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Invited Review Article

Aerosol physical characterization: A review on the current state of aerosol documentary standards and calibration strategies

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ABSTRACT

Aerosols have a wide-ranging impact on the climate, air quality, human health, and agriculture. Despite the ongoing advances in aerosol measurement science and technology, the uncertainties in quantifying aerosol physical properties remain significant in many applications. The accurate characterization of airborne particles - including number and mass concentration, size distribution and light absorption - is critical for understanding their behavior in the atmosphere and environmental fate. We delve into the physical characterization of aerosols, highlighting the measurement and documentary standards that underpin measurement traceability and enable comparison of data collected by instruments based on measurement principles at different times or locations. In particle metrology, recent advances have led to sophisticated primary measurement standards, with relative expanded measurement uncertainties down to 1.1 % (coverage factor $k = 2$; 95 % confidence interval). These standards enable time- and cost-effective instrument calibration to support research, industry, and legislation. We discuss documentary standards and regulations related to air quality and control of particle emissions from vehicles, aviation, shipping, and stationary sources, with the aim to increase awareness of these documents and underline differences in measurement protocols in different sub-fields of aerosol sciences. Importantly, we emphasize the need for further harmonization of measurement procedures, providing specific examples and making suggestions towards this goal. This review, with its comprehensive coverage of aerosol measurement and documentary standards across different sub-disciplines, can serve as a reliable guide for scientists and regulators interested in improving the accuracy of their measurements.

1. Introduction

Atmospheric aerosols can significantly impact climate, human health, and agriculture (Arfin et al., 2023; Davidson, Phalen, & Solomon, 2005; Li et al., 2022; Liu et al., 2021). Aerosol science has therefore received increased attention from state authorities,

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Glossary

AAC	Aerodynamic Aerosol Classifier
AFM	Atomic Force Microscopy
AIST	National Institute of Advanced Industrial Science and Technology (Japan)
APM	Aerosol Particle Mass (Analyzer)
APMP	Asia Pacific Metrology Program
APS	Aerodynamic Particle Sizer
BAM	Beta Attenuation Monitor
BC	Black Carbon
BIPM	Bureau International des Poids et Mesures
CAST	Combustion Aerosol Standard (type of combustion generator)
CCN	Cloud Condensation Nuclei
CCQM	BIPM Consultative Committee for Amount of Substance: Metrology in Chemistry and Biology
CEN	European Committee for Standardization
CERMS	Centrifugal Particle Mass Analyzer – Electrometer Reference Mass Standard
CIPM	International Committee for Weights and Measures
CPC	Condensation Particle Counter
CPMA	Centrifugal Particle Mass Analyzer
DC	Diffusion Charger or Diffusion Charging
DFCAS	Diffusion Flame Combustion Aerosol Source
DLS	Dynamic Light Scattering
DMA	Differential Mobility Analyzer
DPF	Diesel Particle Filter
DUT	Device Under Test
EAB	Electro-gravitational Aerosol Balance
eBC	Equivalent Black Carbon
EI	Emission Index
EC	Elemental Carbon
EM	Electron Microscopy
EU	European Union
FCAE	Faraday-Cup Aerosol Electrometer
GAW	Global Atmosphere Watch Programme of the World Meteorological Organisation (WMO)
GDI	Gasoline Direct Injection
GRPE	Working Party on Pollution and Energy, subsidiary body of the World Forum for Harmonization of Vehicle Regulations (WP.29)
GRV	Global Reference Value
ICAO	International Civil Aviation Organization
ILC/ILS	Interlaboratory comparison/study
IMO	International Maritime Organization
ISO	International Organization for Standardization
LCS	Low-Cost Sensor
LII	Laser-Induced Incandescence
MAC	Mass Absorption Cross-section
MISG	Miniature Inverted Soot Generator
MPSS	Mobility Particle Size Spectrometer (also known as DMAS Differential Mobility Analysing System and SMPS)
MRV	Method-dependent Reference Values
NEDC	New European Driving Cycle
NIST	National Institute of Standards and Technology (U.S.A.)
NMI	National Metrology Institute
NRMM	Non-Road Mobile Machinery
nvPM	Non-volatile Particulate Matter
OC	Organic Carbon
OPC	Optical Particle Counter
OPSS	Optical Particle Size Spectrometer
PAO	Poly-Alpha-Olefine
PEMS	Portable Emission Measurement System
PM	Particulate Matter
PMP	Particle Measurement Programme, working group within UNECE GRPE
PN	Particle Number

PNC	Particle Number Concentration
PTB	Physikalisch-Technische Bundesanstalt (German Metrology Institute)
PTI	Periodic Technical Inspection
PThI	Photothermal interferometry
PS	Polystyrene spheres (used to contain latex and were known as PSL in the past)
rBC	Refractory Black Carbon
QA/QC	Quality Assurance/Quality Control
SAE	SAE International (formerly the Society of Automotive Engineers)
SAXS	Small Angle X-ray Scattering
SEM	Scanning Electron Microscopy
SI	International System of Units
SMPS	Scanning Mobility Particle Sizer (also known as MPSS and DMAS)
SP2	Single-Particle Soot Photometer
SPM	Scanning Probe Microscopy
SPN	Solid-Particle Number
SSA	Single Scattering Albedo
TOA	Thermal-Optical Analysis
TEM	Transmission Electron Microscopy
TEOM	Tapered-Element Oscillating Microbalance
TS	Technical Specification
UFP	Ultrafine Particles
UNECE	United Nations Economic Commission for Europe
US EPA	United States Environmental Protection Agency
UV LIF	Ultraviolet Laser-Induced Fluorescence
VPR	Volatile Particle Remover
WLTP	Worldwide Harmonized Light-Duty Vehicles Test Procedure
XRD	X-ray Diffraction

regulators, and the broader public, especially over the last 20 years, when debates on climate change have become more prominent. The advancement and application of aerosol science, as in many other scientific fields, is driven by experimental measurements, and a vast amount of knowledge regarding aerosol properties has been obtained by this means. Rigorous and accurate measurements can enable aerosol scientists to test new scientific theories, set up reliable computer models and, ultimately, describe the past and future behavior of processes more quantitatively. However, no measurement can be completely free of uncertainties, no matter how carefully planned and executed. With the rapidly growing and maturing market for aerosol instrumentation – with new low-cost PM (particulate matter) sensors for air quality, mid-cost sensors for PN (particle number) emission control, and high-end bioaerosol monitors being introduced to the market every year – the need for reliable aerosol measurement standards and rigorous calibration strategies is becoming increasingly pertinent.

This review aims to inform the scientific community about recent advances in aerosol measurement and documentary standards and calibration strategies and to help experts from different fields (e.g., emission control versus air quality) better understand the differences in the various measurement protocols and the effects these may have on the measurement results. The focus is placed on standards related to the physical characterization of aerosols in the context of particle emission control and air quality monitoring. Methods and protocols for aerosol chemical characterization have been compared and reviewed elsewhere (An et al., 2021; Duarte et al., 2021; Fröhlich-Nowoisky et al., 2018; Gkatzelis et al., 2018; Huffman et al., 2018; Laskin, Laskin, & Nizkorodov, 2012; Palomo-Marín, Pinilla-Gil, Calvo-Blázquez, & Querol-Carceller, 2014; Van Acker, Theiner, Bolea-Fernandez, Vanhaecke, & Koellensperger, 2023; Xu et al., 2020). Documentary standards related to particle filtration would require a dedicated review and are thus also considered beyond the scope of the current document.

The document is structured around measurands (i.e., quantities intended to be measured) – including particle number concentration, particle mass concentration, and particle size – as well as measurement standards specifically targeting black-carbon-related metrics. Efforts were made to review aerosol documentary standards developed in different parts of the world; however, since emission control and air quality monitoring are not as widespread in Africa, Central Asia and Latin America, the discussion in the following sections focuses mainly on standards established in Europe, North America, and parts of Asia. Throughout this review, we highlight opportunities for further harmonization of measurement procedures as well as metrology gaps that need to be addressed through interdisciplinary work.

2. Background

Aerosol metrology pertains to the science of the measurement of aerosols. This has multiple facets, ranging from establishing standards, whether they be documentary standards or reference measurement standards, to verifying aerosol instrumentation.

Calibration is an operation that, under specified conditions, in a first step, establishes a relation between the quantity values with measurement uncertainties provided by measurement standards and corresponding indications with associated measurement

uncertainties and, in a second step, uses this information to establish a relation for obtaining a measurement result from an indication (JCGM, 2012). The term *calibration* thus refers to the act of comparison and does not necessarily include any subsequent software/hardware adjustment of the *device under test* (DUT), which is the instrument or device that is being tested for performance. In practice, very few instruments can be adjusted so that their output/indication matches the value of the applied standard. The calibration standard is usually traceable to a national or international measurement standard held by a metrology body. Measurement uncertainties are either presented as a combined standard uncertainty (used to express the uncertainty of many measurement results at a confidence interval of 68 %, i.e. with a coverage of $k = 1$) or as an expanded uncertainty, i.e. the product of the combined standard measurement uncertainty and a coverage factor larger than the number one. Guidelines for estimating measurement uncertainty in air quality measurements are presented in (ISO 20988, 2007). Details on the accuracy (trueness and precision) of measurement methods and results can be found in (ISO 5725-1, 2023; ISO 5725-2, 2019).

Calibration should not be confused with verification or validation. Verification or validation is simply the provision of objective evidence that the DUT fulfils specified requirements, e.g. that the instrument's counting efficiency (CE) lies within a defined range. The result of the verification/validation procedure is a simple "Pass" or "Fail".

Standards also come in different forms. Laboratory (or measurement or calibration) standards include materials, devices or experimental setups used to perform a calibration. Several types of measurement standards exist, with each intended for different applications and with varying degrees of accuracy. Primary measurement standards are established using a primary reference measurement procedure or created as an artifact that is chosen by convention. A primary reference measurement procedure is a procedure used to obtain a measurement result without relation to a measurement standard for a quantity of the same kind (JCGM, 2012). For instance, a Faraday Cup Aerosol Electrometer (FCAE) can serve as a primary standard for aerosol particle number concentration if calibrated against the primary standards for electric current and aerosol flow. Secondary measurement standards are established through calibration with respect to a primary measurement standard for a quantity of the same kind (JCGM, 2012). For example, a Condensation Particle Counter (CPC) which is calibrated against an FCAE can serve as a secondary standard for particle number concentration. Working measurement standards are routinely used to calibrate or verify measuring instruments or systems, while travelling measurement standards, sometimes of special construction, are intended for transport between different locations (JCGM, 2012). The path of metrological traceability from measurement result to the International System of Units (SI) is illustrated in Fig. 1. This review will mainly focus on primary standards in aerosol sciences, which are often custom-made, state-of-the-art setups developed by National Metrology Institutes (NMIs) to minimize measurement uncertainties.

Interlaboratory comparisons/studies (ILCs/ILSs) contribute to bounding the uncertainties on measurement standards, beyond those that may be captured in a bottom-up uncertainty analysis. These play an important role in establishing the precision and bias (random and systematic uncertainties, respectively) associated with standards. The comparisons can take on different forms, roughly mirroring the traceability pyramid, from formal *key comparisons* organized between NMIs, pilot studies encompassing accredited calibration laboratories, to less formal round robins between non-accredited laboratories. Such comparisons are rapidly increasing in their prevalence in aerosol science, and a number of such comparisons will be highlighted throughout this manuscript.

