenges in scalability of projects (Maccani et al., 2020).

Citizen science can be led by citizens themselves but the most formal projects are designed by professional scientists with citizens as contributors. However, citizen science does not just provide benefits to scientists, it provides an opportunity for citizens to become engaged in scientific investigations and, relevant to this report, to become engaged in air pollution issues (Gabrys et al. 2016, Varaden et al. 2021). Policy-led citizen science projects can be empowering and enabling for citizens and can ensure greater connection between citizens and policy development, implementation and evaluation (Schade et al, 2021).

5.5.2 Community/Citizen led projects

Luftdaten.info / Sensor.community

Luftdaten.info started in 2016 in Stuttgart, Germany as a bottom-up project within a social internet of things (SIoT) framework. Translated as "air data" Luftdaten collects citizen sourced sensor measurements of PM using a low-tech sensor kit. A core team is financed by donations and partnerships. Individuals may buy their own build-it-yourself sensor kit for 30-40 Euro (2020 costs), though many are funded by local initiatives.

The network grew in around three years to over 10,000 sensors in 60 countries in 2020 and by late 2022 it comprised over 13,500 sensors in 77 countries. Now rebranded as

Sensor.Community, all measurements are publicly available on the website: https://sensor.community/en/.

The Luftdaten sensor measures PM using a SDS011 (Nova Fitness Ltd. Co., China), an optical sensor with laser and photodiode. This is coupled with a relative humidity sensor. An evaluation in Santiago, Chile compared the prototype Luftdaten sensor to a BAM. The small sensor showed better performance for $PM_{2.5}$ than PM_{10}, however relative humidity (RH) was key interference with the sensor tending to overestimate PM when RH was greater than 75% and underestimating when it was lower. R^2 values for hourly $PM_{2.5}$ concentration ranged between 0.47 and 0.86 (Tagle et al. 2020).

The public uses of hyperlocal measurements are reported to include community members optimising their household ventilation, deciding when to take exercise or in the case of one elderly Stuttgart resident when to hang out her washing. In Stuttgard, Luftdaten produced automated air pollution information and alerts for the public and media. However, the core network has at times struggled to shake the perception that it is activist led (Hamm, 2020).

Saaf Hava project, Easton, Bristol

The Saaf Hava project in Easton, Bristol was initiated in December 2017 by RADE (Residents Against Dirty Energy), which was itself originally set up as a community campaign against the potential air quality impacts of a proposed diesel-fuelled Short Term Operating Reserve (STOR) plant close to residential and school buildings. Saaf Hava means "Clean Air" in Urdu, to reflect the fact that this is a community-owned project in collaboration with the Council of Bristol Mosques. Afrif Khan, the Chair of Bristol Council of Mosques, has stated: "*Saaf Hava will give the community invaluable information, and bring many more voices into this debate about all our futures. We all breathe the same air, and pollution knows no boundaries. Too often, the BAME communities are not involved. Now we are showing that we both care, and can contribute positively. I am talking to all parts of the Faith Community, asking them to join us, and add sensors to their buildings so that we can create a more and more detailed map of the very air we all breathe*."

The project was born out of an Interest in low-cost sensors by RADE which led to a recognition of the local air quality impacts of wood burning stoves, the use of which in some areas of Bristol had been steadily increasing. For the local community the health effects of domestic wood burning were well-known from the experience of families and growing up in rural Pakistan. The use of low-cost Luftdaten sensors were seen as a valuable tool to highlight to policy makers the effect that wood-burners were having on local $PM_{2.5}$ concentrations, and to counter the wider narrative that traffic was the main problem affecting air quality in Bristol. There are approximately 20 monitoring sites linked to the Sensor.Community platform and a local data visualisation website https://sts.bristolairquality.co.uk/#. These $PM_{2.5}$ sensors are in the process of being supplemented with Telraam citizen science traffic counters, in conjunction with UWE Bristol WeCount project, and NO_2 sensors (https://baggatornexus.org/saaf-havas-air-pollution-developing-stembaggator-project/). The intention is for these data to be collectively mapped by UWE students on the Community Action and Knowledge Exchange (CAKE) module together with other data to create a digital twin of Easton.

The Saaf Hava project has positively impacted local awareness and directly reduced availability of firewood in the local store. As Rashid Majothi. owner of the Bristol Sweet Mart in Easton, stated: "*I used to think air pollution was all about traffic. I forgot seeing the clouds of wood smoke from cooking in Uganda when I was a child. RADE showed me the graphs from previous winters and I was shocked. Bristol Sweet Mart has seen a growing demand for the wood we were selling. Having seen the effect of this we've stopped wo*od." Despite its positive impacts, engagement with the project has been reported as being hampered by lack of funding and limited capacity for technical support. Ongoing funding to provide the necessary technical support has been proposed to be raised through commercial and research-funded projects that wish to utilise the data, although for the public data will be open-source.

5.5.3 Sensors set up by individuals in the UK

Many air pollution sensors are not part of local networks but are established by individuals on their own. To illustrate these applications, AQEG sought information from two members of the public who sometimes correspond with an AQEG member (Fuller). They were asked, "What did you want to achieve from your air pollution measurements?" and "How have your measurements been used?". Both were using their measurements to highlight local pollution problems to central and local government.

Respondent A, lives in rural north Wales and installed a Lufdaten sensor in response to their complex health needs and consequent air pollution sensitivity.

> *"I wanted to know if the PM levels match my own (very sensitive) "nose/lungs". I can always tell if there is smoke or road fumes in the air. I wanted a form of verification. The sensor works well and is reassuring.*
>
> *I have also been able to show others, such as local authority, how the PM level goes up and down over the days, months and years – and how it can vary considerably from the very general ambient Air Quality levels as reported/forecast by Defra or AQWales."*

Respondent B lives in northwest England. Their interest is in living a zero-carbon lifestyle and they have installed six Purpleair monitors at locations in the local authority area.

> *"I began making measurements out of concern about wood smoke pollution from the proliferation of domestic solid fuel burning in Chorley.*
>
> *I wanted to prove to [the] council that the area has a $PM_{2.5}$ air pollution problem and the need for the council to start monitoring $PM_{2.5}$.*
>
> *I collated 2-3 years of daily mean data and calculated the annual and monthly means in a spreadsheet. I then crafted an email to all the local borough counsellors and MP explaining the health impacts and my findings with a copy of the spreadsheet. I also use the real-time levels to lobby various people / agencies on social media".*

5.5.4 Researcher led projects

"I am an air pollution scientist"

The advent of small air pollution sensors allowed the creation of air pollution backpacks that could be worn easily by primary school children to measure their exposure at home, during their journey to and from school and within the school itself. The investigations took a participatory research approach, under the umbrella “I am an air pollution scientist”, to investigate the impact of collecting personalized air pollution exposure data on children and parents in terms of their perceptions of and responses to air quality.

In the first study (Varaden et al. 2018), 400 children were taught about air pollution and measurements were made on a sub-set of 10 children and data shared with the whole school. Participants said that access to personalized data that they collected increased their air pollution awareness and their desire to reduce their exposure. The children’s participation in the project inspired them to think about ways in which they could influence other people’s behaviour including their travel to and from school.

A second study (Varaden et al. 2021) made measurements with 258 school children in five London primary schools (See Figure 5.5). The researchers found that the air at school was not the largest source of highest concentrations, but rather the journey to and from home. Walking along main roads led to greatest exposure and the children that travelled by car breathed more air pollution than those that walked along quiet roads. Such data could help with schemes that are aimed at improving air pollution around schools reducing children’s air pollution exposure. The project showed that using a citizen science approach to data collection, where children are actively involved in the research, facilitated the gathering of a large dataset and also increased children’s awareness of air pollution, encouraging them to adopt behaviour changes to reduce their exposure.

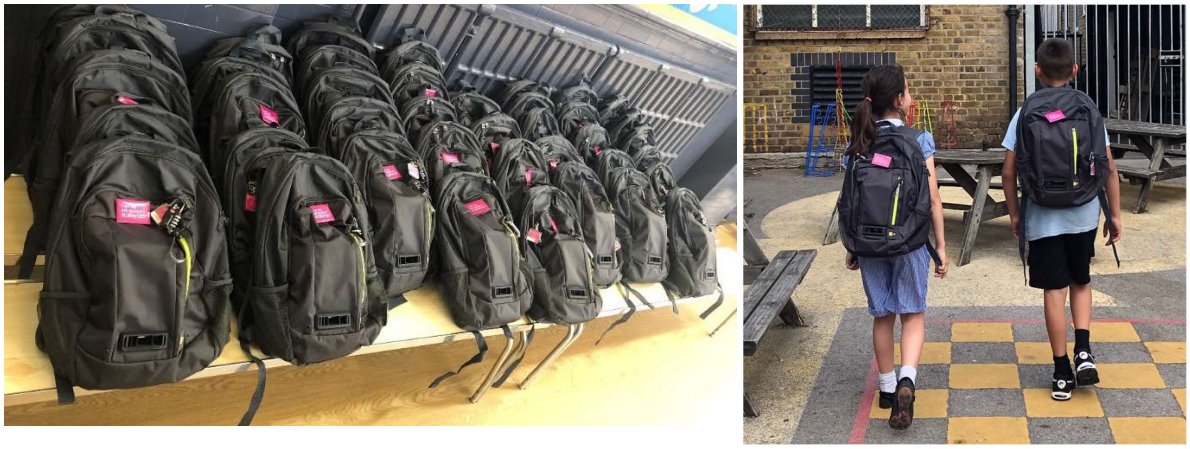


Figure 5.5 Air pollution sensors were incorporated into rucksacks worn by each child. Photo Diana Varaden, Imperial College London.

Breathe London communities programme

The second phase of the Breathe London project began in 2021. Funded by the London Mayor a network of around 150 small PM sensors (Clarity, Node-S, Berkeley, USA) was installed around London, with a focus on schools and areas of local community concern. The Clarity Nodes were selected for their ease of deployment and solar power allowing them to run without mains electricity. The network operated as a hybrid system where Imperial

College London made on-going colocations between nodes and reference instruments which were used to provide correction factors that were responsive to changing air pollution and measurement confounders.

As part of the Breathe London programme community groups could apply for a free Node, funded by Bloomberg Philanthropies. These community nodes also benefitted from quality assurance and the operational support of Imperial College. The community programme aimed to, "*democratise air quality monitoring and create a community of groups who support each other and share their experiences and ideas*."

The community groups in the first tranche of ten provided information about their aims for the measurement project. Common themes include data to support advocacy for solutions and action from local government and other agencies, often from communities that felt that they have been overlooked. Other aims included gathering information to avoid exposure and raising awareness in the local community, as well as data to track changes. Examples of these motivations are captured in the quotes below:

"*As a large BAME community, we feel we have been ignored; our asks haven't been met and we are struggling to have any serious commitment and action to tackle toxic Air Pollution… The Breathe London Programme…will empower our community, by allowing us to monitor the air quality on the road where their families and kids live, play and work. We believe that joining the Breathe London programme can really make a difference and support our campaign for a cleaner, healthier, greener road*." - Rectory Road Residents Association.

"*The data from the Breathe London node will improve our members ability to avoid air pollution and provide them with the data necessary to advocate for measures to improve local air quality*." – Victoria Park Harriers & Tower Hamlets Athletics Club.

"*By harnessing the air quality data captured by this node, we hope to be able to persuade decision-makers at Haringey Council to take action on the ill effects of traffic pollution and implement changes which work for and benefit local residents. It will also help to raise awareness within the community and encourage others to tackle air pollution in North Tottenham*." – Heathly Streets Tottenham.

