Publishable Summary for 16ENV02 Black Carbon
Metrology for light absorption by atmospheric aerosols

Overview

The measurement of particles in the air characterised as black carbon is important both for their role in climate change and as a measure of combustion products associated with health effects. Measurements are made very widely, and compact, precise, real-time, relatively inexpensive instruments are available. Although it is conceptually a simple measure of the light-absorbing properties of airborne particles, the metric does not currently have SI traceability, with consequences for the comparability and interpretation of data. The project made substantial steps towards providing a workable solution to this major problem, using a combination of primary methods (which do not need to be calibrated with a black carbon calibration sample), black carbon reference sources, and a robust calibration protocol that addresses the fact that commonly-used instruments will have more than one type of measurement artefact.

Need

The quantity of airborne particles loosely described as black carbon has been widely measured by various optical methods since the early 20th century, because instruments for this are relatively simple and reliable. The dominant sources have changed over the decades, from domestic and industrial coal burning to vehicle combustion emissions, with more recent contributions from wood-burning.

Black carbon has been identified as the second most important climate forcing agent behind CO_2, contributing an amount of radiative forcing nearly 30 % that of current CO_2 concentrations. Airborne particles have serious human health effects across Europe and worldwide. In 2011, about 430,000 premature deaths in the EU were attributed to fine particulate matter (PM). Studies suggest that black carbon is a better indicator of harmful particulate substances from combustion sources than PM mass concentration.

Although black carbon measurement is in principle a simple optical measurement of absorption, characterised by the aerosol light absorption coefficient, traceability is hampered by the fact that routine monitors determine the absorption of particulate matter collected on a fibrous filter. While the optical absorption measurement itself can be done accurately, the presence of the filter has a large effect, due to internal scattering within the filter, which can increase absorption by a factor of five, and shadowing effects as the filter accumulates material. Empirical but non-traceable correction factors are then incorporated into the conversion from light absorption coefficient into the reported particle mass concentration; these correction factors need to be replaced with properly determined calibration factors in order to standardise the measurement results and ensure confidence and comparability in the field.

Objectives

The overriding objective of the project was, for the first time, to bring SI traceability to field of black carbon measurements, so that their accuracy and value would be greatly increased. The specific objectives were:

1. To establish a set of well-defined physical parameters, such as aerosol light absorption coefficients and mass absorption coefficients, which together can be used to quantify black carbon mass concentrations with traceability to primary standards.
2. To develop and characterise black carbon standard reference materials (SRMs), representative of atmospheric aerosols, together with methods for using them to calibrate field black carbon monitors.
3. To develop a traceable, primary method for determining aerosol absorption coefficients at specific wavelengths that are to be defined for the benefit of users. The method should have defined uncertainties and a quantified lowest detection limit.
4. To develop a validated transfer standard for the traceable calibration of established absorption photometers such as multi angle absorption photometers, aethalometers and particle absorption

photometers. The transfer standard should make use of the black carbon SRMs (developed in objective 2) and associated portable instrumentation characterised by the primary method (from objective 3).

5. To facilitate the take-up of the technology and measurement infrastructure developed in the project by standards developing organisations (CEN, ISO) and end users (e.g. Environmental Protection Agency (EPA), European Environment Agency (EEA), World Meteorological Organisation-Global Atmosphere Watch (WMO-GAW), the ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network) Project).

Progress beyond the state of the art

Before the start of the project black carbon measurements were being widely made, based on a principle that has been used for many decades, with one of several designs of filter-based instrument, such as multi-angle absorption photometers, aethalometers and particle absorption photometers. However, robust calibration techniques and traceability to the SI for these instruments were outstanding issues, such that results from different types of instrument can disagree by up to 30 %. This project has recommended reference aerosols for the calibration of those filter-based techniques that will put traceability and calibration mechanisms in place for the first time, enabling the measurement of black carbon with a target uncertainty of ±10 % (95 % confidence level).

One important element of progress was the realisation that the original strategy of developing a “near-black carbon source” was not a viable approach. This is because filter-based instruments need corrections that depend on both particle size and the mixture of optical properties in the aerosol particles, neither of which are determined directly. Consequently, an instrument “corrected” to provide accurate measurements of the near-black calibration aerosol would be unlikely to give accurate measurements of ambient air, because the size and optical properties of the particles in ambient air would be different to the calibration particles. This problem will be avoided by the requirement for two distinct categories of calibration aerosol, with similar size and optical properties to distinct types of ambient aerosol towards the opposing extremes for what is likely to be encountered.