Documentary standards, by contrast, are technical documents designed to be used as a rule or guideline. They are created by bringing together all interested parties, such as scientists/researchers from academia, industry, and regulators of a particular product or process. Documentary standards can be international, e.g., developed by the International Organization for Standardization (ISO); regional, e.g., created by the European Committee for Standardization (CEN); national; or prepared by a technical standards organization (e.g.

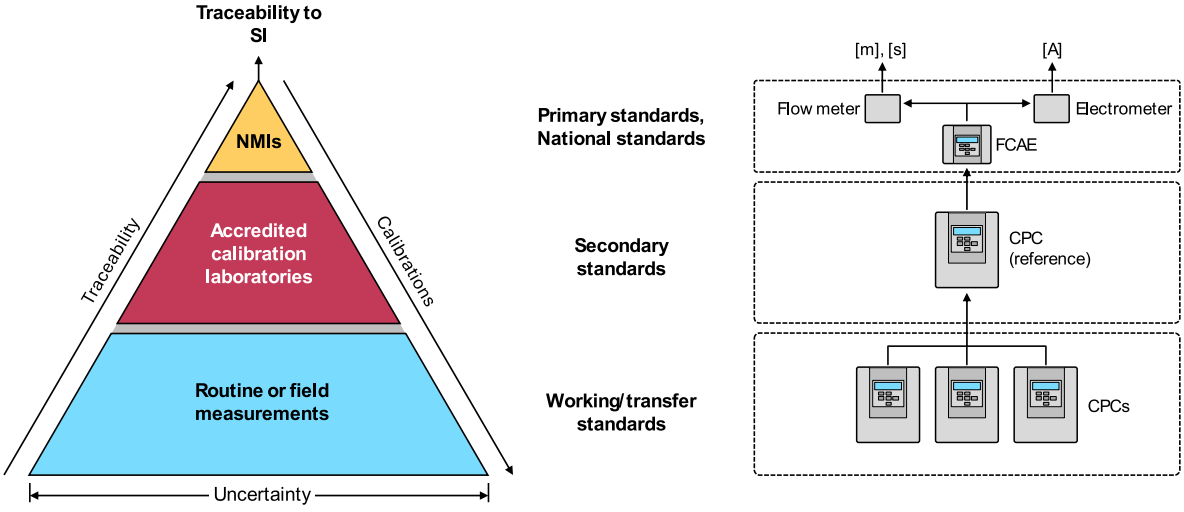

Fig. 1. Left) The *traceability pyramid*, denoting the path of metrological traceability from measurement result to SI units. The measurement uncertainty increases at each step along the calibration chain, being largest at the bottom of the pyramid. (Right) An example of the pyramid for the calibration of condensation particle counters (CPCs) using a Faraday cup aerosol electrometer (FCAE) as a primary standard, where working CPCs are traceable back to a primary standard electrometer and flow meter.

SAE International). Standards differ from acts, regulations, and codes, which have varying levels of legal authority. The majority of documentary standards are adopted on a voluntary basis; however, in certain cases they might be enforced by laws or regulations, often for health and safety reasons. Table 1 provides an overview of major standards organizations having standards related to aerosols.

3. Particle number (PN) concentration

Particle counting is one of the main tenants of aerosol measurement, forming a critical part of many particle sizing systems. There are two primary types of devices used for counting of sub-micrometer particles: condensation particle counters (CPC) and counters based on diffusion charging (DC). CPCs count single particles by detecting pulses of scattered light as the particles pass through a light beam (Cheng, 2011). These counters have a condensation section responsible for growing the particles prior to counting, which enables counting of smaller particles (Cheng, 2011). Multiple configurations exist, with different optical components, working fluids and functionality. The lower size cutoff (D_{50}, i.e. the size at which CE is 50 %) of a CPC depends on the design, typically lying between 3 nm and 23 nm in electrical mobility diameter. CE is the ratio of the number concentration reported by the particle counter under test to the reference number concentration. The electrical mobility diameter is the diameter of a sphere with the same migration velocity in a constant electric field as the particle of interest.

Recently, the use of DC-based instruments has rapidly spread in Europe, especially in the field of vehicle type approval and periodic technical inspection. In a simple DC sensor, unipolar ions are generated in a corona discharge and attach to the aerosol particles. Excess ions are removed by an ion trap, and the charge on the aerosol particles is measured by an electrometer (Dhaniyala, Fierz, Keskinen, & Marjamäki, 2011). Although particle number is not measured directly, it can be evaluated based on the measured charges. More advanced DC sensors have also been reported in the literature (Amanatidis, Maricq, Ntziachristos, & Samaras, 2017; Bainschab, Schriefl, & Bergmann, 2020; Schriefl, Nishida, Knoll, Boies, & Bergmann, 2020). It is estimated that more than 40 000 DC-based instruments have been deployed in Germany, Switzerland, Belgium and the Netherlands alone, and the market is expected to expand further as more European countries are planning to introduce PN-based periodic technical inspection of on-road vehicles.

In the following sections, we will describe the calibration procedures for CPC- and DC-based instruments and discuss applications of PN measurements in the fields of ambient air monitoring as well as vehicle, aircraft and marine emission control. An overview of the regulations and documentary standards related to particle number is presented in Table 2. These will be discussed in more detail in the appropriate sections below.

3.1. Calibration of condensation particle counters

The international standard ISO 27891 (ISO 27891, 2015) describes calibration methods for the determination of the detection efficiency (also known as CE) of CPCs and the associated uncertainty for the particle number concentration range from 1 cm^{-3} to 10^5 cm^{-3} and the particle size range from approximately 5 nm to 1000 nm. In ISO 27891, the detection efficiency is defined as the ratio of the number concentration indicated by the CPC under test (test CPC) to the actual number concentration at the inlet of the test CPC. The latter is determined using a reference instrument while constraining particle size, type, and number concentration. The detection efficiency of a CPC is determined by parallel measurement of particle number concentration of the calibration aerosol consisting of particles of known size and composition between the test CPC and a reference instrument as shown in Fig. 2. The reference instrument can be either a Faraday-cup aerosol electrometer (FCAE) or another CPC calibrated according to ISO 27891(ISO 27891, 2015). Requirements of the reference instrument are specified in the ISO 27891 document. The reference instrument is required to be calibrated with SI traceability by a NMI or an accredited qualified laboratory. A FCAE can be used as reference for the number concentration range above ~10^3 cm^{-3}, and a CPC can be used as reference only in the size range of its plateau region (i.e. the region where CE has reached its maximum value and remains constant). An annex is given for the extension of the number concentration range down to 1 cm^{-3} based on (Owen, Mulholland, & Guthrie, 2012). The calibration aerosol is generated with a stable particle source, conditioned for number concentration and charge, size-classified with a differential mobility analyzer (DMA), which is called differential electrical mobility classifier (DEMC) in ISO 27891, and delivered to the reference and test CPC through a mixing device and flow splitter. Future renditions of the standard may allow for more flow paths from the flow splitter.

There are two major sources of error identified in ISO 27891: (i) the presence of multiply charged particles in the calibration aerosol

Table 1
Major standards organizations or organizations acting as standards producers in some capacity within the realm of aerosol science. Note that those organizations with particle filtration standards are not included as they are out of the scope of this work.

Standards organization	Acronym	Region	Applicable area
International Standards Organization	ISO	International	Broad
European Committee for Standardization	CEN	European Union, European Free Trade Association	Broad
ASTM International	ASTM	International	Broad
SAE International	SAE	International	Aviation/vehicle emissions
International Civil Aviation Organization[a]	ICAO	International	Aviation emissions
Environmental Protectection Agency[a]	EPA	USA	Emissions
United Nations Economic Commission for Europe[a]	UNECE	International, with focus in Europe	Vehicle emissions

[a] Some organizations are unique in that they operate in some capacity to create standards while also being regulatory bodies.

Table 2
Regulations and documentary standards related to particle number concentration (PNC).

Document number	Originating organization	Originating region	Application	Technology/Instrumentation	Calibration/test aerosol
ISO 27891	ISO	International	All applications	CPC, calibration of	Various, e.g. Emery Oil, combustion particles, silver particles
CEN/TS 16976	CEN	Europe	Air quality	CPC, calibration of	Silver
(UNECE R83, 2006) (light-duty vehicles, NEDC) (UNECE R154, 2021) (WLTP) (UNECE R49, 2021) (heavy-duty vehicles) (UNECE R96, 2019) (NRMM)	UNECE	Europe	Vehicle type approval	VPR and engine exhaust CPC, technical requirements and calibration of	Emery Oil and combustion particles (Giechaskiel et al., 2021)
(UNECE R168, 2024);	European Commission	Europe	Real-driving emission testing	PN-PEMS, technical requirements and calibration of	Monodisperse and thermally stable soot-like aerosols (combustion or spark discharge carbonaceous particles) (Giechaskiel, 2018)
EU 2023/688 [a]	European Commission	Europe	Periodic technical inspection of diesel vehicles	PN-PTI, technical requirements and calibration of	Various (depending on national regulations), e.g. combustion, spark discharge carbonaceous particles, NaCl particles
ICAO Annex 16, Volume II (regulatory requirement) and SAE ARP6320B (technical standard)	ICAO and SAE	International	Aircraft engine emissions certification	Non-volatile PM number: VPR and CPC, performance specifications for instrumentation and calibration of	Non-volatile PM number: VPR - 30 nm tetracontane >95% purity; CPC – Emery Oil or equivalent

[a] UNECE's World Forum for Harmonization of Vehicle Regulations (WP.29) has also adopted an amendment to the United Nations Rule No.1 of the 1997 Agreement that allows for the introduction of a more robust test procedure to measure exhaust particle emissions during periodic technical inspection tests for all diesel light-duty vehicles (passenger cars and vans) equipped with diesel particulate filters (UNECE WP.29, 2024).

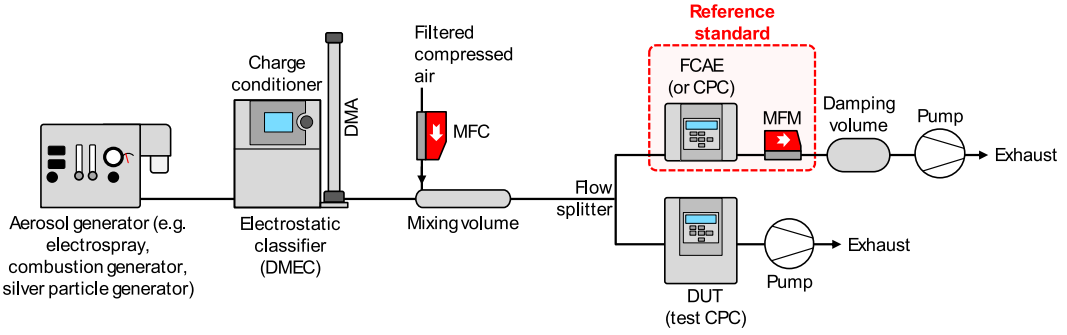

Fig. 2. Simplified schematic of the CPC calibration setup described in ISO 27891.

after the DMA, and (ii) differences in the particle number concentration of the calibration aerosol between the test CPC and the reference instrument due to the flow splitter. The presence of multiply charged particles would cause a bias in two ways: (i) the detection efficiency is underestimated when both a FCAE is used as reference and when all of the particles are assumed to be singly charged in the derivation of number concentration from the FCAE measurements, and (ii) the detection efficiency is overestimated in the cut-off size range because the multiply charged particles are larger than singly-charged particles and are detected with higher efficiencies by the test CPC. To reduce the bias due to multiply charged particles, ISO 27891 provides methods for the evaluation of the fractions of multiply charged particles in the calibration aerosols by measurement and for the correction of the detection efficiency with the obtained fractions. ISO 27891 also requires that the overall multiply-charged number fraction be less than 0.1. For the number concentration bias in the flow splitting, ISO 27891 describes a method to evaluate the bias correction factor by measurement (ISO 27891, 2015). The method works when the inlet flow rates are equal between the test CPC and reference instrument and does not require a knowledge of the detection efficiency of either instrument. The uncertainty evaluation in ISO 27891 then consists of five factors: uncertainty of the reference instrument, uncertainty of the multiple charge correction (not needed when a CPC is used as reference), uncertainty of the splitter bias correction factor, deviation of the flow rate of the reference instrument, and the repeatability of the calibration measurement of the detection efficiency. The detection efficiency thus obtained establishes traceability to the SI through particle charge concentration, as explained in traceability diagrams given in ISO 27891 (ISO 27891, 2015).