"*Being involved in this programme to give young people and the wider community a better understanding of the levels of air pollution in their community and more importantly we hope to be able to empower local people to be able to take action to mitigate this*." - Barking Food Forest.

Slow the Smoke

Slow the Smoke is a community participation project led by Bristol City Council and supported by UWE Bristol and Knowle West Media Studio (Figure 5.6) with aims to highlight the impact of domestic solid fuel burning on air pollution (specifically PM). The project engages with a small number of citizen scientists in the Ashley Ward area of Bristol to use Sensor.Community low cost sensors to measure PM concentrations.

The project aim was to achieve air quality benefits in both the short and long term through the planned monitoring, engagement and awareness raising activities in a pilot area of the city. If successful, the project template could be rolled out more widely in Bristol and recommendations provided for other local authorities to learn from the Bristol experience.

Ashley Ward was selected as the pilot due to high rates of solid fuel use. The area is also highly diverse in terms of ethnicity and economically. There is a history of van dwellers in that area. One of the defining characteristics of the area is the community led self-build housing estate called The Yard, which was built with visions of being sustainable: environmentally, socially and economically. Solid fuel use in the area is both a primary heat source for some, but also a secondary "lifestyle" choice for others.

This project, which was reported in the Guardian (Wall, 2022), used an innovative integration of reference monitoring, community science monitoring and several citizen-led engagement activities and community-based workshops, including one creating a virtual Bristol Minecraft world in which to manage air pollution and another locally produced soundtrack of the air quality data[12].

Figure 5.6 Photo in Slow the Smoke–- Google Photos

Citizen Science Microplastics project

HOMEs Under the Microscope (https://homesunderthemicroscope.co.uk/) is a citizen science project to measure airborne microplastics in the home. Rather than using sensors to measure pollutants, citizens are recruited to collect deposited microfibres onto sticky films in a sample dish. Participants use microscopes provided by the project to take images of the samples which are submitted for python-based automated image analysis. Exposed samples

[12] https://www.youtube.com/watch?v=AhWWO0EtM1c&t=8s

are also analysed using Raman spectroscopy to determine microplastic composition. The citizen science participants have two options for engagement: Phase 1 – high engagement citizens undertaking development and stress-testing (Pilot); and Phase 2 – low engagement citizens collecting data using the passive sampling and analysis approaches developed in Phase 1. Citizens will also interact with industry to understand their perceptions of plastic use, clothing treatment and current behaviour patterns.

For both phases, citizens were identified and recruited through local outreach partners with expertise in engaging a broad cross-section of society. Community-based organisations advertised directly to members of underrepresented communities to recruit citizen scientists. Knowledge exchange between citizens and the textile industry is proposed to ensure integration with other microplastic stakeholders. Understanding the distribution of airborne microplastics indoors is difficult to achieve at scale without citizen scientists. The methods developed, applied and analysed in partnership with citizens can potentially inform both academic and industrial outcomes, embedding their contributions in the early stages of a new and societally relevant field of research.

5.6 Using PM measurements indoors

Air pollution measurements are generally focused on outdoors, however increasing emphasis is being placed on air quality in indoor environments. These locations have the potential to dominate exposure as detailed in the AQEG (2022) indoor air quality report. Monitoring in these environments has been transformed by the availability of compact sensors and associated data and communication systems.

5.6.1 Boston public schools (BPS) network

Over 6,000 air pollution sensors have been installed in public schools in in Boston, Massachusetts as part of their Indoor Air Quality Monitoring and Response Action Plan. Each classroom, nurse's room and main office have a sensor along with over 100 on school roofs. The system deployed SGS Smart Sensor measuring CO_2 (Alphasense IR sensor), CO (Alphasense electrochemical sensor), PM (TERA NEXT) along with temperature and relative humidity sensors.

The real-time data from the sensors is publicly available on-line at https://bostonschoolsiaq.terrabase.com/#close . The system is also intended to allow facilities staff to make scientific and data-driven decisions around making improvements buildings, take action on elevated indoor air quality, including identifying and remediating sources of elevated levels, educating the school community about indoor air quality as well gathering evidence to support investments in mechanical ventilation systems.

The sensors are reported to have revealed air pollution problems that have originated from the outside. In one case carbon monoxide was measured in a classroom and traced back to an electricity generator close to the school air intake. When a house caught fire close to another school, the in-class sensors were used to help staff to close windows and keep the children safe from smoke. They have also detected and helped to prevent the use of cleaning products and room air fresheners that can produce irritating chemicals air and trigger asthma symptoms (Boston Public Schools, 2022, Fuller, 2022)

5.6.2 The SAMHE project

Launched in 2022, the SAMHE project is planning to install air quality monitors to measure CO_2, VOC and $PM_{2.5}$ along with temperature and relative humidity in over 1,000 UK schools. Teachers and pupils will be able to access their data through a specially designed interactive Web App and see how air quality changes over the course of hours, days or weeks and months. The project will also include a range of related activities and experiments, creating opportunities for pupils to be scientists and do hands-on experiments with their monitor. Data will also be placed in a national database.

5.6.3 In2Air

Over the coming decade, the UK Government have committed to a £3.8bn Social Housing Decarbonisation Fund to improve the energy performance of social rented homes, on the pathway to Net Zero 2050. A fabric first approach, with interventions like new windows and

doors, external wall, loft and under floor insulation, is a common low-tech intervention method to reduce energy demand. Yet few studies have evaluated the effect of a fabric first approach on indoor air quality in occupied affordable housing.

In2Air is an NIHR funded collaborative project (researchregistry8208) between Northumbria and Newcastle Universities, Newcastle City Council and their housing management organisation, Your Homes Newcastle, targeting this evidence base. The study is a non-randomized natural experiment on 30 single/dual occupancy bungalows occupied by elderly residents in Newcastle-upon-Tyne, NE England. Baseline data collection (i.e., pre retrofit intervention) on paired indoor-outdoor concentrations of particulate matter ($PM_{2.5}$), temperature and humidity, indoor carbon dioxide (CO_2) concentrations, energy audits and self-reported general health and wellbeing are currently in progress. Post intervention monitoring will take place around 12 months after completion of the retrofit works to allow time for the building to stabilize and for the occupants to experience a range of seasons/operating conditions. This will also allow for seasonally matched pre and post intervention monitoring periods.

5.7 Model development and evaluation using PM measurements

To fully realise the benefit of new measurement technologies, the data must also be used in conjunction with relevant models. This can lead to enhanced scientific understanding of sources and processes, improved capability of models, better designed studies and intervention evaluations, all of which ultimately aid quantitative policymaking.

The opportunities afforded by the combination of new and existing UK PM measurement data for air pollution modelling can be split into applications that directly use the data within the model formulation, e.g., for satellite based forecasts of $PM_{2.5}$ concentrations, Land Use Regression (LUR) models and Machine Learning (ML) applications, and those that don't, but that could make more use of PM measurements to improve emissions estimates within more traditional dispersion and Chemical Transport Models (CTMs).

Examples of the former include PM derived from satellite-based Aerosol Optical Depth (AOD) (see Chapter 3). AOD is an indirect measure of the extinction of light within the atmospheric column and therefore predictions of surface level $PM_{2.5}$ is reliant upon resolving the AOD measure vertically in the atmosphere, often using global atmospheric models such as GEOS-Chem, which derive PM in multiple layers throughout the atmosphere, in combination with ground-based PM measurements. Using these methods, impressive continental and global datasets of ground-level $PM_{2.5}$ have been created (van Donkelaar et al. 2019 and 2016). LUR models use the relationships between air pollution concentrations at measurement sites and amongst other things, land use characteristics, traffic information, population density and altitude. With these relationships the LUR models predict between site locations. LUR methods on their own have difficulty predicting PM and are often combined with another prediction method such as from satellites, with recent examples in De Hoogh et al. (2018).

However, by using measurements in their formulation, satellite and LUR models are unable to forecast policy impacts or predict the future, and so UK air quality reporting and policy development have focused on the use of emissions dispersion models and more complex CTMs. An important question is, therefore, what opportunities exist from PM measurements to help quantify emissions that are poorly understood or are completely missing from emissions inventories, how can these help improve dispersion and CTM models, and therefore improve EU compliance reporting and policy development.

Developing more capable models using measurements: It is important that there are direct links between research into measurements, emissions inventories and air pollution models and that there is timely feedback between the three areas so that new findings are included efficiently into policy and compliance modelling. The development of PM emissions estimates is especially important since they are some of the most uncertain, with examples including wood burning, indoor VOCs, non-exhaust vehicle emissions and cooking. It is also important to recognise that whilst the National Atmospheric Emissions Inventory (NAEI) is an essential and high-quality UK resource, it may not always provide the best solution for model applications.

Examples of how new PM measurements can help:

- Industrial sources of Nickel (Ni): In Pontardawe, South Wales, hourly concentrations of 23 elements were measured alongside meteorological variables and black carbon during a four-week campaign in November–December 2015 (Font et al. 2022). Positive Matrix Factorization (PMF) was used to identify sources and cluster analysis was used to quantify the industrial processes contributing to ambient PM_{10} concentrations. Whilst this project successfully identified these sources, more could be made of this type of analysis elsewhere to characterise industrial emissions rates and release conditions.

- Diesel IVOC emissions: Another potentially significant addition to the UK and European emissions estimates relates to intermediate volatility organic compounds (IVOC), not currently included in the NAEI. Measurements from the UK Clearflo campaign in London and specifically the findings of Dunmore et al. (2015), who showed that Diesel IVOC were 3.2 times higher than Petrol VOCs. Using a simple scaling factor, described by Ots et al. (2016) an additional UK total emission of 99.1 Gg $annum^{-1}$ was added to on-road vehicle VOCs emissions in the UK and this was incorporated into current and future CTM model runs adding to SOA formation.

- Other vehicle studies include measurements of black carbon emissions from an Aethalometer placed in a tunnel in Portugal (Blanco-Alegre et al. 2020), estimating the secondary organic aerosol (SOA) formation from in-use vehicle emissions using a potential aerosol mass (PAM) flow reactor in a highway tunnel in Pittsburgh (Tkacik et al. 2014), the emissions and concentration gradients of Black Carbon and Particle Number from a freeway in North Carolina.

- Cooking and wood burning emissions in the London Atmospheric Emissions inventory (LAEI 2019). PM woodburning (Young et al. 2015) and cooking organic (Ots et al. 2016) aerosol concentrations were used with an inverse modelling

approach to back calculate likely emissions totals and to distribute the emissions using housing stock (wood burning) and food sector employment data (cooking). This approach limited wood burning and cooking emissions to within realistic ranges, based upon the ambient measurements. It's clear that much more needs to be done but it's encouraging that work to standardise the source apportionment methods for organic aerosols is developing across Europe (Chen et al. 2022) and this will help develop organic emissions from a range of sources.

- Elemental analysis of PM has also been used to better characterise non-exhaust emissions for use in air pollution models, going back to the LAEI 2010. In this example use of roadside measurements by Harrison et al. (2012) resulted in revised tyre wear, brake wear and road wear/resuspension emissions factors and improved PM_{10} air pollution model performance at roadside sites in London. More recently, as part of DEFRA's Model Intercomparison Exercise (MIE 2022), the same non-exhaust PM_{10} EFs resulted in a positive bias against ambient measurements. However, new high time resolution tracer and CO_2 measurements have been taken in London, Birmingham and Manchester and have helped establish new non-exhaust PM emissions factors and replacing the old non-exhaust EF's with the new has reduced the PM_{10} model bias identified in the MIE. Another example of non-exhaust emissions estimates includes at inner roads and the ring road in Paris with the road dust contributing 13% of PM_{10} annually (Amato et al. 2016). Measurement of non-exhaust tracer species for emissions development is a time-consuming task but given their future importance and relevance to electric vehicles more measurements are needed from around the UK and under different road, driving and meteorological conditions.