Results

1. Physical parameters for traceable quantification of black carbon mass concentrations

The physical properties of aerosols (and the particles within them) that are relevant to black carbon measurements have been clearly defined, in a way that clarifies how traceability to the SI can be established. A key point in this context is that, because the presence of a filter affects the measured extinction in at least two distinct ways, it is necessary to calibrate filter-based instruments with at least two types of aerosol, with contrasting physical properties. This will prevent the calibration of an instrument in a way that is correct only for aerosol particles with a specific set of properties. From consideration of how filter-based instrumentation introduces artefacts into the measurement, the two key parameters were chosen to be particle size, to characterise the penetration of ambient particles into the filter used by the instrument, and single scattering albedo (SSA, the ratio of scattering efficiency to extinction efficiency) – to characterise the loading and scattering effects of ambient particles within the filter. This result was critical to the successful achievement of the project objectives.

*2. Black carbon Standard Reference Material*s

Standard Reference Materials, together with a method for their controlled introduction to black carbon instruments in the lab and in the field, were developed and tested. The required properties of these two Materials were defined as (1) “fresh combustion particles” with size 50 – 100 nm and Single Scattering Albedo (SSA) 0.05 – 0.2 at 550 nm, and (2) “aged combustion particles” with size 200 – 400 nm and SSA 0.7 – 0.9 at 550 nm. Several methods for generating them were tested and evaluated at TROPOS and at METAS. A new type of premixed flame combustion generator, which provides soot particles with stable EC/OC content independent from the soot particle size, has been found suitable for producing “fresh” aerosol, along with other variations and techniques. The new soot generator combined with a micro-smog chamber to coat the soot particles with organic material has been found suitable for generating an “aged” aerosol. It was found that the particle dilution system should be considered as an integral part of the generation system. A peer-reviewed paper covering this has been published. Again, these results were critical to the successful achievement of the project objectives.

3. A traceable, primary method for determining aerosol absorption coefficients

Laboratory-based, SI-traceable methods for determining aerosol absorption coefficients at specific wavelengths, based on several different principles, were evaluated within the project. The options included established techniques such as extinction minus scattering (EMS), either with separate or combined extinction and scattering instruments, photoacoustic instruments, and the more novel technique of photothermal interferometry (PTI). Several refinements to the methods were made, for example to the baseline correction for extinction measurements and to the design of photothermal interferometry instruments, with several resulting peer-reviewed publications. Methods need to be suitable both for calibrating instruments in the laboratory, or if possible in the field, and for certifying the properties of the Standard Reference Materials of Objective 2. Two potential Si-traceable primary methods were identified. These are the well-known EMS method, based on a combination of nephelometer and extinction cell, and the PTI technique. Due to limitations in portability, these setups are not well suited to field calibration in their current state of development. Instruments combining extinction and scattering, and photoacoustic instruments, are considered as secondary standards, because direct calibration to SI units is lacking. However, these units are better suited as field references.

All methods are subject to a high detection limit when compared with typical values of the absorption coefficient for ambient air. In addition, the EMS methods have higher uncertainties at high single scattering albedos, that is, when the airborne particles are predominantly light rather than dark. Therefore, field calibration of filter-based absorption photometers with ambient air is only possible in rare cases. However, with a transportable black carbon generator to produce black carbon particles with reproducible properties, field calibrations would be feasible and would also reduce the uncertainties due to gas absorption. It should be noted that such a black carbon generator would also support cross-calibration of photoacoustic instruments in the field. These results enabled the project to successfully achieve its objectives.

4. A validated transfer standard for calibration of field absorption photometers

A substantial field comparison of black carbon instruments was carried out in Pallas, Finland in June/July 2019, at low concentrations. A second field trial took place in Athens, Greece in November 2019 – January 2020, to investigate higher concentrations. Both campaigns were used to trial provisional calibration procedures developed during the project.

These results have provided a clear route for calibrating commonly-used black carbon instruments, and so allowed the project to successfully achieve its objective.

Impact

Impact activities within the project included establishing a Stakeholder Committee; presentations to relevant standardisation and metrological committees, and scientific conferences, including the ETH Conference on combustion generated nanoparticles, the European Aerosol Conference and the International Aerosol Conference; training to 24 groups of people provided at TROPOS and at monitoring sites; and 5 papers published in peer-reviewed journals. One paper, *Radiative properties of coated black carbon aggregates: numerical simulations and radiative forcing estimates*, by Baseerat Romshoo et al, won the best poster award at the 2020 European Aerosol Conference.

The results achieved in the project were appropriately communicated to the stakeholders and end-user community of EC Directives and European standardisation activities such as CEN and BIPM (CIPM). Input and feedback was obtained from this community to improve the project impact and its outcomes.

During the course of the project the consortium ran a regular series of intercomparison and training workshops covering absorption photometers (for black carbon measurements) and integrating nephelometers (for scattering measurements).

Several of the partners were directly involved with the operation of black carbon monitoring sites for scientific and national network purposes, so that the results of the project were being implemented directly, for example through the establishment of improved calibration facilities and procedures.