To establish metrological traceability to SI units for the measurement of particle number concentration with CPCs by calibration, NMIs hold the responsibility to develop and maintain national measurement standards for particle number concentration. To check the equivalence of the measurement standards among the NMIs, they participate in international comparison studies (Yli-Ojanperä et al., 2012). For example, a comparison study CCQM-K150 (Brown et al., 2023) was conducted with participation by nine NMIs and designated institutes (DIs) to compare the measurement results of particle number concentration by CPCs as well as particle charge concentration by FCAEs. For the comparison study, the NMIs brought CPCs and/or FCAEs, which were their primary or secondary instruments, to the pilot laboratory (TROPOS, Germany) and measured number/charge concentrations of particles distributed from the same source through a manifold. The comparison was performed with particles of 40 nm and 50 nm in electrical mobility diameter at number concentrations in the range from 100 cm^{-3} to 20 000 cm^{-3} for the comparison with CPCs, and at charge concentrations in the range from 0.15 fC cm^{-3} to 3 fC cm^{-3} for FCAEs, which approximately corresponds to the number concentration range from 1000 cm^{-3} to 20 000 cm^{-3} for singly charged particles. Based on the results of such comparisons, NMIs claim their calibration and measurement capabilities (CMCs) to be recognized internationally in the key comparison database (KCDB) of the mutual recognition arrangement (MRA) by the International Committee for Weights and Measures (CIPM). Through the CCQM-K150 study and other past international comparisons (Quincey et al., 2014; Schlatter, 2009), the equivalence of national measurement standards at number concentrations in the range from 100 cm^{-3} to 20 000 cm^{-3} has been established, with expanded uncertainties with a coverage factor of 2 (95 % confidence interval) of as low as 1.1 % for one NMI (Sakurai et al., 2023).

To investigate the equivalence of national measurement standards at lower number concentrations, the NMIs in Switzerland and Japan conducted a study to compare their measurement results at number concentrations as low as 1 cm^{-3} through the calibration of the detection efficiency of a transfer CPC (Sakurai et al., 2023). The two countries established national measurement standards for number concentration that do not rely on direct comparison of CPC with FCAE in the concentration range below 10^3 cm^{-3}. The Swiss standard is based on a custom-made reference Optical Particle Counter (OPC) (for more details see Subsection 5.2.3) whereas the Japanese standard relies on a reference FCAE and aerosol diluters. The obtained results were in good agreement within the stated uncertainties (4 %–5 % for each NMI at the number concentration level of about 1 cm^{-3}), which provided evidence for the validity of their individual measurement procedures and the equivalence of the two national measurement standards.

Note that ISO 27891 does not prescribe a specific test aerosol for CPC calibration and that calibration certificates are only valid for the specific particle type used as calibration aerosol. In practice, different research and standardization groups work with several different test aerosols, which often leads to misunderstandings and hinders data comparison. In the sections below, we will provide a comprehensive overview of the test aerosols used in vehicle and aircraft emission control as well as in air quality monitoring.

3.1.1. Particle counters for on-road and non-road vehicle type-approval and periodic technical inspection

In Europe, emission limits based on solid PN (SPN) concentration were first introduced for light-duty vehicles in 2011, followed by heavy-duty vehicles in 2013, and non-road mobile machinery in 2019 (European Parliament, 2008a, 2011, 2012; Giechaskiel, Melas, Martini, & Dilara, 2021). The standards for the measurement of PNC emitted by vehicles or internal combustion engines are described in the UNECE Regulations No. 83 (light-duty vehicles New European Driving Cycle NEDC), No. 153 (World Harmonized Light Vehicles Test Procedure WLTP), No.49 (heavy-duty vehicles) and No. 96 (non-road mobile machinery NRMM). A standardized PN measurement protocol for the automotive industry was developed by the Particle Measurement Programme (PMP), a working group within UNECE GRPE (Working Party on Pollution and Energy of the United Nations Economic Commission for Europe). PMP-compliant measuring systems consist of a volatile particle remover (VPR) and a particle number counter, typically a so-called engine exhaust CPC, i.e., a CPC with a CE of 0.50 ± 0.12 at a mobility diameter of D_{50} = 23 nm and a CE above 0.90 at 41 nm (Giechaskiel et al., 2009). Once the new Regulation Euro 7/VII comes into force, the specifications are expected to become more stringent (CE of 0.65 ± 0.15 and >0.90 at 10 nm and 15 nm, respectively). Note that these are laboratory-based systems (as opposed to portable systems described below), which sample from a dilution tunnel, and that the VPR and CPC are calibrated separately, not as unit. It must also be highlighted that the particle losses between vehicle and particle number system (i.e. connecting tube up to 6 m and dilution tunnel), which are typically up to 15 %, are not taken into account (Giechaskiel et al., 2021). The main calibration requirements are summarized in (Giechaskiel et al., 2022). Here, we will focus on the calibration of the CPC; information on technical specifications and calibration of VPR can be found elsewhere (Giechaskiel et al., 2022; Giechaskiel, Melas, et al., 2021).

Although the test aerosol is not strictly defined in the legislation (emery oil or any soot-like materials are allowed), engine exhaust CPCs are commonly calibrated according to the ISO 27891 standard using Emery Oil (Poly-Alpha-Olefin, PAO 4) or soot particles generated by a Combustion Aerosol Standard (CAST) generator (Giechaskiel et al., 2011). It is known, however, that the lower cut-off curve of a CPC strongly depends on the properties (i.e., chemistry and morphology) of the test aerosol. It has been shown that the CE is ~0.15 and ~0.06 units higher for Emery Oil than for CAST combustion particles at mobility diameters of 23 nm and 41 nm, respectively (Giechaskiel et al., 2011; Giechaskiel et al., 2022; Terres et al., 2018; Wang, Caldow, Sem, Hama, & Sakurai, 2010). Though Emery Oil particles bear no resemblance in terms of chemistry or morphology to combustion particles emitted by vehicle engines, CPC calibration with Emery Oil has become standard practice in the automotive industry, and it is used by many instrument manufacturers for in-house CPC calibration.

In addition to the laboratory measurements, on-road measurements with portable emissions measurement systems (PEMS) are required in the EU since 2017 ((EU) 2017/1154, 2017; Giechaskiel, Bonnel, Perujo, & Dilara, 2019). PEMS are smaller and lighter than their laboratory counterparts and sample directly from the tailpipe. They are comparable with the reference laboratory-based (i.e., PMP-compliant) systems in terms of performance, with measurement deviations typically within 40 % (Giechaskiel, Bonnel, et al., 2019; Giechaskiel et al., 2020; Riccobono, Giechaskiel, & Mendoza Villafuerte, 2016). PN-PEMS (also known as SPN-PEMS to emphasize that only solid particles are measured) consist of an optional sampling line and a PN analyzer, which combines a dilution

and thermal pre-conditioning unit with a particle detector, typically a CPC- or DC-based particle counter. Current technical requirements prescribe a 23 nm lower cut-off size, but in view of the upcoming regulations, PEMS with improved detection efficiency at 10 nm are currently being developed (Giechaskiel et al., 2020; Giechaskiel, Mamakos, Woodburn, Szczotka, & Bielaczyc, 2019; Zinola et al., 2019).

Technical requirements (e.g., size-dependent CE, linearity as a function of PN concentration (PNC), volatile removal efficiency) are specified for the PN analyzer as a whole unit in Regulation ((EU) 2017/1154, 2017). The CE of PN-PEMS is determined with monodisperse and thermally stable soot-like aerosols, such as combustion particles by CAST-based burners or carbonaceous particles by spark discharge generators (Giechaskiel, Melas, et al., 2021; Riccobono et al., 2016). The most straightforward way is to determine the CE of the complete PN-PEMS, but a separate calibration of the thermal pre-conditioning unit and the particle detector (CPC- or CD-based) is also permissible. In the second case, the two calibration results have to be combined to yield the total penetration efficiency of the PN-PEMS (Giechaskiel, 2018). A schematic illustration of the calibration setup can be found in (Giechaskiel, 2018). Note that a second aerosol neutralizer must be placed after particle size-selection by the DMA in the case of DC-based counters since the pre-existing charges of the test aerosol will have a strong effect on the sensor's CE (Qi, Asbach, Shin, Fissan, & Pui, 2009). As a result, DC-based counters cannot be calibrated directly against an FCAE, but only against a reference CPC, i.e. a CPC which has been calibrated against an FCAE according to the ISO 27891 standard. PEMS ILCs at a non-NMI level have shown a variability of $\pm 1 \times 10^{11}$ particles per km for PN (Simone Casadei et al., 2021).

Experiments at the Swiss Federal Institute of Metrology (METAS, unpublished data) indicate that certain DC-based counters integrated in PN-PEMS exhibit a higher CE with spark-discharge particles compared to combustion particles of the same mobility diameter by about (0.3 to 0.4) units. This is in agreement with a recent study that observed differences in CE of about 0.5 units (Melas, Trikka, Valentini, Cotogno, & Giechaskiel, 2024). Such deviations in CE are 2 to 3 times larger than the expanded measurement uncertainty in the PN-PEMS calibration against a reference CPC or electrometer (Kazemimanesh et al.). Any effort to further reduce the uncertainties in the PN-PEMS calibration or improve metrological traceability will be in vain unless a suitable proxy for diesel soot is selected by the scientific community and clearly specified in the regulation.

To ensure that vehicles in operation undergo proper service, so that they can maintain their performance as guaranteed by type-approval throughout their lifetime, several European countries recently introduced new periodic technical inspection (PTI) procedures based on PNC measurements. At present, PN-PTI is limited to diesel vehicles equipped with a Diesel Particle Filter (DPF), aiming to identify defects or tampering thereof. The technical specifications of the PN-PTI instruments and relevant calibration procedures are defined at a national level (Melas, Selleri, Suarez-Bertoa, & Giechaskiel, 2022; Vasilatou, Kok, et al., 2022). While those methods are similar, they do differ in certain aspects, e.g. in the required CE of the particle counters or the test aerosol used for PN-PTI instrument calibration and annual verification. To avoid new measurement methods (other than the existing ones) being developed and established in the EU in the future, a common set of minimum requirements for PNC measurement has been recommended by the European Commission ((EU) 2023/688, 2023). A schematic representation of the experimental setup for the calibration/verification of PN-PTI instruments is shown in Fig. 3. Similar to PN-PEMS, PN-PTI instruments consist of a dilution and thermal pre-conditioning unit with a particle detector, typically a CPC- or DC-based particle counter (Melas, Vasilatou, Suarez-Bertoa, & Giechaskiel, 2023). However, as PN-PTI instruments need to be more compact and affordable, the design and performance of the PTI counters might not necessarily meet the same standards as those defined for PEMS. Recent studies on the performance of DC-based PN-PTI sensors revealed that PTI counters are highly sensitive to the properties of the test aerosols (Hammer, Roos, Giechaskiel, Melas, & Vasilatou, 2024; Vasilatou et al., 2023). More specifically, the use of sodium chloride (NaCl) and graphitic particles from spark discharge generators led to a change in CE by up to a factor of 2 compared to combustion particles. The choice of aerosol generator (e.g. nebulizer model/design) also played a role. With EU countries, such as the Netherlands, Belgium and Germany, allowing the use of these materials as test aerosols for the annual verification of PN-PTI instruments, it is important to define suitable correction factors that account for the effects of the test aerosol on the CE of the instruments (Vasilatou et al., 2023); otherwise, there is a risk that instruments placed on the European market might fail to reliably detect malfunctioning DPFs. It must be highlighted that such correction factors will not be universal; they need to be defined for every instrument type and every calibration setup separately.