- In contrast to the examples above a lack of measurement data limits our understanding of PM sources. In the recent report for the Clean Air Fund the importance of industrial wood burning as a source of PM in the NAEI was highlighted with sources spread throughout the UK. This precipitated the need for model sensitivity tests to establish whether air pollution model assumptions could be improved and also led to a better understanding of the National Atmospheric Emissions Inventory (NAEI) methods. Despite this and because there are no measurements at the hundreds of locations predicted with high PM it remains unclear as to whether these sources are a cause for concern or not.

The ability of model users to contribute to the measurement-emissions-model process is made more difficult because of the relatively small numbers of PM mass measurements compared to other pollutants such as NO_2. Examples of the lack of data include, limited long time series of co-located PM_{10} and $PM_{2.5}$ measurements, the Industrial wood burning example given above and more urban sites to better understanding the spatial variability of PM concentrations/emissions. Furthermore, because of the complexity and cost of PM compositional measurements there are relatively few sites making such measurements, and finally, in future, routine analysis will become more difficult because of the need to measure low concentrations. In response, DEFRA has rightly focused its attention on expanding PM measurements across the UK and alongside the expansion of other 'low cost' sensors this represents an opportunity to increase the measurement-emissions-model analysis.

The application of complex statistical methods to CTMs using ground-based PM data.

Complex air pollution models often require large computer resources and are time consuming to run. As a consequence, whilst examples do exist e.g. Moonen et al. (2015), it is rare to see sensitivity and uncertainty analysis undertaken on such models. In the Moonen paper the authors quote 10^6 – 10^8 lower computational costs by using a statistical metamodel as a surrogate for the model in their analysis. Similar computational benefits were realised in a CMAQ emulator described by Beddows et al. (2017) and as a consequence, this allowed an understanding of the influence of a comprehensive range of input and model variables on O_3 predictions, beginning with a list of over 200. Use of the emulator was taken further and using a Monte Carlo analysis of model inputs and assumptions, resulted in improved predictions of peak O_3 concentrations during a 2006 episode, a difficult task for CTMs. This type of analysis would have been impossible using CMAQ alone. The application of emulators to the complex assessment of modelled PM concentrations in the UK would be an attractive option, however it is worth noting that there is a cost in the significant time it takes to set up and test the model emulators in the first place.

Machine learning (ML) approaches (see also section 5.2) are increasingly applied to models of air pollution concentrations, with results often created using 10-fold cross validation and leading to good comparisons against ground-based measurements. An example of applying ML to CMAQ $PM_{2.5}$ predictions in China, is given by Lyu et al. (2019) in which tests were made against held out PM data (data not used in the ML process), a tougher test than using cross validation and showing more modest improvements. Whichever approach is used it is essential to have a full understanding of the ML methods, and a detailed knowledge of how they have been applied to avoid of problems such as model overfitting, a danger of ML approaches. In addition, it is important to understand that R^2 values can overestimate the performance of data-driven models when predictions are made at locations which have physical characteristics not represented in the training data. The application of ML in modelling exercises can also risk a 'black box' understanding of the model results, limiting interpretability and discussion of model weaknesses and causes of bias. Having said that a well-run and clearly described application of ML to a CTM for PM predictions would have potentially significant benefits for the model user and emissions inventory developer, and it would be wise to consider such an approach in future analyses.

Inverse modelling to increase the spatial detail of air pollution predictions

To increase the spatial detail of air pollution predictions, Chemical Transport Models such as CMAQ have been coupled with local scale dispersion models such as ADMS (Beevers et al., 2012) herein after called CMAQ-urban. However, despite this, there remains significant limitations to the spatial detail of within city predictions, due to the lack of road traffic emissions road by road for use with these models. One way to increase modelled detail is to utilise roadside measurements and to 'back calculate' the associated road emissions. The number of measurements required for this task is large but is of potential future importance due to the proliferation of small sensors, including for PM.

The approach entailed mapping all UK roads segments (~5 million) into separate emissions points every 10 m across the network (~60 million). The major roads were characterised by traffic counts and the road emissions calculated using a bottom-up approach, with smaller

roads, albeit those that are often important locally or within cities, back calculated from roadside measurements. An example of where this has been demonstrated at scale in the UK is through the use 16,800 NO_2 diffusion tubes in 2019.

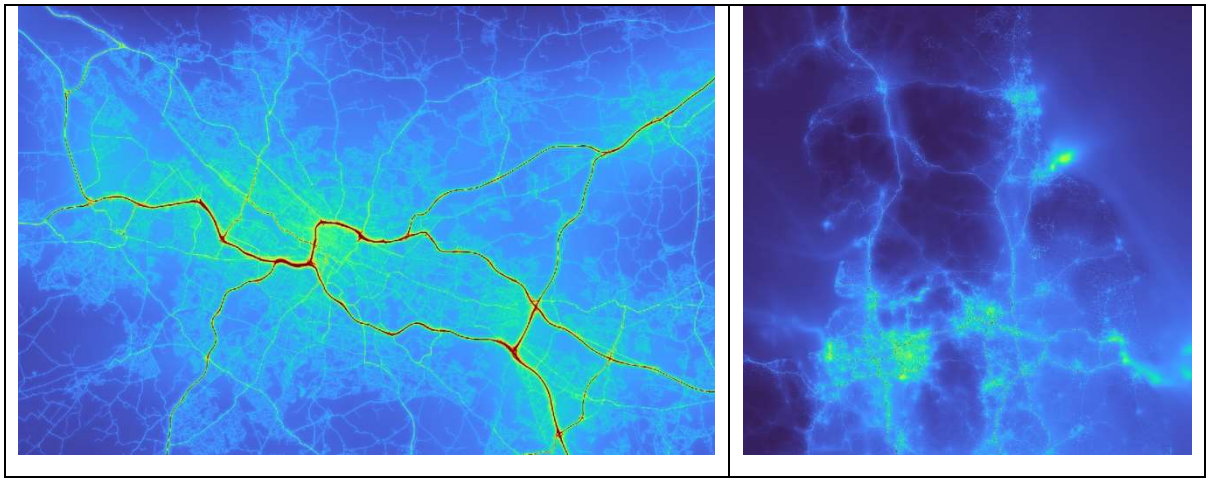


Figure 5.6 Left pane, example NO_2 concentrations in Glasgow, Right pane, results of NO_2 for Northern England.

The method to estimate emissions on these roads used a response curve of NO_2 concentration to emissions changes for each measurement site within a Monte Carlo calibration exercise. This created previously unavailable road emissions estimates, as well as calibrating emissions on those roads that were already in the model and resulted in a spatially detailed model across the UK (see Figure 5.6). However, there were a number of uncertainties including; sites with very low sensitivity to road emissions changes, difficulty in assigning monitors to particular roads at junctions, diffusion tube measurements having relatively high uncertainty and issues of recording location and site type.

Chapter 6 - Emerging PM science

6.1 Microplastics

Microplastics are recognised as an emerging environmental pollutant arising from use of synthetic polymers and leading notably to accumulation in various aquatic environments. Small, airborne microplastics are now known to be a component part of airborne PM. This acts as a route for distribution of microplastics to other environments. Because of their chemical and physical nature, microplastics require a different approach to measurement and quantification compared to other PM types.

6.1.1 Background

'Microplastic' defines particulate plastic debris measuring less than 5 mm in size. These particles and fibres originate from the photochemical and mechanical degradation of a diverse range of plastic materials, such as packaging and clothing. 'Microrubbers', a substantial proportion of which originate from tyre wear, are a closely related material sometimes included in the definition. Several studies have identified microplastic releases from tyre wear in road dust (Kang et al. 2022, Kole 2017), known to be resuspended into air. In addition, microrubbers including styrene butadiene rubber, butadiene rubber and natural rubber, from tyre wear have also been found in the environment (Carrasco-Navarro, et al. 2022) with emerging evidence of their ecotoxicity in the marine environment (Halle et al. 2020), but little evidence for their presence and impacts in air.

With non-exhaust emissions (brake, tyre and road wear) representing 10% of all PM_{10} emissions in 2020 (Defra, 2020), and annual mean modelled lifetime of $PM_{2.5}$ tyre-wear particles estimated to be 28 ± 2.7 days (range 18–37 days), and for PM_{10} 8.3 ± 1.0 days (range 5.5–11 days), a more robust understanding of microplastics and microrubbers within this size fraction is required. An example SEM micrograph of tyre and road wear particles with characteristic morphology of tread rubber and mineral incrustations, is shown in Figure 6.1 (from Panko et al. 2019).

Figure 6.1: From Panko et al., 2019. Scanning electron microscope (SEM) photo of tyre and road wear particles with characteristic morphology of tread rubber and mineral incrustations

Microplastic integrates a variety of plastic polymer types and shapes, spanning several orders of magnitude in size, and of various 'age' and weathering states. Originally discovered as a marine pollution issue, the definition (i.e., < 5 mm) has been fit for purpose for environmental monitoring studies. However growing discoveries of microplastic in a range of human exposure matrices (air, food, water, dust, soil) has emphasised the importance of smaller size distributions with greater relevance to human exposure (Levermore et al. 2020) with hazards, including oxidative stress, inflammation, metabolic disruption.

The recent and growing observations of microplastic in atmospheric deposition and particulate air samples confirm that plastic materials are a source of both ambient and indoor PM. Potential sources of emissions include roads, marine environment, agriculture, and dust emissions generated downwind of population centres (Brahney et al. 2021). However, the field is in its infancy, with technological and analytical challenges, substantial knowledge gaps and hence a wealth of future measurement opportunities to inform the development of, and compliance with, policy needs. Several NERC-supported research projects are exploring airborne microplastic in various contexts in addition to a Horizon 2020-funded consortia under the CUSP Cluster.

The most frequently observed plastic polymers in air are polyethylene and polyethylene terephthalate, with polypropylene, polystyrene, polyamide, and epoxy resin particles also commonly observed. Most studies find that fragments are the predominant shape, followed by fibres; however, these variables depend on the sample type, environment, geographical location, and analytical methodology employed (reviewed in Wright and Borm, 2022), and published microplastic particle number concentrations range from <1 to 1,000 strands m^3. Controlled laboratory wear experiments suggest plastic nanoparticle and fibril release from the wear of synthetic textiles, up to 2.1 x 10^{11} nanoparticles and 5.3 x 10^5 fibrils per gram of synthetic clothing, but these are an analytical challenge for environmental measurement.

There is currently no international or national legislation relating to airborne microplastics; however, intentionally added microplastics are, through amendment to Annex XVII of Regulation (EC) No 1907/2006, expected to be restricted in certain products. Both the WHO and the All Party Parliamentary Group for microplastics, have articulated the need to reduce microplastic releases to the environment, but more data are required to inform and evaluate individual policy interventions. The measurement and characterisation methods set out below may help this.

6.1.2 Methods for sampling

Passive: The earliest studies (since ~2015) first documented microplastic in bulk atmospheric deposition at varying levels spatially, ranging from less than a hundred to several hundred particles m^{-2} d^{-1} (particles deposited per square metre of surface per day). The exact procedures vary by sampling device and duration, ranging from passive sample collection in beakers and bottles, to bowls, purpose-built samplers (Klein and Fischer, 2019) and rain gauges (Wright et al, 2020) (including wet and dry collectors (Brahney et al, 2020)) collecting typically for 1 day (*e.g.,* in indoor studies) to 1 month per sample. Samplers have been deployed outdoors, in a range of environments (pristine, rural, marine, urban), and indoors, with some studies estimating human exposure due to passive ingestion (Fang et al.