In the short term, further impact will follow from laboratory calibration using reference sources of both “fresh” and “aged” combustion aerosol that are traceably-characterized in the laboratory at the time of calibration. Comparability and accuracy for aerosol particle light absorption measurement will be significantly improved. It would be premature to give definite figures for this, but there are indications that uncertainties will be reduced from around 30 % to around 10 % (at 95 % confidence).

In the medium term, as reproducibility of the aerosol sources is improved, and further validation of the laboratory calibration procedures is carried out, it should be possible to perform field calibrations of black carbon instruments, either with aerosol sources combined with a reference absorption instrument, or reference aerosol sources on their own.

Impact on industrial and other user communities

In commercial terms the project will give a direct advantage to European black carbon instrument manufacturers, who will have early access to traceable calibration facilities for their current instruments, and who will also make use of the facilities to develop innovative designs much more quickly than would otherwise be the case. It will also offer a great advantage to European manufacturers of aerosol generators of the type that have been selected within the project as suitable to be calibration sources. End users would include government, environmental and citizen monitoring groups, who all employ black carbon measurement devices.

Impact on the metrology and scientific communities

The simplest direct impact of the research will be that measurements of black carbon become more accurate and more comparable in the aerosol monitoring networks across Europe, through the development of reference calibration sources for black carbon, primary national facilities and traceable calibration mechanisms.

Impact on relevant standards

The project outputs are expected to provide the basis for new documentary standards for monitoring black carbon by European and International standards-developing organisations like CEN and ISO. No such standards currently exist, and project partners regularly participated in the standardisation groups which are likely to develop them in future.

Longer-term economic, social and environmental impacts

In terms of socio-economic benefits, the project outputs will potentially lead to revised air quality legislation, based on black carbon, for which reliable measurement methods will be available.

Indirectly, the impact will be very widespread. In terms of scientific benefits, the improved measurements will be used directly within EU atmospheric aerosol projects, refining climate change models and mitigation proposals, and improving the quality of conclusions from cohort health studies looking at the effects of air pollution. Air quality measures to reduce black carbon emissions such as emission reduction and low emission zones have already been taken. However, traceable black carbon metrics to reliably quantify the success of these measures are not yet available. The results of the project will allow them to be addressed.

List of publications

1. Michaela N. Ess & Konstantina Vasilatou (2019) Characterization of a new miniCAST with diffusion flame and premixed flame options: Generation of particles with high EC content in the size range 30 nm to 200 nm, Aerosol Science and Technology, 53:1, 29-44, DOI:10.1080/02786826.2018.1536818 https://www.tandfonline.com/doi/full/10.1080/02786826.2018.1536818
2. Rob L. Modini, Joel C. Corbin, Benjamin T. Brem, Martin Irwin, Michele Bertò, Rosaria E. Pileci, Prodromos Fetfatzis, Kostas Eleftheriadis, Bas Henzing, Marcel M. Moerman, Fengshan Liu, Thomas Müller, and Martin Gysel-Beer, (2021) Detailed characterization of the CAPS single scattering albedo monitor (CAPS PMssa) as a field-deployable instrument for measuring aerosol light absorption with the extinction-minus-scattering method, Atmospheric Measurement Techniques https://doi.org/10.5194/amt-14-819-2021
3. Michaela N. Ess, Michele Bertò, Martin Irwin, Robin L. Modini, Martin Gysel-Beer &Konstantina Vasilatou, (2021) Optical and morphological properties of soot particles generated by the miniCAST 5201 BC generator, Aerosol Science and Technology https://doi.org/10.1080/02786826.2021.1901847

4. Sascha Pfeifer, Thomas Müller, Andrew Freedman, and Alfred Wiedensohler, (2020) The influence of the baseline drift on the extinction values of a CAPS Pmex, Atmospheric Measurement Techniques https://doi.org/10.5194/amt-13-2161-2020

5. Visser, B., J. Röhrbein, P. Steigmeier, L. Drinovec, G. Močnik, and E. Weingartner, (2020) A single-beam photothermal interferometer for in-situ measurements of aerosol light absorption, Atmospheric Measurement Techniques, https://doi.org/10.5194/amt-13-7097-2020

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

<table>
<tr><td colspan="2">Project start date and duration:</td><td>01 July 2017, 42 months</td></tr>
<tr><td colspan="3">Coordinator: Paul Quincey, NPL   Tel: +44(0)208 943 6788   E-mail: paul.quincey@npl.co.uk
Project website address: www.empirblackcarbon.com</td></tr>
<tr><td>Internal Funded Partners:
1 NPL, United Kingdom
2 IL, Finland
3 LNE, France
4 PTB, Germany</td><td>External Funded Partners:
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6 TROPOS, Germany</td><td>Unfunded Partners:
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