The methodology for measuring solid PNC of vehicle emissions, initially introduced in the EU in 2011, has since then been adopted by several countries, particularly in Asia. For example, China (China 5 for light-duty diesel vehicles, China 6/VI all vehicles), India (BS VI all vehicles since 2020), and Korea have introduced solid PNC limits both under laboratory test conditions and real-driving conditions (Deo et al., 2023; Giechaskiel, Melas, et al., 2021; Park, Shin, Lee, & Lee, 2021). Discussions on PTI based on PNC are ongoing in many European countries, while the first PN-PTI measurements have been performed in Mexico City, Bogota, Lima and Santiago de Chile (Botero, Londoño, Agudelo, & Agudelo, 2023; CALAC+). Considering the wide application of CPCs and PN-PEMS worldwide and the large number of PN-PTI instruments already on the European market (>40 000 units), further harmonization of the calibration procedures (e.g., choice of test aerosol) would be desirable to ensure that data from different parts of the world are comparable and that meaningful long-term trends can be drawn.

3.2. *Condensation particle counters for air quality monitoring in Europe*

Although PNC in ambient air is not a regulated metric, several air quality monitoring stations or atmospheric observatories measure PNC of sub-micrometer particles because of their impact on human health and the environment. Ultrafine particles (UFP), i.e. particles with a diameter smaller than 100 nm, are considered as an air pollutant of emerging concern according to a recent report by the EU Joint Research Center and might be regulated in the near future (Monforti-Ferrario et al., 2022).

Number concentration of sub-micrometer particles in ambient air is measured according to the technical specification CEN/TS

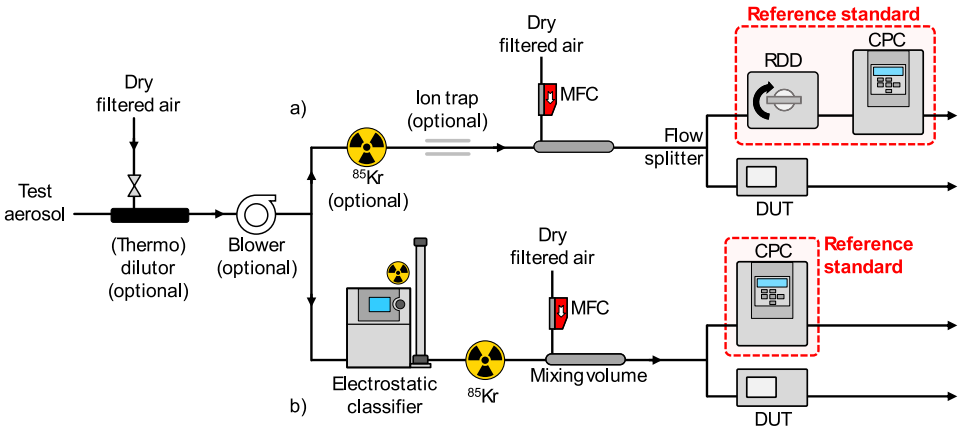

Fig. 3. Example of an experimental setup for the calibration/verification of PN-PTI instruments against a reference CPC, using a) polydisperse and b) monodisperse aerosols. RDD, CPC and DUT stand for rotating disc dilutor, condensation particle counter and device under test respectively. Figure adapted from (Vasilatou et al., 2023).

16976 (CEN/TS 16976, 2016). This document describes a standard method for PNC measurement based on a CPC operated in the counting mode and a dilution system for concentrations exceeding the counting mode range. CPCs for ambient monitoring are required to have a D_{50} and D_{90} CE at 7 nm ± 0.7 nm and <14 nm, respectively. The instruments are calibrated against an FCAE (or a traceable CPC) according to ISO 27891 using silver particles produced by the evaporation/condensation method as test aerosol. Silver particles with a mobility diameter smaller than about 15 nm are quasi spherical, while larger particles have a fractal-like morphology (Hammer et al., 2022; Scheibel et al., 1983; Tuch et al., 2016). If necessary, the latter can be sintered by passing through one or multiple sintering stages. The main advantages of using silver aerosols are the generation of high particle number concentrations even at particle sizes below 20 nm and the narrow size distribution which makes the aerosols quasi monodisperse (Hammer et al., 2022; Tuch et al., 2016). Moreover, silver particles result in a similar cut-off curve of the CE as soot particles by CAST-based generators (Giechaskiel and Melas, 2022; Kiwull, Wolf, & Niessner, 2015; Terres et al., 2018; Wang et al., 2010).

3.3. *Condensation particle counters for aircraft engine emissions*

Similar to the requirements for particle counters for vehicle type-approval, global aircraft engine certification requirements have set emission limits based on non-volatile particulate matter (nvPM) number emission indices (EI), that is the number of particles per kg of fuel consumed (particles kg_{fuel}^{-1}), with mathematical definitions provided in (Lobo et al., 2015). nvPM is defined as emitted particles that exist at the gas turbine engine exhaust nozzle exit plane that do not volatilize when heated to 623 K, which is functionally equivalent to the solid particles counted for vehicle emissions (see also Sec. 5). The EI represents the number of nvPM particles emitted per kg of fuel burned by the engine. The limits for nvPM number, introduced as standards for certification of in-production and new type aircraft turbofan/turbojet engines with rated thrust >26.7 kN (ICAO, 2023), were adopted by the International Civil Aviation Organization (ICAO) Council in 2020 and have been enacted in national legislation by almost all ICAO member engine-producing countries as of 2024. The technical requirements for a standardized system to sample and measure nvPM mass and number concentrations for aircraft gas turbine engines (SAE ARP6320B, 2024) were developed by the SAE E−31 Aircraft Engine Gas and Particulate Emissions Measurement committee in response to a remit from ICAO. For nvPM number, the method built upon knowledge developed by PMP, with a standardized sampling approach and a measuring system consisting of a volatile particle remover (VPR) and a CPC with a CE of ≥50 % (D_{50}) at a mobility diameter of 10 nm and ≥90 % at 15 nm (Lobo et al., 2015). The specification for aviation is for smaller size cutpoints (D_{50} and D_{90}) than that for vehicles, due to small size of particles emitted by modern gas turbine engines, often with GMDs in the range of 15 nm to 50 nm (Saffaripour, Thomson, Smallwood, & Lobo, 2020), acknowledging that vehicle standards are also moving to more stringent size cutpoints.

Calibration of the nvPM number measurement system is performed in two parts (ICAO, 2023; SAE ARP6320B, 2024). First, the VPR must demonstrate that it has >99.5 % removal of 30 nm tetracontane ($CH_3(CH2)_{38}CH_3$, >95 % purity, particles with an inlet concentration ≥10 000 cm^{-3}). The calibration to demonstrate this performance must be conducted annually. Secondly, the CPC must be calibrated annually with Emery Oil (or equivalent), with a counting accuracy against a traceable standard of ±10 % from 2000 cm^{-3} (or expected lower threshold) to the upper threshold of the single particle count mode.

Due to the size of the largest gas turbine engines and the noise and vibration levels in the engine certification test cells, the instruments must be located at a distance where they are unaffected by the engine operation. Further, the sample is collected in the engine exhaust at high temperatures and high velocity, such that dilution is required to avoid condensation of water vapor and coagulation of nvPM particles. The sampling system, which has been standardized as much as possible regardless of engine size, results in a dilution factor of ~10 and a sampling line length of ≤35 m from the sampling probe tip to the entrance of the measurement instruments. An ILC of three different reference sampling and measurement systems conforming to the SAE and ICAO specifications was conducted and found that the reference systems generally agreed to within 15 % of the average nvPM number emission index

(Lobo et al., 2020).

The long sampling lines result in high levels of nvPM particles being lost between the sampling probe tip and the instruments, due primarily to thermophoresis and diffusion. As a result, to aid in estimating the engine-out emissions, a method has been developed based on the measured number and mass concentrations and established theoretical losses to estimate the system loss correction factor. This correction is to be applied to the nvPM number EI in order to predict the engine exit plane emissions, with further details provided in SAE ARP6481 (SAE ARP6481, 2019). While not a certification requirement, there is a reporting requirement (ICAO, 2023) to provide this correction factor for engine nvPM number emissions so that the health, climate, and local air quality impacts due to nvPM emissions may be assessed. The correction factors for nvPM number have values in the range from 2 to 10 (Durand et al., 2023). Improved correction methods, using measured particle size, are under development (Durand et al., 2023) and are being assessed for future regulation.

3.4. Particle number measurements for marine emissions

The International Marine Organization (IMO), the body governing particulate emission internationally, does not currently require particle number measurement. To detect defective diesel particle filters (DPF), Switzerland has recently introduced a new procedure for the technical inspection of ship emissions based on PNC (VASm, 2020). Inspection is carried out with the instruments approved for PTI of diesel vehicles (see Subsection 3.1.2). National regulations define a limit value of 250 000 cm^{-3} (AB-VASm, 2022). If this limit is exceeded, the particle filter must be replaced by a new one.

3.5. Summary of test aerosols used for particle counter calibration

Table 3 provides a summary of the test aerosols used for calibration of different particle counters employed for emission control and air quality monitoring. The test aerosols include combustion particles produced by a CAST generator (Ess, Bertò, Irwin, et al., 2021; Ess et al., 2018; Terres et al., 2018; Vasilatou et al., 2023; Wang et al., 2010) or a miniature inverted soot generator (Bischof, Weber, Bundke, Petzold, & Kiendler-Scharr, 2020; Hammer et al., 2024; Kazemimanesh et al., 2018; Moallemi et al., 2019; Senaratne et al., 2023), carbonaceous particles by spark discharge generators (Burtscher, 2019; Mamakos, Giechaskiel, & Drossinos, 2013; Terres et al., 2018), Emery Oil (Giechaskiel et al., 2011; Mamakos et al., 2013; Terres et al., 2018; Wang et al., 2010), silver particles (Hammer et al., 2022; Hermann et al., 2007; Terres et al., 2018; Tuch et al., 2016; Wang et al., 2010), NaCl (Hermann et al., 2007; Vasilatou et al., 2023; Wang et al., 2010) and NaI particles (Krasa, Kupper, Schriefl, & Bergmann, 2023). Tetracontane is used to evaluate the volatile particle removal efficiency of VPRs (Giechaskiel, Melas, et al., 2021), a pretreatment stage in vehicle and aviation PN measurement.