2022). Due to the high sample mass and organic content, bulk deposition samples are usually processed to purify and concentrate the target microplastic material ahead of analysis. For historical assessment of microplastic deposition, peat cores (Allen et al. 2021) have been utilised.

Active: Studies have increasingly adopted active sampling methods to quantify the particle number concentration of suspended microplastic. The sampling instruments used in microplastic studies to-date are routine for air quality and include low, mid, and high-volume samplers, with TSP or size selective inlets, cascade impactors (Kernchen et al. 2022), and cyclone samplers (Levermore et al. 2020). There are few publications on the presence of microplastic in PM_{10} and below, due to the analytical challenges this brings; however, these publications observed higher concentrations and a smaller average particle size (reviewed in Wright and Borm 2022). Such datasets will fill a crucial research need.

Whilst most measurements have been undertaken using existing active and passive sampling methods, microplastic-specific measurement collection devices are being developed, including the Norwegian Institute for Air Research (NILU) Atmospheric Microplastics Collector, and a variant of this (Postma, 2022). Both are adaptations of the passive deposited dust measurement Bergerhoff Gauge.

A key consideration in sample collection is the prevention of contamination by background microplastic. Aside from avoiding the use of plastic materials and wearing cotton clothing, field and transport blanks should be collected as is routine for PM field campaigns. Additionally, it is recommended that samplers are trialled to evaluate the potential background contribution from, *e.g.,* plastic components, before sampling commences.

6.1.3 Methods for analysis

Visual assessment using light microscopy may allow for the discrimination between plastic and non-plastic particles of a relatively large size (*e.g.,* >~500 µm), but suffers poor accuracy, with a high possibility of bias, and false negatives and positives. Small microplastic particles, and those particles with an indistinct appearance (transparent, white, yellowish, brown) may be overlooked, whilst some brightly coloured mineral particles may be included. Fluorescence and polarised light modalities have also been employed; however, with visual assessment alone, the type of plastic polymer, and hence source material cannot be determined. This is also the case for Scanning Electron Microscopy-Energy Dispersive X-ray Spectroscopy (SEM-EDS), where a dominance of carbon in a particle is used to discriminate plastic from non-plastic (Tiwari et al. 2019). Thus, standard criteria within the field includes the adoption of an analytical technique capable of fingerprinting chemical composition.

Infrared or near-infrared micro-spectroscopies are analytical methods routinely used to detect and classify the composition of microplastic particles in a sample (reviewed in Mariano et al. 2021). Fourier-transform Infrared (FTIR, absorption or reflectance modes) and Raman micro-spectroscopy are most used, with varying spatial limits of detection depending on the instrument model and configuration. Examples of Raman spectral images are shown in Figure 6.2, below.

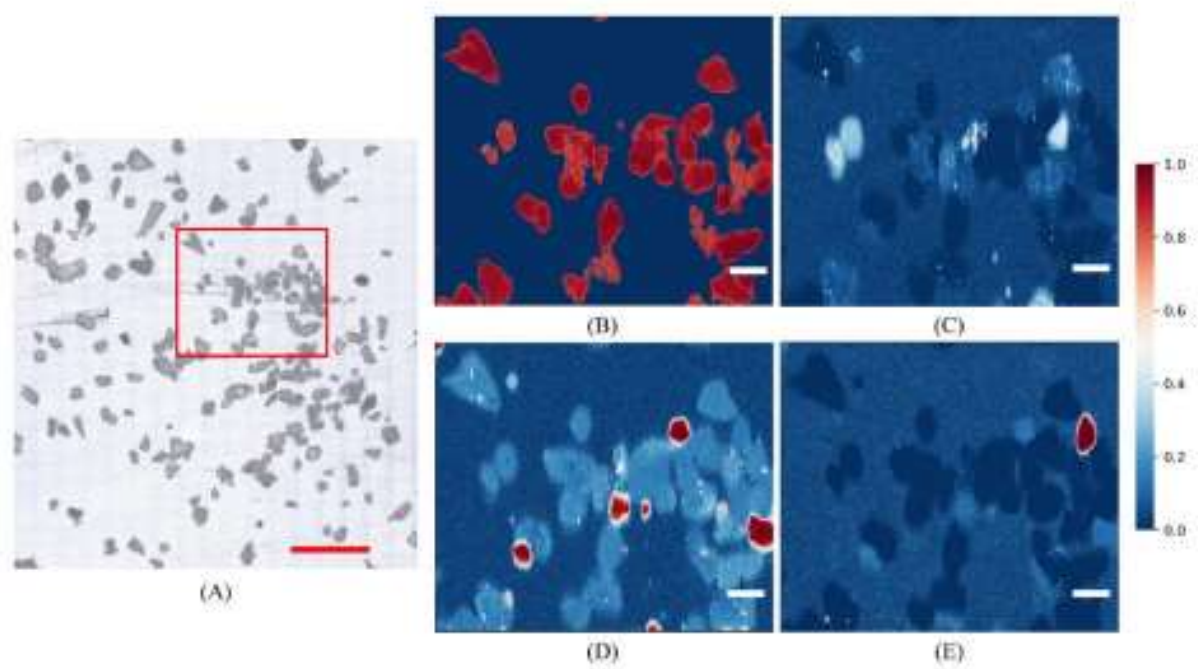


Figure 6.2: Raman spectral images of environmental microplastics dried on to an aluminium slide. (A) A micrograph of environmental microplastics dried on to an aluminium slide (scale bar: 1000 µm). The approximate image area is outlined in red. Spectral analysis identified polyethylene (B), copper phthalocyanine (C), polypropylene (D), and polystyrene (E). The pixels shown in red, refer to positive correlations, while blue denotes the corrected negative or independent correlations (0; Scale bar: 250 µm). Source: Levermore et al., 2020, 10.1021/acs.analchem.9b05445

Considering routine instruments, Raman micro-spectroscopy typically has a higher resolving power, theoretically down to ~300 nm depending on instrument configuration, although there are high specification specialist hybrid instruments capable of nanoscale classification. Depending on the number of particles for analysis these methods can be resource intensive and subject to bias if manually pre-screening and selecting particles for analysis first. Instruments and software which enable either semi-automated particle-finding or automated spectral imaging may remove some of this bias. However, such approaches generate large spectral datasets, governed by the size of the image area, and hence chemometrics are required. These semi- and automated approaches still likely provide an underestimate, since pre-defined instrument and data analysis parameters (e.g., laser power, aperture and magnification, correlation thresholds) will likely result in false negatives for microplastic particles with a poor spectral quality. However, as analytical methods have improved, the evidence to confirm the occurrence of microplastic in smaller size classes relevant to human exposure and health has emerged (Levermore et al. 2020).

To further improve spatial resolution and classify chemical composition at the nanoscale, hybrid techniques can be employed. These include a high-resolution microscopy, such as SEM or Atomic Force Microscopy, coupled to FTIR or Raman spectroscopy (e.g. Schmidt et al. 2021, Brahney et al. 2021). Typically, this correlative approach relies on coordinate matching in a sample, which may not be optimum for microplastic depending on their stability in a sample, although there are some specialist integrated systems available. It is

likely that these techniques will have longer data acquisition times, with fewer particles being analysed. There are no publications on this application and environmental samples.

The above methods generate particle-based data, which can be extrapolated to a particle number concentration. Alternative techniques include thermal methods, such as pyrolysis-gas chromatography mass spectrometry (py-GC MS), whereby plastic polymers are detected and quantified based on their thermal degradation or pyrolysis products. This derives mass metrics, and is thus limited by plastic polymer mass, which is a function of particle size. Figure 6.3 presents a pyrogram of polystyrene (left) and of Nylon-6 (right) with an analytical ion chromatogram (AIC) overlaid with an extracted ion chromatogram (XIC). Depending on the model, instrumental limits of detection can be on the order of picograms to micrograms.

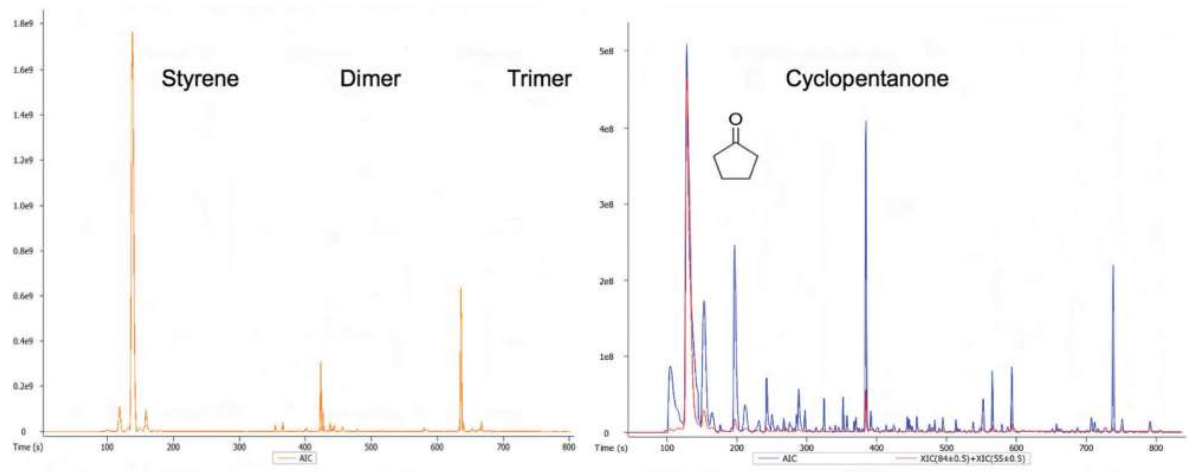


Figure 6.3: Pyrogram of Polystyrene, and of Nylon 6 (AIC and XIC overlaid). Source: Jones et al., Chromatography Today, May/June, 2021.

6.1.4 Future measurement opportunities

Whilst the monitoring and measurement of microplastic spatial distribution in coarser fractions is advancing rapidly, their characterisation in the <10 µm fraction is poorly understood. Key exposure sources and pathways, both indoor and outdoor, are yet to be sufficiently quantified. To improve this understanding and to aid health and environmental policymakers, measurements should be undertaken to:

Identify key sources of airborne microplastics. With just one publication on the modelling of sources of emissions, targeted measurement campaigns of potential sources are needed, enabling the development of emission factors and supporting the development of dispersion modelling.

Refine spatio-temporal variation in microplastics concentrations, including seasonal and meteorological variations

Filter-based methods within the AURN, such as the Beta-Attenuation Monitor, may provide an opportunity to refine the spatio-temporal resolution of airborne microplastics. Similarly,

pre-cleaning filters for other methods, including chemiluminescent analysers, may also support this opportunity. Care is needed with these methods to ensure contamination is kept to a minimum.

Increase measurements in the <PM_{10} size fraction. Measurement and characterisation of microplastic in the <PM_{10} size fraction is required to generate empirical data for use in epidemiological studies. Improving understanding of these parameters may aid in the development of microplastic specific policies.

Inform policy development. To aid the policymaking pathway, the following recommendations are made:

- Define the evidence need for which airborne microplastic measurement is required, to support policy development. The method by which airborne microplastics are collected will likely vary by policy need.

- Once policy is established, appropriate sampling methodologies, verified via field intercomparison exercises, should be undertaken to support comparable and robust measurement.

- Laboratory standardisation and regular contamination reviews, similar to the approach undertaken within the Air NO_2 PT Scheme for assessing the analytical performance of those laboratories, should accompany robust field measurements.

6.2 Biological particles

The highly complex and varied microbiological component of oair is a key determinant of human health yet is poorly understood. A very rapid pace of innovation is occurring in the ability to sample, quantify and characterise BioPM. This includes nucleic acid based methods and real-time discrimination using spectral methods. There is an emerging need for standardisation and unification of methods to harmonise research and to develop opportunities to understand and set exposure limits for BioPM.

6.2.1 Biological particles present in the air - BioPM

Bioaerosols are a complex mixture of airborne particles with a biological origin (BioPM). Particles range from > 10 nm - 100 μm in size with the majority in the range 0.1-10 μM. Contributing to BioPM are airborne microorganisms including eukaryotes (eg. fungi and plant pollens), prokaryotes (bacteria and archaea), viruses and including fragments of cells and aeroallergens released by animals (such as dust-mites and pets). BioPMs also include by-products of microbial metabolism such as microbial volatile organic compounds (MVOCs), endotoxins and mycotoxins. Accordingly, measuring the range and complexity of BioPM composition has necessitated innovation in capture and analytical techniques.

The need to rapidly innovate the detection of a BioPM has been exemplified by COVID-19, where the novel nature of the infection required rapid development of methodologies to quantify SARS-CoV-2 in bioaerosols in order to better understand epidemiologically-relevant variables such as exposure assessments and the success of interventions. However, the wider risk to humans posed by background and expected BioPM requires nuanced and data-rich approaches. This is because, on one hand, all air contains BioPM and long-term exposures of populations to BioPM are normal and healthy. One the other hand, short term exposures to high density bioaerosols - pathogens, allergens or toxins - can lead to poor health outcomes that include death as was recently exemplified by a tragic UK case that was specifically attributed to indoor exposure to moulds (Judiciary 2022). These exposures can occur in the homes, the outdoor environment and in the workplace, and vary widely depending on the global context, the type of built environment, and occupation and the presence of other pollutants.

Defining safe exposure limits for BioPM is not simple owing to their complexity and the lack of simple dose-response relationships. The consequences of exposure to BioPMs vary from person-to-person and include individuals risk-factors such as age and clinical conditions, for instance respiratory sensitisation and predisposition to asthma. Therefore, in order to better understand aspects of biologically-healthy (and unhealthy) air alongside the effectiveness of their mitigation there is a pressing need to standardise acceptable limits for BioPM through the use of toxicological and epidemiological data in order to establish No-Observed-Adverse-Effect Level (NOAEL) and Lowest-Observed-Adverse-Effect Level (LOAEL) guidelines.

A necessary prerequisite for developing epidemiologically-guided threshold limits and targets is predicated on using accurate, standardised and reproducible measurements of BioPM, alongside better monitoring of human exposures; for instance measuring SARS-CoV virus in the air in order to monitor the efficacy of interventions (Ereth, Fine et al. 2021). The rapid

pace of innovation in analysing complex mixtures of BioPM includes the development of culture-independent high-throughput metagenomic and metatranscriptomic molecular approaches alongside increasingly comprehensive nucleic-acid databases and bioinformatic analyses to characterise and quantify BioPM. In parallel, methods for the online characterisation of bioaerosols using real-time fluorescence detectors entrained by machine-learning are leading to an increased discrimination of BioPM in realtime. We note here that there are no current UK established protocols for mould testing that enable benchmarking of exposures to moulds and their products in homes (UKCMB, 2018) although these protocols do exist for some environmental bioaerosols at regulated facilities (Environment Agency, 2018).

6.2.2 Culture-independent - Offline - measurement of BioPM

The diverse nature of the species that comprise BioPM necessitates diverse methods to sample their composition, distribution and abundance, recently reviewed by Whitby et al. (2022). While some collection methods have been in use for decades, there is rapid pace of innovation in the techniques used to assay BioPM post-capture that can be used in different environments and to answer a range of questions. Broadly, sampling BioPM uses either passive or active sampling approaches:

Passive sampling relies on gravitational settling or electrostatic attraction onto a sterile surface such as a petri dish, and therefore process low volumes of air. As the equipment is non-intrusive and simple, passive sampling designs are useful for extended sampling periods, in home environments or in citizen science BioPM surveillance. The method preferentially collect heavier particles and is therefore only semiquantitative; The Index of Microbial Air contamination (IMA; (Viani et al. 2020)) has standardised passive collection methods to an extent, expressing colony forming units (CFU) as CFU m^2, dm^2 or cm^2 per unit time as a scale of BioPM capture. Such culture-based techniques suffer from extensive bias as only a subset organisms are culturable and even non-viable microorganisms can still provoke disease. However, while classically used for culture-dependent enumeration of microorganisms, passively collected samples are suitable for analysis using molecular culture-independent methods such as quantitative polymerase chain reaction (qPCR) of target species (McKinstry et al. 2007), metabarcoding, metagenomic or metatranscriptomic pipelines to classify BioPM composition (Ferguson et al. 2022).

Active sampling screens a known volume of air by actively processing air into a liquid or particle collection surface. As active sampling can screen higher volumes of air compared to passive approaches, it is more suitable when concentrations of BioPM are low, such as in a hospital operating theatre, or where BioPM needs to be sampled in a time-proscribed manner. An increasingly wide variety of active sampling devices exist that actively collect particles of different size classes. Accordingly, the equipment and protocols are different when designing studies to collect small particles such as viruses compared against those studies analysing pollen and fungal spore densities. Modern active airsampling systems (Whitby et al. 2022) rely on (a) *filtration* onto cellulose, nylon, polycarbonate, gelatin or glass fibre filters using either high volume machines (eg. Research International SASS 3100) or personal samplers (eg. SKC IOM samplers), (b) *impactors* which select samples onto culturable surfaces (eg. Andersen impactors), (c) *impingers* that collect bioaerosols into liquids, which has been used for viral exposures assessments (eg. SASS 2300 Wetted Wall

Cyclone Sampler (Guo, Wang et al. 2020)), (d) *cyclones*, (e) *droplet-based microfluidic chips* for use in lab-on-a-chip analyses, (f) *electrostatic precipitation*, (g) *condensation* and *thermal precipitation* gradients and (h) detection of *volatile organic compounds* (MVOCs) of microbial origin.

Which air-sampler and method is best for which biological particle depends on the characteristics of the targeted microorganism(s) and the downstream analytical techniques. Downstream culture-based enumeration are limited as are time-consuming and constrained to the numbers of viable particles that will grow on the selective media and are biased to rapidly-growing taxa. However, culture is applicable if specifically targeted, for instance *Aspergillus fumigatus* and MCERT (Environment Agency, 2023) approved emission monitoring (Environment Agency, 2018); furthermore modern culturomics approaches uses high-throughput (including robotic) approaches to analyse microbiologically complex samples using multiple culture conditions in tandem with nucleic acid sequencing to further categorise species (Lagier et al. 2016). Increasingly culture-independent approaches now rely on the detection and classification of the nucleic-acid signatures of BioPM using molecular approaches.

Targeted approaches to quantifying BioPM using molecular methods relies on combining PCR with primers that are either specific to the microbe in question (such as SARS-CoV-2; (Guo et al. 2020)) or are universal with the most commonly-used gene-targets being the 16S ribosomal RNA gene for bacteria and the internal transcribed spacer (ITS) region of the ribosomal RNA gene for eukaryotes (Whitby et al. 2022). Developments on basic PCR include the use of probe-based detection and quantification (qPCR) and loop-mediated isothermal amplification (LAMP), both of which can be configured to detect RNA as a proxy for viable organisms. An advantage of PCR is that it has extremely high sensitivity and specificity, enabling the detection of the very low sample biomass that often characterizes indoor BioPM samples from clean environments such as hospitals.

The rapid increase in sequencing capacity driven by innovation in both short- and long-read technologies is associated with rapidly decreasing costs, with the result that it is now possible to analyse BioPMs using metagenomic and metatranscriptomic techniques. Previously necessitating high concentrations of input DNA, ultra-low biomass BioPM pipelines are now available for both active and passive-sampling approaches in order to subsequently characterise biological species at the genus level, see Figure 6.4 (Luhung et al. 2021). Substantial progress is still needed in several areas however: Data analysis and storage requirements are computationally challenging, as are the bioinformatic skills needed to process and interrogate high-dimensionality metagenomic datasets. Moreover, the methods are semiquantitative as the inferred abundances of taxonomic species are relative rather than absolute owing to variations in protocols, the presence of nucleic-acid inhibitors, molecular reagents and queried databases. However, these barriers are not insurmountable and metagenomic analysis will become a key component of future BioPM characterisation. In future, the innovation of single-molecule sequencing using portable real-time devices (e.g. MinION nanopores) that can be used in field-settings and can directly interact with online nucleic-acid databases means the real-time discrimination of metagenomic and metatranscriptomic BioPM is now achievable (see below). The small size and microfluidic nature of nanopore sequencing lends itself to integration into autonomous robotic workflows.

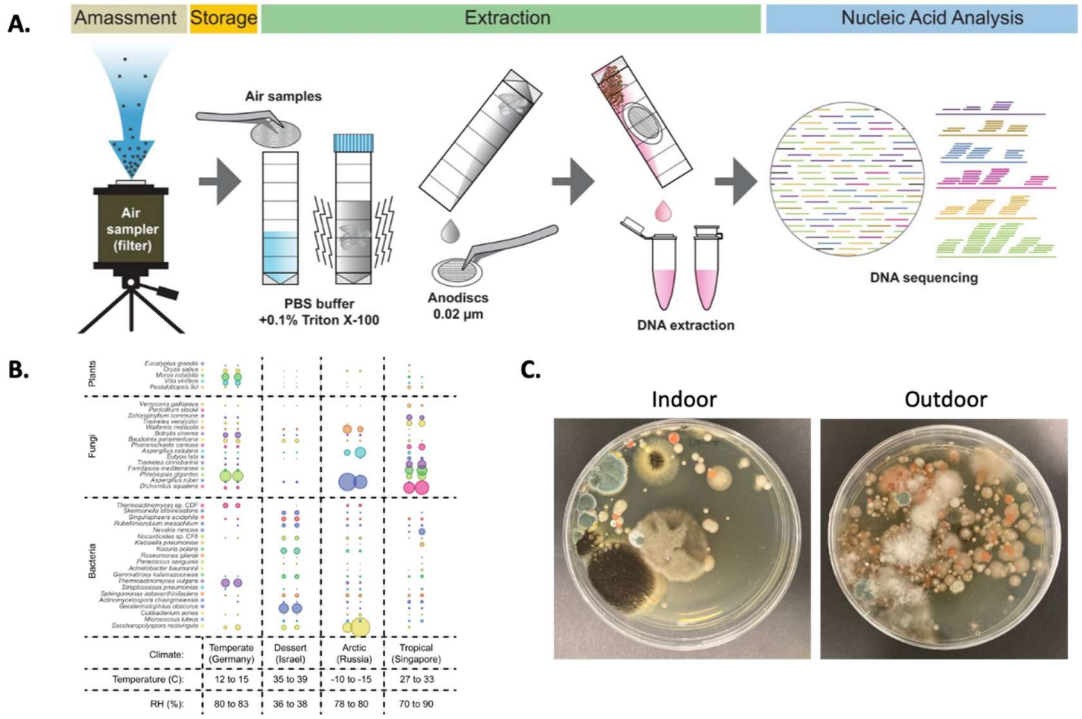


Figure 6.4: A. Example pipeline for the recovery of ultra-low biomass BioPM suitable for qPCR, metabarcoding or metagenomics using active air sampling and nucleic acid extraction; B. Detection, discrimination and enumeration using illumina short read sequencing and metagenomics of the relative abundance of the most frequently encountered species from exemplar temperate, desert, sub-arctic and tropical climates (source: Luhung et al. 2021); C. Gravitational settling (passive air sampling) onto sterile petri plates followed by subsequent culture from an indoor and outdoor exposure

Understanding the air resistome - The increasing public health burden of antimicrobial resistance (AMR) necessitates surveillance of environments that may harbor and transmit drug resistant microorganisms as bioaerosols. An Environment Agency review (Environment Agency 2020) concluded that studies on airborne AMR are lacking, and recent evidence suggests that ~5% of airborne *Aspergillus fumigatus* mould spores are resistant to agricultural and clinical azole fungicides in the UK (Shelton et al. 2022), which can be detected using suitable passive air sampling and culture-based BioPM protocols. Moreover, the comprehensive nature of metagenomic datasets means that antimicrobial resistance genes (ARGs) or resistance alleles present in the genome or plasmids will be found if the depth of nucleic acid sequencing is adequate and ARGs / alleles are represented in genetic databases.