As the CE of both CPC- and DC-based counters depends on the properties of test aerosols, there is a strong need for harmonization in the calibration of particle counters. With the European Commission considering to extend the Air Quality Directive to include number concentration of UFPs, this need will become even more pressing. Lack of harmonization in the performance of the particle counters used for emission control and air quality monitoring will, as an example, hinder efforts to link the impact of transport-related UFP sources on air quality in terms of ambient UFP number concentration levels.

CPCs with a D_{50} cutpoint at 10 nm are already specified in aviation, and are being contemplated for specification to control other engine emissions (Samaras et al., 2022). From 2025 onwards, once the Euro 7 regulation for on-road vehicles comes into force, limit values for the emission of solid particles with a mobility diameter starting from 10 nm (PN_{10}[1]), instead of 23 nm as in Euro 6, will be introduced. This will drive the need for stable aerosol calibration sources that generate smaller particle sizes, as commercial combustion aerosol sources produce limited or no solid particles in this size range.

Flame spray pyrolysis (FSP) is an emerging approach, which may prove capable of expanding the size range and types of particles available for calibration purposes. The technique has been shown to produce combustion particles with high elemental carbon (EC) content and small sizes consistent with aviation emissions (Kholghy et al., 2021; Scott et al., 2024; Trivanovic, Kelesidis, & Pratsinis, 2022).

4. Particle mass concentration

4.1. Measurement methods

Numerous studies have shown a correlation between PM mass concentration and the onset of many diseases, such as respiratory, cardiovascular, and cancer (WHO, 2021). Ambient PM mass concentration has therefore been regulated in many parts of world, while in North America and elsewhere this metric is also used for vehicle emission control. Non-volatile (nv) PM mass concentration measurements are also carried out alongside PN concentration measurements in the field of aviation. Multiple measurement methods are available to measure PM mass concentration. For decades the most common measurement was gravimetric, referring to weighing filters before and after being exposed to an aerosol stream with a known flow rate for a certain amount of time (US EPA, 1971–2024). In the last (25 to 30) years, several semi-continuous instruments for near-real time monitoring of PM mass concentrations, such as the Tapered-Element Oscillating Microbalance (TEOM) and the Beta Attenuation Monitor (BAM), have also been used to measure total PM

[1] Note that the terminology around PN size fractions is not harmonized. For example, CPCs with a cutoff at 23 nm and 10 nm are often referred to as CPC_{10} and CPC_{23}, respectively. However, a CPC with a 10 nm cutoff could also be designated as $PN_{0.01}$.

Table 3
List of test aerosols commonly used in the calibration of CPC- and DC-based particle counters, relevant physicochemical properties of the particles and application range.

Generator	Typical GMD_{mob} (nm)	Typical GSD	Chemical composition	Morphology	Advantages	Disadvantages (compared to engine soot)	Application range
miniCAST or CAST	20 to 200	1.6	Combustion particles, EC and OC mass fractions depend on operation point	Fractal-like to spherical particles	Particles simulate satisfactorily the physicochemical properties of vehicle engine soot	Wide range of operating points allow for particles that are not fractal, especially at smallest sizes	Any type of CPC, PN-PEMS, PN-PTI (for both type-examination and verification)
Miniature Inverted Soot Generator (MISG)	95 to 200	1.9 to 2.2	Combustion particles, mostly EC	Fractal-like	Affordability, particles simulate satisfactorily the physicochemical properties of vehicle engine soot	$GMD_{mob} \geq$ 90 nm	Possibly for PN-PTI (instrument verification only)
Spark discharge	5 to 150	1.7 to 1.9	Carbonaceous, mostly EC	Fractal-like	Affordability	Smaller primary particle size, lower particle effective density, high particle charges	PN-PEMS, PN-PTI (instrument verification only[a])
Silver	5 to 70	1.1 to 1.4	Silver (pure or oxidized depending on carrier gas)	Fractal-like (compact if sintered and at diameters below 10 nm)	High number concentration of particles below 20 nm, CPC cut-off curve similar to that obtained with combustion particles	Does not simulate the chemical properties of engine soot, different aerodynamic properties	CPCs for ambient monitoring [b]
NaCl (through nebulization)	20 to 100	1.6 to 2.1 (depending on nebulizer)	NaCl	Cubic crystals	Affordability	Does not simulate the physicochemical properties of engine soot	PN-PTI (verification only[a])
NaI (through nebulization)	20 to 100	1.6 to 2.1 (depending on nebulizer)	NaI	Cubic crystals	Affordability, CPCs exhibit constant CE for NaI particles above ~15 nm (i.e. no need for particle size selection)	Does not simulate the physicochemical properties of engine soot	Field testing of engine exhaust CPCs
Emery Oil[c]	20 to 60	1.1	PAO 4 cSt	Spherical droplets	Almost no multiply charged particles if nebulized with electrospray generator	Does not simulate the physicochemical properties of engine soot, volatile material	Engine exhaust, including aviation, CPCs
Tetracontane	15 to 50	1.4	$CH_3(CH_2)_{38}CH_3$	Spherical droplets	Completely evaporates at VPR temperatures	Only challenges the removal efficiency of organic material	Volatile removal efficiency of VPR (vehicle and aviation)

[a] Correction factors must be applied to the CE of DC-based PN-PTI counters (Vasilatou et al., 2023).
[b] PMP is currently discussing the possibility of recommending silver particles (along with Emery Oil and CAST-based soot) for calibration of engine exhaust CPCs.
[c] Historically, the use of Emery Oil was based on its resemblance to the major constituent of volatile particles that, at the time when the PMP method was initially developed, was lubricant oil.

mass or, in conjunction with an impactor or cyclone, to determine different PM mass fractions (Weingartner, Burtscher, Hüglin, & Ehara, 2011). As such, air quality and aerosol research observation stations are increasingly turning to real-time optical instruments (sometimes also referred to as dust monitors or aerosol spectrometers) because of their affordability, compact size, and easy maintenance (Amacker et al., 2023; Ardon-Dryer, Kelley, Xueting, & Dryer, 2022). This section will focus predominantly on the manual reference gravimetric method, measurement methods benchmarked against this reference, and the calibration of mid- and low-cost optical PM monitors/sensors.

A list of documentary standards related to the mass of particulate (PM mass) is provided in Table 4. These standards will be discussed in detail in the following sections.

4.2. Air quality monitoring of $PM_{2.5}$ and PM_{10} mass concentrations

Air quality standards (here referring to allowable concentrations) date back to the Environmental Protection Agency (EPA) in the early 1970s (US EPA, 1971–2024) and vary between regions (Nazarenko, Pal, & Ariya, 2021). Procedures for measuring air quality have been reviewed a number of times over the years (Cao, Chow, Lee, & Watson, 2013; Chow, 1995; McMurry, 2000; Patel et al., 2022), typically with a focus on atmospheric sampling. In all cases, air quality requires measurements of airborne particulate matter (PM) and some of its constituents. One of the most important regulated metrics for airborne particle monitoring is the mass concentration, or more specifically the total mass per unit volume of air of particles which are small enough to pass through a size-selective inlet. Common demarcations include total suspended particles (TSP), covering all particle sizes that can stay suspended; PM_{10} (coarse mode); and $PM_{2.5}$ (fine mode), which have 50 % efficiency cut-offs at 100 μm, 10 μm, and 2.5 μm aerodynamic diameters, respectively. Note that there is overlap between these quantities, e.g., all of $PM_{2.5}$ is contained in PM_{10}. A final size class, ultrafine particles (UFP), refers to much smaller particles with a mobility or geometric diameter <100 nm, which are measured using counting instruments but are not widely monitored. These size classes are shown relative to representative atmospheric aerosol size distributions in Fig. 4. Gravimetric measurements are universally used as the standard reference method, with different allowances for other methods of measurement depending on the region. Inlets used in size-selection also vary between regions, with most modern air quality standards taking a dichotomous approach by requiring reporting of both PM_{10} and $PM_{2.5}$.

In the EU, air quality monitoring – as laid down in the Air Quality Directive 2008/50/EC (European Commission, 2022; European Parliament, 2008b) – is mandatory and requires quantification of airborne particulate matter (PM) and some of its constituents. Reference measurements are performed with the manual gravimetric method, a filter-based method described in detail in the standard EN 12341:2014 (CEN/TC 264, 2014). Note that the inlet design for the sampling of PM_{10} and $PM_{2.5}$ is standardized in Europe and that the sampling flow must be kept at 2.3 $m^3\ h^{-1}$. It is worth noting that the measurement uncertainties for PM mass concentration in the directive are 25 %, and thus much higher than those for gaseous pollutants (typically 15 %). As previously mentioned, the main drawback of the manual reference gravimetric measurement is the poor time resolution (filter measurements are made once every 24 h). In recent years, automated PM instruments providing high-time resolution (or even real-time measurements) are increasingly being used at monitoring stations for regulatory purposes. These monitors must be tested for equivalence with the manual gravimetric reference method in monitoring sites using real ambient air (EC Working Group, 2010), which requires time-consuming and expensive testing field campaigns in an attempt to include all representative meteorological conditions and the temporal and spatial variations of the ambient air composition.

Mid-cost monitors (typically based on optical measurements) – used in industrial, workplace, or indoor applications – do not necessarily need to be tested for equivalence with the reference gravimetric method and are usually calibrated with simple aerosols by the manufacturer. Likewise, low-cost PM sensors are either tested through collocation studies with a reference station (see (Karagulian et al., 2019) and references therein) or calibrated in the laboratory with simple model aerosols, such as dust or salt particles (Hogrefe, Drewnick, Lala, Schwab, & Demerjian, 2004; Liu, Zhang, Jiang, & Chen, 2017; Omidvarborna, Kumar, & Tiwari, 2020; Papapostolou, Zhang, Feenstra, & Polidori, 2017; Schwab, Hogrefe, Demerjian, & Ambs, 2004) or dried organic particles, e.g. sucrose and adipic acid (Zhang, Marto, & Schwab, 2018). Currently, there exist no documentary standard on the calibration of mid- and low-cost PM sensors in Europe (or worldwide), but CEN/TC 264/WG 42 is about to amend CEN/TS 17660–1:2021 on gas sensors with a second technical specification fully dedicated to PM sensors (CEN/TS 17660-1, 2021).

Optical PM sensors based on light scattering measure optical diameter and particle number concentration, which is then converted to mass concentration by assuming either a constant or a size-dependent particle effective density (Wu, Horender, Tancev, & Vasilatou, 2022). Light scattering is strongly affected by particle hygroscopicity, refractive index, and particle composition, with all of these factors exhibiting high seasonal and spatial variability. A single calibration with a simple model aerosol in the laboratory or a collocation study at one reference station will introduce significant biases when the sensor is installed at a different site. Multiple collocation studies or a rigorous laboratory-based calibration with tailored aerosols, i.e. aerosols representative of the environment of their intended use, are needed (Jayaratne et al., 2020; McNamara, Noonan, & Ward, 2011). Efforts are currently being directed towards standardizing laboratory-based calibration procedures for automatic PM measuring instruments under well-controlled and reproducible experimental conditions. Multi-component model aerosols containing fresh and aged soot particles, inorganic salt, and mineral dust particles are generated in order to reproduce the main properties of real ambient air in terms of particle size distribution, chemical composition, and number/mass concentration, including volatility and hygroscopicity as shown in Fig. 5 (Horender, Auderset, et al., 2021; Horender, Tancev, Auderset, & Vasilatou, 2021). The aerosols can be conditioned in a wide temperature and relative humidity range to simulate different outdoor environmental conditions. Expanded measurement uncertainties ($k = 2$; 95 % confidence level) are typically below 10 %.