6.2.3 Culture-independent - Online - measurement of BioPM

Online measurements of BioPM are designed to provide ‘real time’ measurements of BioPM through interrogation of spectral and morphological properties of detected particles. Converting these instrument response functions into real-time analytics, or classification of BioPM, exists in varying levels of maturity. This is in part driven by challenges in accessing appropriate standards of controlled experiments of BioPM combined with varying levels of information held within these instrument response functions as we increase spectral resolution and optical processing technologies. This also results in some online technologies

that are able to sit within existing networks of sensors and deliver characterisation of detected BioPM through appropriate (Application Programming Interfaces (APIs). Others remain directly useful to the academic community where a number of post processing steps and interrogative analytics are required to infer the broad type of BioPM that has been sampled.

As identified in the previous section, various offline methods require manual interpretation of samples collected onto filters using a mixture of semi-automatic microscopical assay and post analysis DNA extraction techniques. The existing UK pollen detection network relies on a seven-day volumetric spore trap from which daily average pollen counts can be obtained. Lack of temporal resolution, and problems with collection efficiencies, makes quantification of actual airborne concentrations and links to changes in environmental factors difficult (Huffman et al. 2020). Ultraviolet Laser Induced Fluorescence (UV-LIF) instruments measure the size, shape and the fluorescence of individual airborne particles across multiple wavelength bands, corresponding to the emission maxima for important bio-fluorophores (e.g. Tryptophan and NADH). For example, the WIBS-4A, and more recently the WIBS-5 (Wideband Integrated Bioaerosol Spectrometer) instrument [DMT Inc., Boulder, CO] records particle fluorescence over two wavebands, 310–400 and 420–650 nm; the UV-APS (TSI Inc., Shoreview, MN) records particle fluorescence across 64 channels from approximately 400-650nm. The WIBS-5 uses a particle size calibration based on the calculated MIE theory curve determined by the cavity and laser design. It collects forward scattering forward scattered light captured by a Quadrant detector to determine an estimate of particle shape, or more correctly, scattering asymmetry. The working size range is stated as 0.5 to 30 micrometres.

The Rapid-E, developed by Swiss company PLAIR uses a deep-UV laser (320 nm) to induce a fluorescent spectrum captured over 32 channels within a spectral range of 350–800 nm. This is combined with eight sequential acquisitions with 500 ns retention and recorded lifetime which has been shown to lead to improved particle identification (Šauliene˙ et al. 2019). By default, the deep-UV laser works for the 5–100 μm particle size range though the instrument can be used to sample particles in the range 0.5–100 μm (Šauliene˙ et al. 2019). Particles interact with the 400 nm laser light source and the scattered light is captured by 24 time-resolving detectors distributed at different angles. A number of studies have now demonstrated the use of deep learning architectures for pollen identification with the Rapid-E (Šauliene et al. 2019, Matavulj et al. 2021), by combining the fluorescence and scattering information. The Poleno/Jupiter instrument made by Swiss company Swissens uses laser scattered triggers to reconstruct two focused images at 90 degrees from each other using digital holography as in Sauvaugeat et al. (2020). The instrument also records UV-induced fluorescence spectra and lifetime measured at three different excitation wavelengths (280, 365, and 405 nm) using five measurement emission windows between 320 and 720 nm. Finally, a measurement of the time-resolved optical polarization characteristics of the particle is acquired before it exits the device. Swissens provide pollen classifiers through a series of data portals and real time analytics as part of the package. Combined with the PLAIR Rapid-E we see movement to novel real time classification of pollens across a number of case studies (Matavulj et al. 2021).

Realtime classification of other BioPM components remains an active area of research. This is not only connected to development of appropriate classifiers (e.g. deep learning

frameworks), but also appropriate training data. Numerous studies in the literature have evaluated the utility of laboratory generation of spores, bacteria and a range of fluorescent interferants (Forde et al. 2019). Sample generation and replicating atmospheric characteristics remains a challenge (Crawford et al. 2020; Pöhlker et al. 2012), though integration of multiple metrics from emerging UVLIF instruments previously noted may lead to development of accurate classifiers for non-pollen BioPM. Alongside supervised learning applications, converting instrument response to distinct observed BioPM types is achievable using unsupervised learning such as traditional cluster analysis. Multiple studies have demonstrated the utility of distance-based clustering methods which then require expert interpretation for assigning a likely BioPM type (e.g. Ruske et al. 2017). This requires comparison with ancillary meteorological data and creation of diurnal profiles to identify patterns that one might expect with, for instance, spore release. Unfortunately, distanced based clustering methods suffer from increasing computational complexity and cost for even moderately sized ambient datasets. Whilst alternative methods remain largely unevaluated, there remains a need for improved controlled environments for improved evaluation of classification potential (e.g., Forde et al. 2017).

6.3 Toxicity metrics, including proxies

Oxidative stress has been shown to be linked to inflammation and to a variety of associated adverse effects in the human body (Ayres et al., 2008). It has been postulated that airborne particulate matter exerts effects on health by causing oxidative stress and this has led to development of a number of *in vitro* acellular and cellular methods for assessing Oxidative Potential (OP). These various tests are primarily sensitive to the main chemical components of particulate matter, and, to date, there is limited evidence that any individual OP test is a better predictor of health effects than $PM_{2.5}$ mass.

Ayres et al. (2008) defined three mechanisms by which particles exert Oxidative Stress Potential: chemical groups on fracture surfaces (associated with exposure to quartz), Fenton chemistry (welding fume, PM_{10}, asbestos) and organic chemical redox cycling (diesel exhaust particles, PM_{10}). All mechanisms generate Reactive Oxygen Species (ROS) capable of causing inflammation and damaging tissue, and the various OP tests respond to different mechanisms of ROS production, and different ROS species. Hedayat et al. (2015) compared four tests (DTT, ascorbic acid, DCFH-DA and BPEA-nit in DMSO), all of which respond primarily to different particle components. According to Fang et al. (2019), H_2O_2 and O_2^- production are driven by Fe and Cu ions, while OH radicals are produced mainly from organic compounds and Fenton-like chemistry of metal ions. They report that humic-like substances can contribute substantially to ROS generation from biomass burning aerosol. Probably the most widely used test is the dithiothreitol (DTT) assay which is most sensitive to PAH and quinones, while ascorbic acid consumption relates most closely to transition metal concentrations. The dichlorofluorescein-diacetate (DCFH-DA) method depends upon catalysed oxidation by H_2O_2, while the BPEA-nit reagent is responsive to a range of radical species.

Daellenbach et al. (2020) carried out extensive measurements of both chemical composition and Oxidative Potential (by the DTT, DCFH and ascorbic acid methods) on particulate

matter samples from across Europe. They concluded that the mass concentration of PM was related mainly to secondary inorganic components, crustal material and secondary biogenic compounds. In contrast, anthropogenic sources, in particular fine-mode secondary organic aerosols largely from residential biomass burning and coarse-mode metals from vehicular non-exhaust emissions were the main contributors to OP.

Daellenbach et al. noted that if oxidative potential was causally linked to the major health impacts of PM, it may be more effective to control specific sources of particulate matter rather than overall PM mass. However, while COMEAP (2022) acknowledged that oxidative stress is believed to play a role in the capacity of inhaled PM to cause cell and tissue damage, they cautioned that OP metrics derived in acellular models reveal only the inherent capacity of PM to directly drive damaging oxidation reactions. Many PM constituents which are not directly redox active can generate reactive oxidative species (ROS) within cells, for example through the stimulation of cellular enzymes, as a function of their metabolism, or by induction of inflammation (COMEAP, 2022). COMEAP reviewed a number of epidemiological studies which had used OP as an exposure metric. The results were very mixed, with some studies showing an OP metric to be a better predictor of the health outcome than mass, some concluding that mass was the better predictor, and some finding OP to be an effect modifier of outcomes attributed to mass (COMEAP, 2022).

Chapter 7 - Data and information systems

7.1 Data infrastructure

The ever-increasing quantity and variety of air quality and PM data from an increasingly diverse range of instruments and models creates new demands for data dissemination and archiving. Data must be appropriately standardised, organised and be made 'findable' to researchers, practitioners, and the general public. Broadening data access creates new opportunities for improved information, analysis and understanding of air pollution.

Once data is collected, there need to be appropriate mechanisms for processing, archiving, sharing and thus generating insights. At each stage there are several software and data processing options, including the use of manual offline post processing through to automated streaming of data in near to real-time. For example, we might consider a mass spectrometer to require significant expert intervention, from calibrations through to post processing and then manual archiving on local and/or national database servers for offline analysis. In this example there are linear dependencies from data collection and then post processing of instrument response functions using vendor supplied and/or community derived software. On the other hand, a distributed network of $PM_{2.5}$ sensors, for example, might deliver data thorough a vendor specific API [application programming interface] to a user defined database or directly through a bespoke portal with no user intervention. Both divergent approaches however share the need for clear use of standardised approaches and reproducible best practices from the point of processing an instrument response through to data archiving and delivery. This section focuses on the latter. There are a number of community standards and database technologies available to improve accessibility and sustainability of platforms.

Standards allow many technologies to be interoperable within a network (OGC Reference Model. The Open Geospatial Consortium Reference number: OGC 08-062r7 http://www.opengis.net/doc/orm/2.1 Version: 2.1.) A definition of interoperability provided by the Data Interoperability Standard Consortium states that "*data interoperability addresses the ability of systems and services that create, exchange and consume data to have clear, shared expectations for the contents, context and meaning of that data*". Standards facilitate more agile integration of new networks of sensors or emerging sensing technologies. However, standardised approaches are not necessarily widespread across the air quality domain. In 2010 a DEFRA-commissioned study recommended a standardised approach for linking data, defined UK data formats, metadata standards and the need to reach compliance with the INSPIRE Directive (Spatial Information in the European Community). As a result, adding new sources to the open UKAIR service are required to comply with the INSPIRE Directive; a set of rules on interoperability that define how to publish and share spatial data across the public sector. These broad directives provide overarching guidance and data management principles. For example, '*Data should be collected only once and kept where it can be maintained most effectively*' (https://inspire.ec.europa.eu/inspire-principles/9). These broad Directives include guidance on relevant *ontologies* and *metadata*

standards, both of which provide more formal structures to define the software required at the interface between the air quality sensor and database.

Ontologies: Ontologies provide the vocabulary framework that describe different components and flow of information such that data can be exposed through APIs, for example. From an air quality data access perspective, the API exposes the chosen ontology. A number of ontologies are used in the environmental monitoring sector. These include:

- The Semantic Sensor Network (SSN) Ontology: The SSN Ontology describing sensors and their observations, the involved procedures, the studied features of interest, the samples used to do so, and the observed properties, as well as actuators (https://www.w3.org/TR/vocab-ssn/. It provides a vocabulary for describing environmental sensors and the data they produce, as well as the relationships between different sensors and observations.
- The Sensor Observation Service (SOS) Ontology: The SOS Ontology extends the SSN Ontology and provides a vocabulary for describing sensor observations, including spatial and temporal information.
- The OGC Observations and Measurements (O&M) Ontology.