In North America, air quality allowances vary between countries, e.g., the National Ambient Air Quality Standards (NAAQS) in the

Table 4
Sample standards related to PM mass. TSP is the total suspended particles.

Document	Originating organization	Originating region	Application	Citing regulation[b]	Technology/ Instrumentation	Particle type
EN 12341:2014	CEN	Europe	Air quality	Directive 2008/50/EC	Gravimetric	$PM_{2.5}$, PM_{10}
CEN/TS 17660–1:2021[a]	CEN	Europe	Air quality	–	Low-cost sensors	$PM_{2.5}$, PM_{10}
US 40 CFR Part 50	EPA	USA	Air quality	National Ambient Air Quality Standards (NAAQS, e.g., US 78 FR 3228, 2013; US 85 FR 82684, 2020)	Gravimetric (Federal Reference Methods, FRMs)	$PM_{2.5}$ (Appendix L), PM_{10} (Appendix J), $PM_{10\text{-}2.5}$ (Appendix O)
US 40 CFR Part 53	EPA	USA	Air quality	NAAQS	Federal Equivalent Methods (FEMs)	$PM_{2.5}$, PM_{10} (Subpart D), $PM_{10\text{-}2.5}$
Method No.: 8.06/2.0/M	ECCC	Canada	Air quality	Canadian Ambient Air Quality Standards (CAAQS)	Gravimetric (NAPS RM, time integrated)	$PM_{2.5}$
GB/T 15432-1995/XG1-2018		China	Air quality	GB 3095-2012/XG1-2018	Gravimetric	TSP
(UNECE R83, 2006) (light-duty vehicles, NEDC)		EU	Vehicle		Gravimetric	–
(UNECE R154, 2021) (light-duty vehicles, WLTP)						
(UNECE R49, 2021) (heavy-duty vehicles)						
(UNECE R96, 2019) (NRMM)						
(ISO 8178-1, 2020)	ISO	International	Vehicle			
US 40 CFR Parts 1065 and 1066	EPA	USA	Vehicle		Gravimetric	–
US 40 CFR Parts 9 and 86	EPA	USA	Vehicle (heavy-duty diesel trucks)		PEMS	
EN 13284-1	CEN	EU	Stationary source			
EPA Method 5 and 17	EPA	USA	Stationary source			
US 40 CFR Part 51 Method 201A and 202	EPA	USA	Stationary source			Total filterable PM, filterable PM_{10}, and filterable $PM_{2.5}$, condensable PM
ICAO Annex 16, Volume II (regulatory requirement) and SAE ARP6320B (technical standard)	ICAO and SAE	International	Aviation	Aircraft engine emissions certification	nvPM mass: performance specifications for instrumentation and calibration of	nvPM mass: diffusion flame combustion aerosol source with >80 % EC

[a] To be amended to include low-cost sensors.
[b] Note that the term *standards* has two meanings in the field. An air quality standard refers to regulations or codes that govern acceptable levels of particulate in the air, which is not consistent with the formal definition of a documentary standard (e.g. CEN and ISO) used elsewhere in this work.

United States and the Canadian Ambient Air Quality Standards (CAAQS). In the US and Canada, the categories of mass concentration have changed over time. Initially, measurements targeted TSP, before switching to a dichotomous approach (PM_{10} and $PM_{2.5}$) in the 1980s, with corresponding changes in the measurement principle. Measurement procedures in North America often reference those set out by the Environmental Protection Agency (EPA) and are traceable to the National Institute of Standards and Technology (NIST, USA). Measurement technologies are similar to those used in the EU. The EPA has defined both Federal Reference Methods (FRMs), which amount to gravimetric measurements using pre-weighed Teflon filters and size-selective inlets, and Federal Equivalent Methods (FEMs), which, similar to the situation in the EU, allows for a broader range of technologies that must be benchmarked against gravimetric measurements.

Measurements have also expanded to use a broader set of technologies that are not yet adopted into standards. Most notably satellite-based measurements (e.g., the Multi-Angler Imager of Aerosols mission (NASA)) provide an additional wealth of spatially resolved data but are not as reliable or accurate as ground-based sampling, which is often used to benchmark the measurements. This makes it challenging to standardize these measurements.

4.3. Light- and heavy-duty vehicle PM emissions

The standards for the measurement of PM emitted by a vehicle or internal combustion engine are described in the UNECE

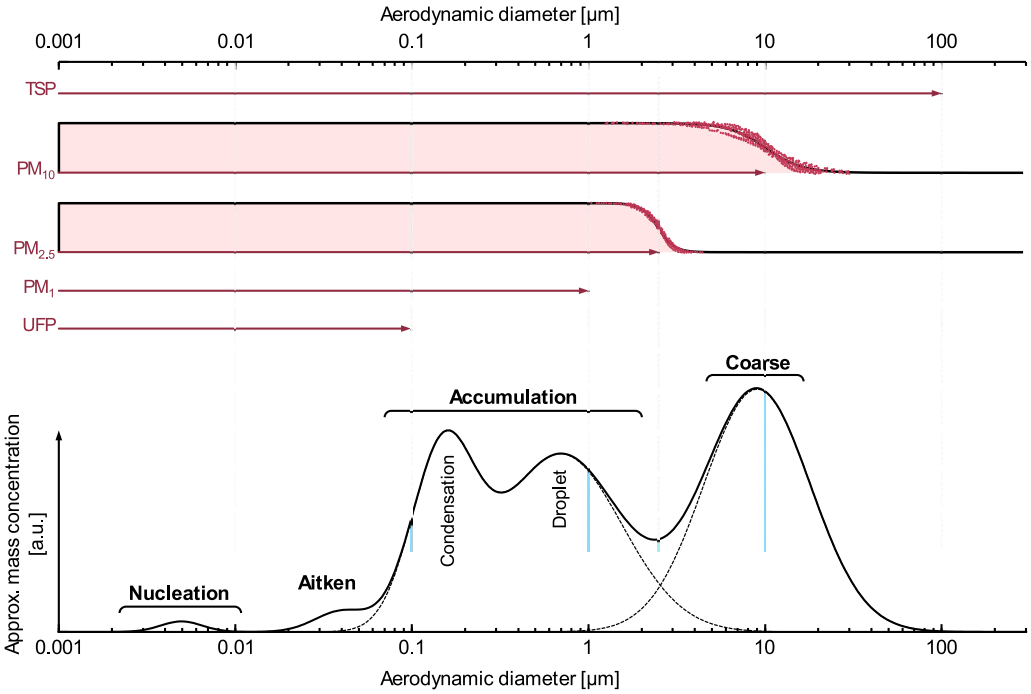

Fig. 4. Schematic demonstrating the persisting model of atmospheric aerosol size distributions, originally laid out by Whitby and co-workers (Whitby, 1978; Whitby, Husar, & Liu, 1972) and adapted by many works since, including Hinds (Hinds et al., 2022), Watson (Cao et al., 2013; Watson, 2002). Sample data for measurements of a range of size selected inlets are overlaid on the PM_{10} and $PM_{2.5}$ curves, with data digitized from Patel and Aggrawal (Patel et al., 2022) and Cao et al. (Cao et al., 2013).

Regulations No. 83 (light-duty vehicles NEDC), No. 153 (ligh-duty vehicles WLTP), No.49 (heavy-duty vehicles) and No. 96 (NRMM), and the US Code of Federal Regulations (CFR) Title 40 Parts 1065 and 1066, or the Global Technical Regulation (GTR) No. 4 (UN/ECE, 2006, 2007; UNECE R49, 2021; UNECE R83, 2006; UNECE R96, 2019; UNECE R154, 2021; US CFR, 2024; US EPA). The basic sampling system is similar in all three standards and consists of: (i) an exhaust dilution tunnel, (ii) a transfer tube (iii) a pre-classifier (e.g. impactor or cyclone), to remove large particles, (iv) a temperature-controlled filter holder, and (v) a flow control system. The exhaust dilution tunnel is required to reduce the concentration of water and other semi-volatile species to reduce the likelihood of their condensation in the sampling system. The transfer tube is designed to minimize particle losses to the filter holder. The pre-classifier cut-off efficiency depends on the region; the US requires that the pre-classifier must not remove more than 1 % of PM at an aerodynamic diameter of 1 μm but also it must remove at least 50 % of PM at an aerodynamic diameter of 10 μm. Europe also requires that the pre-classifier must not remove more than 1 % of PM but that the 50% removal efficiency can be anywhere between 2.5 μm and 10 μm; while the GTR only reguires a 50 % removal efficiency between 2.5 μm and 10 μm. The filters are either polytetrafluoroethylene (PTFE) coated glass fiber or PTFE membrane, usually 47 mm in diameter. Filter holders must be kept at 47 °C ± 5 °C in the US and GTR standard while the European standard only defines an upper limit of 52 °C. The flow control system is required to control the face velocity of the filter and measure the volume of air sampled. The filter face velocity must be near 100 cm s^{-1} in the US, must not exceed 100 cm s^{-1} in the GTR, and be between 20 cm s^{-1} and 80 cm s^{-1} in Europe.

It is important to realize that the *particulate matter* measured by these standards is operationally defined. As the GTR No. 4 says "particulate matter means any material collected on a specified filter medium after diluting exhaust with clean filtered air to a temperature between 315 K (42 °C) and 325 K (52 °C), as measured at a point immediately upstream of the filter." Therefore, PM will consist of solid carbon, ash, condensed hydrocarbons, sulphates, nitrates, and even gas-phase substantaces that adsorb to the filter surface during sampling. Vehicle PM measurements are very challenging because the amount of PM emitted from modern vehicles is very small, requiring the use of ultra-high resolution balances, temperature- and humidty-controlled clean rooms, and careful filter handling and conditioning procedures.