Metadata: Unlike an ontology, Metadata provides information about data, such as its format, creator, source, etc. Metadata solutions do not rely on a chosen ontology, but are complementary components. The adoption of a given meta-data schema can depend on the sensor characteristics, but also the supplier's business model. From an air quality perspective, take the example of a city collecting data across a network from multiple sensors vendors. Two separate vendors of networked air quality sensors, using the same underlying technology, may not share a meta-data standard. Air quality data can be provided with a set of metadata descriptors consisting of:

- name of the quantity measured (i.e., NO_2, $PM_{2.5}$, etc.)
- unit of measurement
- time of the observation
- location

Information could also be provided about the devices generating the data and any associate provenance that might be useful when interrogating interventions, for example:

- id of the device
- location
- measurement count
- power status (battery voltage, battery charge, external voltage)
- operational status:
- current operational status of the device
- 'calibration flag'

The Central Digital and Data Office of the UK government has compiled a guide for APIs and data standards *(guide for APIs and data standards)*. This includes recommendations on adoption of the JSON format delivered through an API query, for example. JSON objects are much preferable to a numerical array since each observed value could be associated

with the sensor state metadata, for example. The previously mentioned INSPIRE directive requires implementing rules around metadata standard maintenance are adopted. It has been recognised that enforcing existing metadata requirements can quench wide scale integration of diverse data streams with varying volume (Klump et al. 2023). The Open & Agile Smart Cities (OASC) network introduced the concept of Minimum Interoperability Mechanisms (MIM) that allow different data solutions provided by different sensor vendors provided there are agreed 'critical' components defined by the relevant stakeholders.

All of the above can be modified for a given set of air quality networks. For example, the UKCRIC Urban Observatory programme moved towards standardisation of air quality data streams across Newcastle, Birmingham and Manchester as inspired by the SSN/SOSA ontology.

Database technologies: Whether an appropriate metadata schema and/or ontology is adopted for expanding air quality networks, there are also dependencies on how and where the data is stored and archived separate to any near-to-real-time delivery. There is now a spectrum of options, ranging from local 'on premise' to cloud native options (e.g. Amazon Web Services), each with resource and maintenance skillset considerations to be made. Local, or 'on premise', solutions typically involve a dedicated server with a software stack that could be built around one of the ontologies outlined above. As previously mentioned, whilst some research grade instrumentation requires rigorous expert intervention to then provide manual data deposition on a central repository, there are also streaming options available. As an example, a local solution could include a 'front end' that any user interacts with (e.g. written in ReactJS) and a 'backend' that maintains a metadata instance (e.g. written in MongoDB) and in turn pulls data from the network of sensors through the provided API services. An additional archive could then store the streamed data using e.g. InfluxDB. The choice of database software can vary. On the other hand, a cloud native solution could be built around cloud service provider toolkits. For example, this could include IoT specific toolkits provided by AWS who could handle availability, scaling and security. This last component is important; particularly if the data streams are to be used to inform agile interventions such as Digital Twins.

Summary: Integrating wider networks of air quality sensors can be supported through a combination of standards and software choices. This could enable researchers to integrate specialised datastreams into existing data portals. A review of appropriate overarching metadata standards and, perhaps, ontologies would be important to understand the most appropriate mechanism for integration across sampling technologies. Whilst adoption of the INSPIRE directive enforces alignment of community standards, a review of vendor supplied data standards would be needed. Modification of existing metadata standards for, say, indoor environments could be required. There are wider developments that will enforce these issues, including the development of Digital Twins. Whilst there is ongoing debate on the utility of Digital Twins, or Digital Shadows, for environmental and city scale monitoring (Topping et al. 2021), it is likely this will lead to proposed solutions to improve air quality through automated interventions. Wherever interventions are made, ensuring provenance in the data on which those decisions have been made is essential. Thus, any integration of new network data, or services built around that data, should present a clear roadmap for use or adoption of standards as described above.

7.2 Public dissemination

This section discusses opportunities for public dissemination of PM data, within the broader context of air quality information. It does not address issues or recommendations of the Defra AQIS Review (https://uk-air.defra.gov.uk/research/aq-system-review), which in progress at time of writing this report.

Public dissemination of information on air quality can help to raise awareness of concentrations, associated health effects, and sources of pollutants. The former can help vulnerable individuals to protect their health from exposure to episodic peaks in concentration e.g., the Daily Air Quality Index (DAQI), and airTEXT alerts (airTEXT.info) for Greater London and the south-east. Knowledge about sources can help the public more generally link behaviours to emissions and facilitate policy implementation, e.g. reducing solid fuel burning in urban areas.

Current sources of information about air quality include the DAQI, UK-Air datasets, air quality apps, weather apps, text alerts, newspapers, and local authority websites. In most of these examples, the provision of information is passive and therefore limited to those that opt in to finding out, i.e., via subscribing to text alerts (where available) or checking relevant websites or news articles, which often misses the most vulnerable populations (Ramírez et al., 2019). It is also recognised within behavioural science literature, that provision of information does not necessarily translate into action (McCarron et al., 2022; D'Antoni et al., 2019; Ramírez et al., 2019), and awareness raising without the apparent power or means to affect the situation can cause psychological stress and inaction (Heydon and Chakraborty, 2020).

There are opportunities to consider the advised given to citizens to encourage then to protect themselves and reduce emissions of PM, and on the wider action that is being taken and by whom. In this way public dissemination of PM data might be linked to messaging around what action is being taken by responsible authorities in response to pollution in order to reassure the public. Where behaviour change is a desired outcome, messaging might for example provide the public with constructive advice and illustrate what is being done to facilitate these changes in a way that is directly relatable to the individuals concerned. Further advice may be provided on how they can effectively action further improvements in air quality, e.g., though engagement with Local Government, NGOs or health charity campaigns, working with businesses, etc. (McCarron et al., 2022).

The 2022 update to the LAQM Policy Guidance in England (Defra, 2022) recognised the limitations of existing air quality data provision. A chapter on consultation and community engagement encouraged local authorities to provide communities with clear, accurate and timely information about local air quality to facilitate behaviour change. According to the guidance, local authorities should consider the broadest range of communication opportunities, including healthcare setting, supermarkets and sports facilities.

There is a need to communicate air pollution issues beyond those that may be most directly affected or engaged, including reaching more affluent sectors of society that may be responsible for, on a per-household basis, a larger fraction of emissions (Barnes et al., 2019). There are NGO-led activities with these wider communication aims, for example Global Action Plan's national Clean Air Day

(https://www.actionforcleanair.org.uk/campaigns/clean-air-day). Public health campaigns on air pollution, mirroring those for smoking, healthy diets and road safety, have the potential to improve the public consciousness of key sources of PM such as transport and home heating. Observational evidence from monitoring would likely form a key backdrop.

An approach to increasing uptake of air pollution policies is by involving the public in the development of interventions, e.g., through citizen assemblies (The Involve Foundation, 2020) where communities are actively engaged in creating policies that match their needs and lived experiences (McCarron et al., 2022, Bovaird and Loeffler, 2012). These approaches aim to remove more adversarial aspects of traditional top-down consultation and encourages buy-in, empower individuals and to shift normative behaviours through peer and social support for measures. Participative engagement methods can also utilise citizen science projects which may be led by researchers, policy makers or communities themselves, e.g., Air Quality Data: The Bristol Approach and HackAIR - How clean is the air you breathe? (Schade et al., 2021) (see also section 5.5). An evidence review conducted for the Scottish Government provided a summary discussion of public engagement on air quality, including participatory approaches (Barnes et al., 2020). As with any public communication, it is important to recognise the diversity of publics and ensure information and data dissemination is as inclusive as possible, using a wide range of media, modes and mechanisms (Ramírez et al., 2019). Engaging harder to reach groups can be facilitated through trusted gatekeepers and change agents.

There are issues specific to PM pollution and associated data that require targeted public dissemination. With some sources of PM emissions increasing (e.g. woodburning in cities, wildfires from extreme dry weather events) there is a need to communicate the effect that these trends are having on local air quality. There are also emergent sources that are gaining media attention, such as microplastics, that would benefit from improved messaging about the current state of scientific knowledge on prevalence and effects, and the research being undertaken to develop this understanding and inform policy. There may also be value in raising awareness of the contribution that secondary/background PM makes to local air pollution, particularly in light of the more restrictive national targets and WHO Air Quality Guidelines. Again, more nuanced observational data on the composition of PM may help support that process.

The National Environmental Public Health Tracking Program, from the United States Centers for Disease Control and Prevention (CDC), is a good example of a publicly accessible platform that brings together environmental and societal data from a number of sources in real-time to help users to observe and relate these datasets (Strosnider et al., 2019). This tool allows users to view and interrogate data on air pollution (e.g. $PM_{2.5}$) with potential sources (e.g. wildfires) and health outcomes (e.g. COPD prevalence). The Tracker is available as a web-based Data Explorer and API that can be incorporated into any website and provides a number of visualisation options at multiple spatial and temporal scales.

Pervasive technology, or smart devices, present an opportunity to directly engage individuals with air pollution (Carminati et al., 2017). This may be through wearable sensors (Fahimi et al., 2019) (with the user expectation that these represent real-world concentrations at a relatively fine spatial resolution). Unobtrusive sensors can also measure the indoor environments and help advise householders and landlords about the contribution from

different sources (e.g. wood-burners) and where ventilation may be required (Smeenk, 2023). Similarly, in-car/cab sensors can be used to detect build-up of pollutants within vehicles (Waworundeng and Adrian, 2021). Further improvements in sensor technology may create an opportunity for proactive emissions assessment, including PM and NO_x, outside of annual vehicle tests.

Social media and social networking platforms, such as Facebook, Instagram and TikTok, also have the potential for widespread sharing of air pollution data and messages, capturing of public experiences through multimedia and exploration of public perceptions through hashtag searches and content analysis. The key value of social media, however, is the ability to engage widely, not only with those that are primarily interested but also with their wider social networks. The opportunity this presents for affecting subjective norms makes this a particularly useful tool.

Air pollution apps providing information about local air pollution levels in real-time have become commonplace (e.g. Breathe London and Plume Labs App: Live and forecast air quality data). Google Earth also includes air quality data in some locations (although not the UK currently) and forthcoming immersive view maps using AI-generated air quality data will enable users to plan their journeys avoiding pollutant hotspots (I/O 2023: Google Maps updates Immersive View and launches new tools for developers (blog.google)). How such data is generated is however often unclear, or the extent to which it is based on observational data, or models. These apps have varying degrees of functionality, from just providing air pollution estimates for a chosen location, to providing advice on the lowest pollution walking route from one location to another or advising on the best time of day to exercise. These apps are also available on smart watches. This represents an opportunity, through making a problem that was invisible now potentially 'visible' at any time, in any place, but also a risk that people are frequently pushed potentially inaccurate information about a substantial risk with no actionable information about what they can do about it.

7.2.1 Communication through other stakeholders

Health professionals are a trusted source of information about risks to health, and research has shown that publics may be more receptive to information provided by trusted institutions than that provided by local or national government (Riley et al. 2021). However, existing training and current systems and resources have not always provided sufficient information to enable health professionals to discuss air pollution risk with patients. Addressing this was a specific recommendation of the Chief Medical Officer's annual report 2022 which said "*the training of healthcare staff should include the health effects of air pollution and how to minimise these, including communication with patients"*.