As PM emission regulations tighten in the future (e.g. a proposed limit of 0.5 mg $mile^{-1}$ for light-duty vehicles in the US in 2027) and more vehicles use aftertreatment particulate filters, the fraction of solid particulate collected on the filter will continue to decrease dramatically, while the fraction of PM that is volatile or semi-volatile increases. This exacerbates the uncertainties in the filter measurement. For example, a vehicle with a nominal PM emission rate of 0.7 mg $mile^{-1}$ was tested 28 times in three different test cells and the coefficient of variation (that is, the standard deviation relative to a reference value) of the tests was 28 % (Hu et al., 2014). Even at current emission levels, filter artifacts (adsorption of gas-phase material from the dilution air or filter transport, handling, and storage) and filter blank measurements can be comparable to the filter mass collected (Swanson et al., 2018). Thus, consideration is being given to updating the standards to increase the mass loading on the filter by increasing the filter face velocity, lowering the

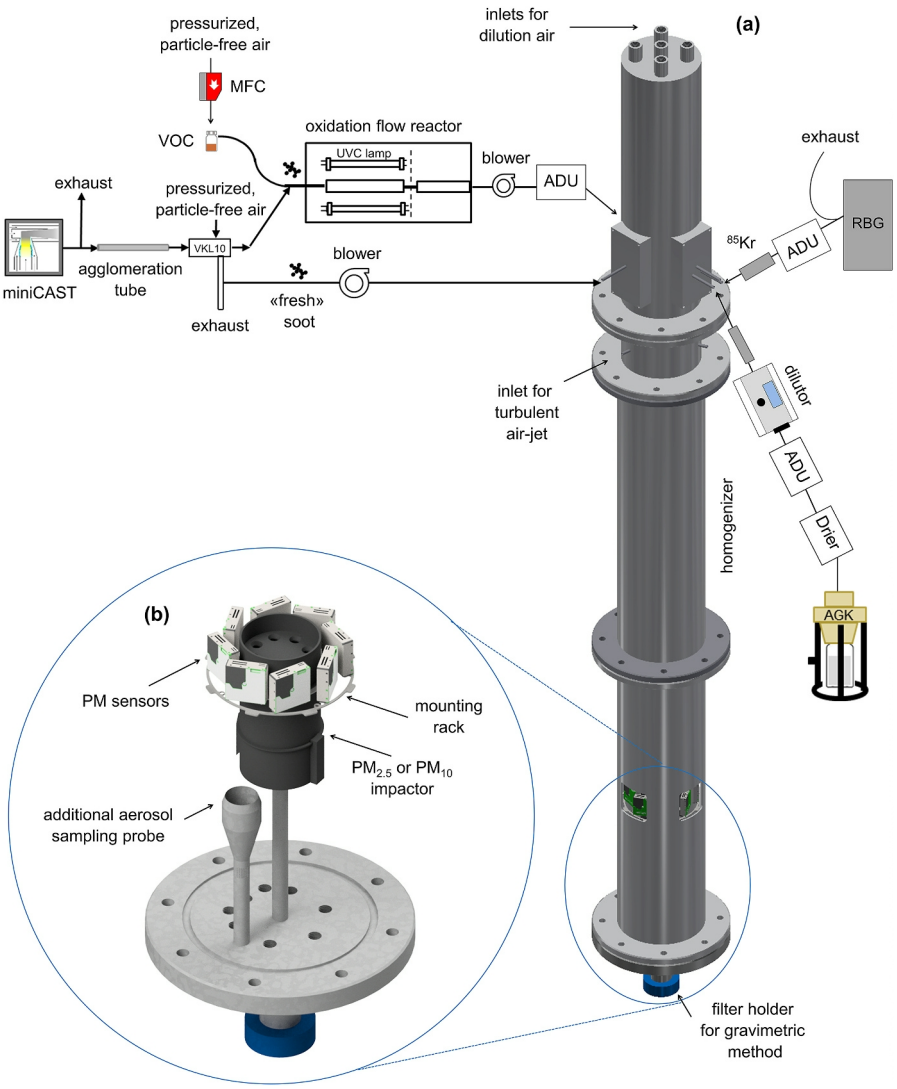

Fig. 5. Schematic illustration of the experimental setup for the calibration of low-cost PM sensors with synthetic ambient aerosols. The setup is equipped with temperature and humidity control systems (not shown in the picture). Figure taken from (Horender, Tancev, et al., 2021); licensed under a Creative Commons Attribution (CCBY) license.

dilution ratio, and using one cumulative filter for all drive cycle test phases (Xue et al., 2018).

The difficult and time-consuming nature of this measurement method means that these standards are not typically used for research in engine or aftertreatment development. Thus, many researchers use other instruments that measure surrogates of PM. These include particle sizing instruments where the relationship between particle size and mass is either measured separately or assumed (Olfert et al., 2019). This has driven the development of a number of real-time sizing instruments, including the differential mobility spectrometer (DMS500 (Reavell, Hands, & Collings, 2002)), engine exhaust particle sizer (EEPS (Johnson, Caldow, Pöcher, Mirme, & Kittelson, 2004)), and electrical low-pressure impactor (ELPI (Maricq, Xu, & Chase, 2006)); however, post-particulate filter emissions from modern vehicles can be near or below the detection limit of these instruments.

In the US, heavy-duty diesel trucks are also subject to emission limits during in-use testing under real-world driving conditions (EPA Title 40 CFR Parts 9 and 86) (US CFR, 2024). Testing requires the use of portable emission measurement systems (PEMS) and, in practice, a real-time PM sensor. The PM sensor in these systems typically consists of a unipolar diffusion charger (DC), an ion trap, and an aerosol charge (current) measuring device (Ntziachristos, Fragkiadoulakis, Samaras, Janka, & Tikkanen, 2011; Rostedt, Arffman, Janka, Yli-Ojanperä, & Keskinen, 2014). The current is a function of the number and size of the particles which indirectly correlates with the mass concentration of the aerosol. Since the DC method is not a direct measurement of PM, the PEMS system also incorporates a PM filter for gravimetric analysis to calibrate the DC sensor. Such systems have received 'alternate system approval' under provisions in 40 CFR Part 1065 (e.g. EPA Compliance Guidance Letters CD-12-08 (HDE) and CD-17-14 (HDE) (EPS DIS)). Alternatively, a BC sensor can be used in combination with the gravimetric filter method (e.g. as incorporated in the AVL M.O.V.E PM PEMS iX

instrument).

4.4. PM emissions from aircraft engines

Similar to the discussion for nvPM number regulations for aircraft engines, global aircraft engine certification requirements have set emission limits based on nvPM mass emission index (EI), that is the mass of nvPM particles emitted (in mg) per kg of fuel burned by the engine (see Eqs. (1) and (2) in (Lobo et al., 2015)).

The limits for nvPM mass, introduced as standards for certification of in-production and new type aircraft turbofan/turbojet engines with rated thrust >26.7 kN (ICAO, 2023), were adopted by ICAO Council in 2017 and have been enacted in national legislation by almost all ICAO member engine-producing countries by 2020. The previous regulation on particulates was based on smoke number. This was intended to limit visible emissions from aircraft engines, which was effective in reducing visible emissions from the 1970s to 2020, but has now been phased out and replaced by a new nvPM mass regulation (SAE ARP6320B, 2024). As with nvPM number, the technical requirements for nvPM mass were developed by SAE E−31. For nvPM mass, a new approach was needed, as other approaches did not limit the measurement of mass to only non-volatile particles. Stringent performance specifications were set on the nvPM mass instruments, including calibration accuracy of ±10 %. Two instruments that have been shown to meet all the requirements: (i) the Artium Technologies LII 300 (based on laser-induced incandescence) and (ii) the AVL Micro Soot Sensor (MSS, based on photoacoustic principle), both of which have a limit-of-detection of ~1 μg m^{-3} (Lobo et al., 2015). An assessment of three regulatory measurement systems for the determination of the non-volatile particulate matter emissions from commercial aircraft engines is presented in (Lobo et al., 2020). The calibration and regulatory apparati for measuring nvPM aviation emissions are shown in Fig. 6.

Calibration of these nvPM mass concentration instruments is currently performed by comparison to elemental carbon (EC) mass concentration as determined from thermal-optical analysis (TOA), with further information on this measurement provided in Sec. 5 on black carbon (BC)-related measurements. EC was adopted as a surrogate for nvPM, acknowledging that it is an operational quantity that is not directly equivalent to the nvPM mass (Corbin et al., 2020). The calibration must be performed at a minimum of four mass concentration levels from 0 μg m^{-3} to 500 μg m^{-3}. The particles for the calibration must be from a diffusion flame combustion aerosol source (DFCAS), which may include the Miniature Inverted Soot Generator (MISG), miniCAST, or a gas turbine engine. The source must have an elemental carbon to total carbon mass fraction (EC/TC) ≥ 80 % and must be demonstrated in concert with the mass instrument to elicit the same response as nvPM emissions from a gas turbine engine (ICAO, 2023; SAE ARP6320B, 2024). The calibration to demonstrate this performance must be conducted annually. There is a desire to reduce the uncertainty of the nvPM mass EI, much of which is associated with the determination of EC with TOA, and to make the measurements more directly traceable to mass standards. The CPMA (Centrifugal Particle Mass Analyzer) – Electrometer Reference Mass Standard (CERMS) is demonstrating strong potential as the reference to calibrate nvPM mass concentration instruments with markedly lower uncertainties (Corbin et al., 2020).

The sampling system is the same as was discussed in Sec. 3.3 for nvPM number. The high exhaust temperatures result in sampling losses of the nvPM particles due to thermophoresis, which must be corrected for prior to reporting the nvPM mass EI (ICAO, 2023). As

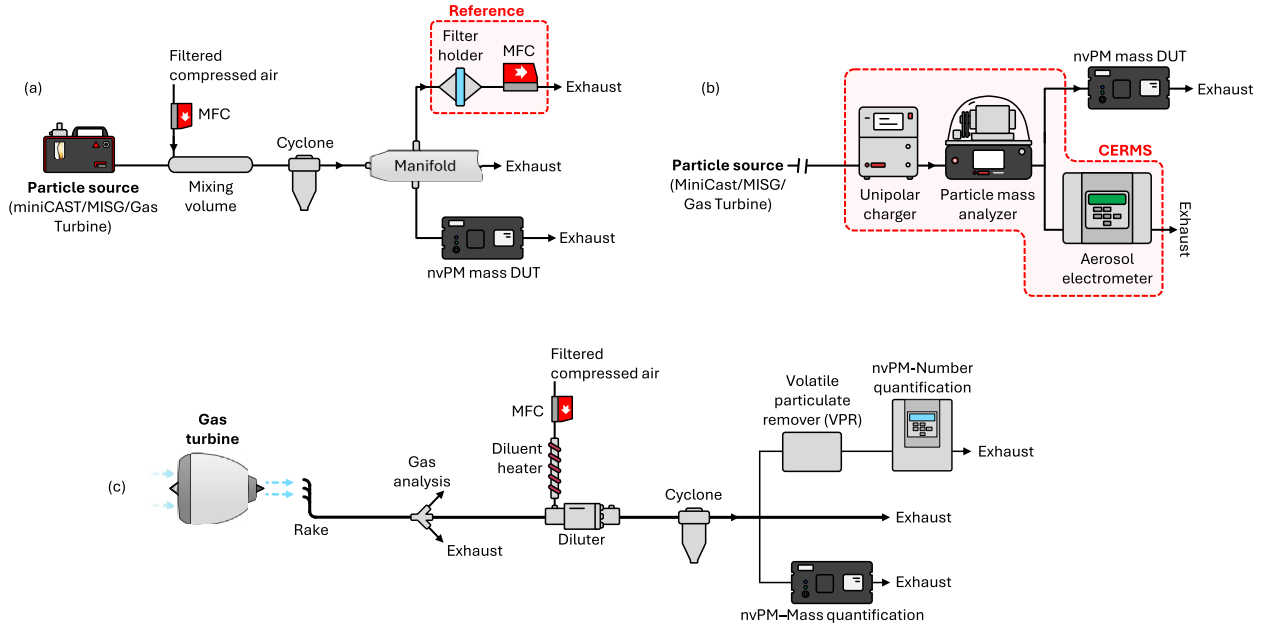

Fig. 6. The (a, b) calibration and (c) regulatory apparatus for measuring nvPM aviation mass concentration emissions. (a) Standard calibration setup employing filters for analysis by TOA. (b) The CPMA-Electrometer Reference Mass Standard (CERMS) in a proposed configuration as the reference to calibrate mass concentration.

with nvPM number, an ILC of three different reference sampling and measurement systems conforming to the SAE and ICAO specifications was conducted, in this case finding that the reference systems generally agreed to within 30 % of the average nvPM mass emission index (Lobo et al., 2020). With the dilution, for the lowest emission modern engines, the mass instruments are often operating at or below their limit-of-detection, and improvements are needed to reduce the uncertainty associated with the measurement of nvPM mass EI. In contrast to nvPM number, the system loss correction factors (used to predict the engine exit plane emissions when assessing health, climate, and local air quality impacts) for nvPM mass have lower values in the range from 1.1 to 2 (Durand et al., 2023).