Great Ormond Street Hospital (GOSH) are partnering with Imperial College London to use air pollution measurement data to support health professionals and inform the public (Hayden et al. 2023). The hospital recognised that clinicians did not have easy access to information that would allow them to provide patient-specific advice on air pollution risk. GOSH surveyed clinicians and parents of patients to define the needs of these two groups. Only 15% of clinical staff felt confident discussing air pollution with their patients and 75% of families surveyed would like to know about air pollution risks in their area.

Figure 7.1 illustrates the approach taken to supporting clinicians by embedding air pollution data for patients' postcode with supporting information, and curated actions, directly into the electronic medical record (EMR). Since November 2022 annual average air pollution estimates for home postcodes have been displayed on the EMR Epic system and a communications strategy will follow to enable clinicians at GOSH to use this information. A trust-wide quality improvement project is now underway to expand teaching resources, increase staff engagement and to evaluate the project. There has been interest in this approach from other NHS trusts and local and national government. The way this has been designed means that this intervention can easily be replicated by other trusts and GOSH are discussing its implementation with interested parties.

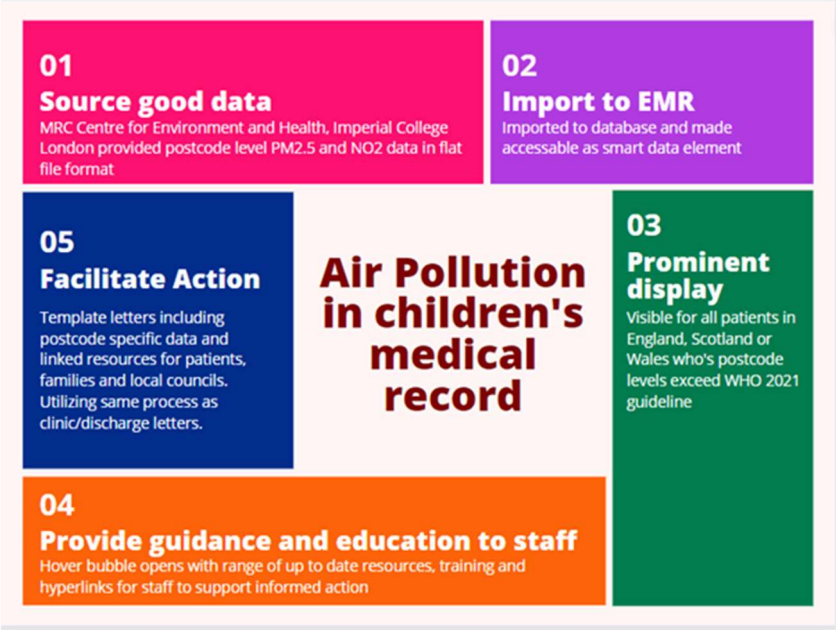


Figure 7.1: GOSH approach to introducing the inclusion of air pollution in children's medical records.

7.2.2 Future communications systems

There are many opportunities for improving and broadening the communication of information on air pollution concentrations, distribution, emission sources and health impacts in the UK, and new instruments and data methods can underpin some those. For example, future information systems may provide greater time or spatial resolution to forecasts, or place increased emphasis on actions that limit emissions of pollution during events. New systems may also be needed to improve understanding and communication of air pollution data for relevant professions such as healthcare.

There is currently a major review ongoing in Defra, titled Air Quality Information Systems (AQIS) that is considering these issues and opportunities for the future. See: https://uk-air.defra.gov.uk/research/aq-system-review for current information. AQEG is currently providing input to that review. At time of writing AQIS is approximately half-way through its two-year work programme, which includes a number of commissioned studies of existing provisions, public engagement and how advice on air pollution may reach new audiences. In order to avoid conflicting or partially complete advice, this report will not detail further detailed areas for future communication systems.

7.3 Active air quality management

Historically, most air quality management interventions have been permanent or semi-permanent responses to protracted air quality issues, typically relating to annual mean objectives or limit values. The improved availability and affordability of highly time-resolved measurements, coupled with faster communication and data processing, allows for rapid-response interventions which react to air quality observations. These rapid interventions might still serve to reduce long-term exposure, but more often focus on reducing exposure to short-term episodes.

7.3.1 Measurements Informing Action to Control Emissions

It is common to use near real-time PM monitors to both regulate and control emissions from construction sites. Most often using nephelometers, instruments are placed around the construction site boundary, with results compared in real time to predefined trigger levels (Figure 7.2). Exceedances of these trigger levels can then prompt alerts to the local planning authority and to the site manager (e.g. DfT, 2017). In principle, sending an alert to the mobile phone of the site manager allows rapid action to be taken to either increase dust suppression or to stop the activity until a remedy is found. In practice, the effectiveness depends on both ability and willingness to impose remedial action. The measurements nevertheless provide a helpful safeguard even at well managed sites.

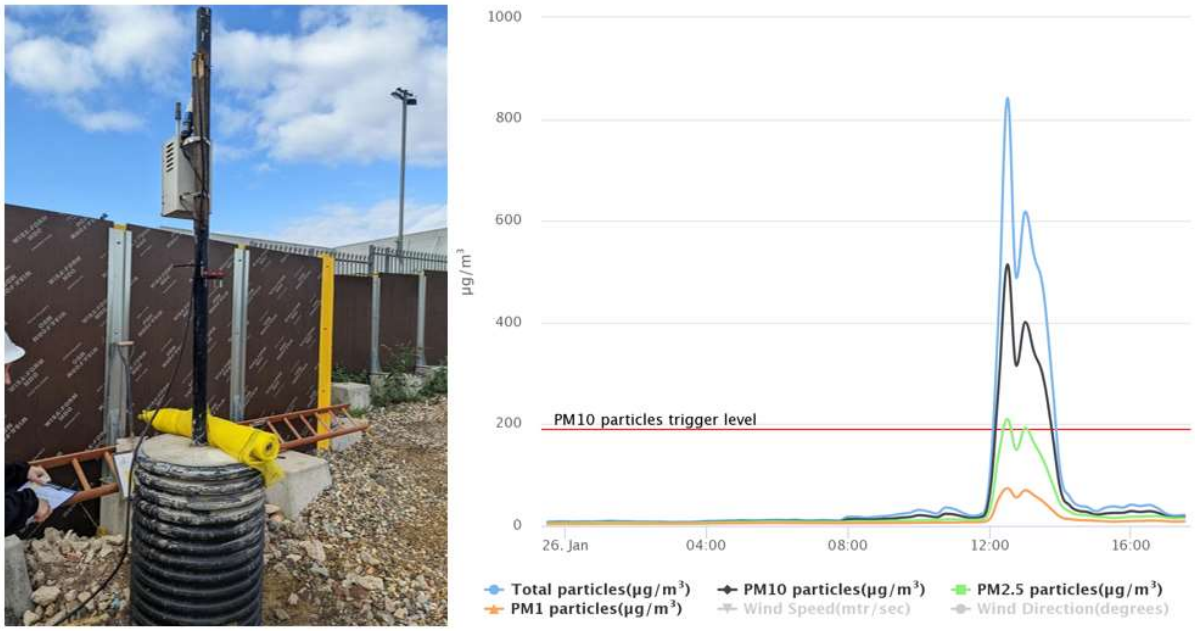


Figure 7.2: PM Monitoring by Nephelometer at a Construction site with live data feed. Photo Joe Rondell, Air quality consultants.

Ambient $PM_{2.5}$ measurements have also been used to control domestic wood burning. The 'burner alert' system (Chakraborty et al., 2023) links measurements from sensors in Sheffield with online and app-based alerts which seek to temporarily discourage stove owners from lighting their appliance based on the previous 24-hour mean concentrations.

When using ambient measurements to control emissions more directly, it is most practical to focus on the pollutants which give the clearest signal from that source. Since most combustion sources generate larger relative local increments of pollutants such as CO or NOx than of PM, it is unlikely that PM measurements will provide an optimum solution controlling these sources regardless of the measurement technology used. This may, however, change as the spectrum of emissions sources evolves, for example with widespread electrification of the road vehicle fleet (AQEG, 2019).

Urban Traffic Management and Control (UTMC) systems coordinate groups of traffic signals to better manage traffic flows around towns and cities. While they primarily target congestion and journey times, they can also consider air quality (e.g. DfT, 1999). UTMC systems have also been linked with ambient measurements via emissions and/or dispersion models (e.g. Figure 7.3). UTMC algorithms then calculate optimal traffic solutions in real time based on the operators' chosen priorities (Galatioto et al., 2011, Rose et al., 2012, Gustafsson et al, 2012). Published studies have demonstrated the potential for integration with UTMC systems but have not sought to quantify the air quality benefits. Systems are nevertheless now being installed commercially (e.g. Liverpool City Region, 2021).

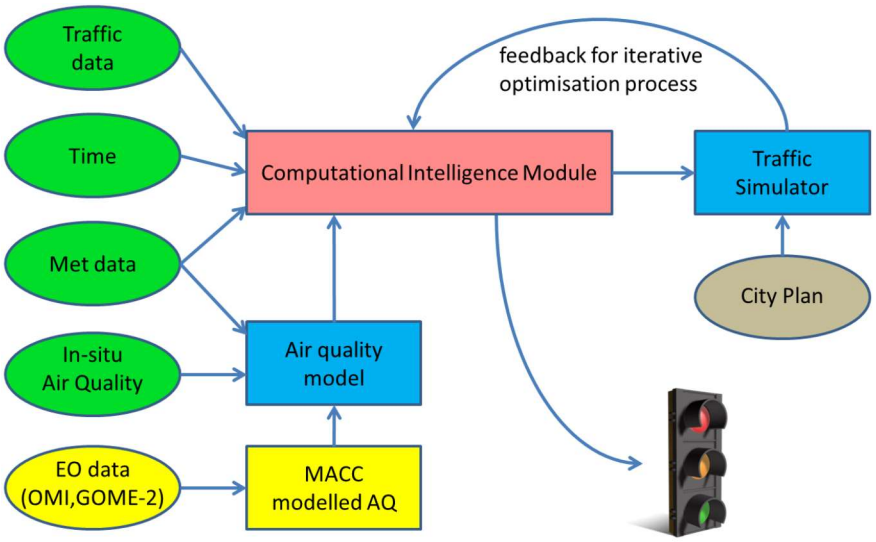


Figure 7.3: Integration of Traffic Management and Air Quality Control (Gustafsson et al., 2012).

Within-stack measurements are used routinely to control industrial emissions, with the measurements reported to the regulator (thus acting as a safeguard to excessive emissions) and also often providing diagnostic feedback to optimise the process and thus minimise average emissions. While ambient measurements of other pollutants have been used to actively control industrial activity (for example SO_2 measurements were used to control activities at the Fawley oil refinery - NFDC, 2018), using PM measurements in this way is often complicated by first needing to determine the precise emission source, since industrial areas can include multiple operators and releases (e.g. AQEG, 2011).

Using PM measurements to actively control emissions from other sources is uncommon. However, the use of PM monitors to control construction emissions demonstrates the effect that real-time observations can have when coupled with readily useable and practicable actions.

7.3.2 Measurements Informing Action to Control Exposure

People might chose to limit their personal exposure to elevated ambient PM concentrations if data made them aware of risks. This might be from published alerts (see Section 7.2), involvement with citizen science or from information linked to personal monitoring devices. Real-time air quality information is also increasingly provided as a commercial opportunity via smart devices and other products. This may combine dispersion modelling, local monitoring of air quality and activity data, and machine learning / data analytics, to provide a near real-time concentration field. Reported commercial applications include navigation (automotive, eBike, and hand-held products), sports (advice on exercise location and timing), ventilation control (indoor and in car cabin), and even cosmetics (marketing products linked to personal environmental exposure) (e.g. BreezoMeter, 2023).

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