4.5. Marine PM emission monitoring

At IMO (International Maritime Organization), BC emissions from marine vessels have been discussed for over a decade, but there is no activity that would lead to a regulation for international shipping. However, recently, IMO has approved the establishment of an Emissions Control Area (ECA) in the Arctic waters of Canada and Norway to limit the impacts of BC on the fragile northern climate (CAA, 2024). The state of marine emissions is covered in a recent review (Aakko-Saksa et al., 2023). An International Technical Working Group (TWG) has been established to develop a standardized sampling and measurement methodology for marine BC emissions, but limited funding for field work to evaluate options has hampered progress. Decisions about calibration methods, source particles, and what elements need to be standardized have not been adopted.

4.6. PM emissions from stationary sources

PM emissions from many different stationary sources are regulated. These sources include, but are not limited to, electrical powerplants, refineries, pulp and paper mills, cement plants, steel and aluminum plants, and waster combustors. There are hundreds of regulated emission limits depending on the source of the emission and the jurisdiction. For example, in the US, existing coal-fired power plants must emit less than 0.010 pounds of filterable PM per million British thermal units of heat input, while for new Portland cement kilns the emission limit is 0.02 pounds of PM per ton of produced clinker (40 CFR Part 63). In the US the measurement method of PM emissions from stationary sources is described in EPA Method 5 or Method 17 (US EPA, 2017; 2020a) and in Europe in EN 13284-1 (Stationary source emissions, 2017). The measurement of PM from stationary sources is unique because the sample must be collected directly from the stack (i.e., without dilution) and the filter must be either placed: (i) directly in the exhaust stack or (ii) in a heated filter holder maintained at stack temperature (or other minimum temperature, typically 120 °C in the US or 160 °C in Europe). PM in this case is defined as the mass of particles collected by filtration at the specified temperature and pressure and measured after the filter has been dried under specified conditions. The PM is collected on the filter with an isokinetic sampling probe and no aerosol pre-classifier is used before the filter (i.e., total filterable particulate is measured rather than PM_{10} or $PM_{2.5}$).

Some exhaust compounds maybe in the gas-phase at stack temperature but would quickly form an aerosol when they cooled. Thus, US 40 CFR Part 51 Method 201A and 202 (US EPA, 2016; 2020b) account for this by measuring the 'filterable' PM and the 'condensable' PM. The filterable PM is defined as the mass of 'particles that are emitted directly by a source as a solid or liquid at stack or release conditions and captured on the filter of a stack test train', while condensable PM is defined as the mass of 'material that is vapor phase at stack conditions, but condenses and/or reacts upon cooling and dilution in the ambient air to form solid or liquid PM immediately after discharge from the stack. The method also distinguishes between total filterable PM, filterable PM_{10}, and filterable $PM_{2.5}$. The measurement system for filterable PM uses an isokinetic sampling probe, a PM_{10} cyclone, a $PM_{2.5}$ cyclone, and a PM filter (controlled to within ±10 °C of the stack temperature), as per Fig. 7. The method defines filterable $PM_{2.5}$ as the mass collected on the filter after it has been desiccated, defines filterable PM_{10} as $PM_{2.5}$ plus the desiccated mass recovered from the $PM_{2.5}$ cyclone using mechanical cleaning and acetone rinse, and defines the filterable total suspended PM as the sum of $PM_{2.5}$, PM_{10}, and the desiccated mass recovered from the PM_{10} cyclone and upstream sampling probe. To determine condensable PM, a series of dry impingers and a

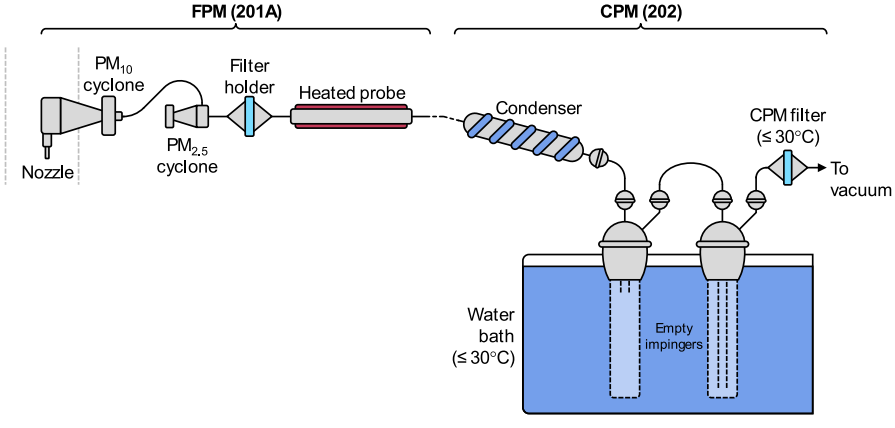

Fig. 7. The filtered particulate matter (FPM, left) and condensed particulate matter (CPM, right) from US 40 CFR Part 51 Method 201A and 202 setup. Figured adapted from Gong et al. (Gong et al., 2016) and the original EPA documents.

filter controlled to temperatures below 30 °C are added to the end of the filterable sampling train described above. Condensable material condenses in the impingers or nucleates and is captured by the filter. The condensable PM is then determined by weighing the organic and aqueous fractions in the impingers and filter after they have been desiccated.

5. Black carbon and elemental carbon metrics

BC is a class of mature carbonaceous particles often found in the atmosphere and is characterized by being strongly light absorbing. Naturally, then, aerosol light absorption is the metric used worldwide by climate scientists for studying and quantifying BC direct radiative forcing. Meanwhile, the mass concentration of BC is used for studying the effects of particulate air pollution on health. Neither metric is adequately standardized (Liu et al., 2023). In Europe, BC-containing particles have recently been characterized as an air pollutant of emerging concern, and BC mass concentration in ambient air might be regulated in the near future (Monforti-Ferrario et al., 2022). This is expected to drive standardization forward, at least in Europe, to ensure measurement data comparability between different monitoring stations.

BC mass concentration represents a subset of PM mass concentration, with specific methods targeting its measurement. Different terminology applies depending on how the carbon is measured (Petzold, Ogren, et al., 2013). Measurements in the atmosphere are typically carried out either as elemental carbon (EC) mass concentrations measured by thermal–optical analysis (TOA) or as equivalent BC (eBC) mass concentrations, where BC mass is derived from particle light absorption coefficient measurements together with a suitable mass absorption cross-section (MAC) for the conversion of light absorption coefficient into mass concentration. In the past few years, ambient measurements of refractory BC (rBC) mass concentrations – defined based on the incandescent emission from the particles (via laser-induced incandescence or LII) – have become more common. Definitions for the terms BC, EC, eBC, and rBC are provided in Petzold et al. (Petzold, Ogren, et al., 2013). BC-containing particles are also a major component of non-volatile PM (nvPM) emitted by most internal combustion engines, and so make up the majority of nvPM mass in diesel engine and aviation turbine emissions. In the following sections, we will present a short overview of methods for calibrating filter-based absorption photometers with respect to light absorption and for measuring EC and rBC mass in ambient air.

5.1. *Calibration of filter-based absorption photometers with respect to aerosol light absorption*

The light absorption coefficient of ambient aerosols is typically measured with filter-based absorption photometers, such as the Aethalometer, the Particle Soot Absorption Photometer (PSAP), the Continuous Light Absorption Photometer (CLAP) and the Multiple Angle Absorption Photometer (MAAP). These instruments are relatively affordable, robust, and suitable for unattended operation. They measure light attenuation through a sample collected on a filter relative to the clean filter, convert this to light absorption and then to eBC mass concentration by using a fixed MAC value. Measurements often suffer from artifacts, e.g., due to the scattering properties of the filter medium and the reduction of sensitivity as the filter gets progressively loaded, and thus necessitate corrections (for a more detailed discussion see (Drinovec et al., 2022) and references therein). As a result, they need to be calibrated against reference methods with known measurement uncertainties. Since the artifacts may depend on sample properties, calibration must be carried out with well-defined test aerosols covering the whole single scattering albedo (SSA) range.

In situ methods for measuring light absorption, such as the extinction-minus-scattering (EMS) or photoacoustic methods, have often been used as reference methods for the characterization of filter-based BC photometers (Petzold, Onasch, Kebabian, & Freedman, 2013; Sipkens et al., 2023). EMS relies on separate measurements of the aerosol extinction and scattering coefficients; the absorption coefficient is then calculated as the difference between these two parameters. Since EMS is an indirect method for determining the aerosol light absorption coefficient, it suffers from high uncertainties – sometimes exceeding 100 % – when the aerosols have a high SSA (Singh, Fiddler, Smith, & Bililign, 2014). Even when strict measurement and data post-processing algorithms are applied, measurement uncertainties remain significant (Modini et al., 2021). At low SSA values, on the other hand, the fractal nature of BC-containing particles and its effect on the scattering truncation correction of the nephelometer presents another systematic source of measurement uncertainty (Modini et al., 2021) – this is a specific manifestation of a broader EMS systematic uncertainty: the truncation correction of the nephelometer depends on the sample properties, such as size distribution. It should also be noted that the nephelometer response is a function of the refractive index of the particles (Massoli et al., 2009).

Photoacoustic spectrometers (PAS) (Nakayama et al., 2015) and photothermal interferometers (PThI) (Sedlacek et al., 2007) allow for a direct measurement of the aerosol absorption coefficient. Briefly, aerosol is drawn through the detection chamber, where the aerosol particles are illuminated with a powerful modulated or pulsed pump light source (laser or light-emitting diode) (Cao et al., 2021; Sharma, Arnold, Moosmüller, Arnott, & Mazzoleni, 2013; Tikhomirov et al., 2005). The light absorbed by the aerosol is converted to heat, causing a modulated increase in local temperature and changes in air density which create sound waves. PAS detect the generated acoustic waves while PThI probe the change in the refractive index of the air (see (Drinovec et al., 2022; Nakayama et al., 2015) and references therein). The greatest advantage of in situ methods is that at short (i.e. near ultraviolet) wavelengths these can be calibrated with gases, such as NO_2, which have a known absorption cross section in the visible spectral range (Arnott et al., 2000; Lack et al., 2006; Nakayama et al., 2015; Davies et al., 2018). Although ozone has been used in the past for calibrating PAS instruments, it is known to photodissociate at wavelengths up to approximately 1120 nm (Fischer et al., 2018). To reduce uncertainties due to poor stability of reference gas mixtures in the nmol mol^{-1} range in pressurized cylinders, SI-traceable NO_2 reference gas mixtures at low mole fractions can be generated dynamically with a mobile permeation generator (Haerri et al., 2017). Light-absorbing particles can be used instead of gases for calibrating absorption instruments at longer (infrared) wavelengths including: soot particles (Nakayama et al., 2015), Aquadag (Foster, Pokhrel, Burkhart, & Murphy, 2019), light-absorbing polystyrene spheres (Lack et al., 2006) and nigrosin

(Bluvshtein et al., 2017; Foster et al., 2019; Lack et al., 2006). Nigrosin is water soluble and forms spherical aerosols, which can be well described using Mie theory (Foster et al., 2019). However, the refractive index of nigrosin must be accurately known, and it must be noted that it's slightly variable between batches (Bluvshtein et al., 2017) and pH dependent (Radney et al., 2015). For the PTAAM-2λ (Haze Instruments, Slovenia) PThI, uncertainties of 4 % and 6 % (coverage factor $k = 1$, 68 % confidence interval) in the absorption coefficient have been achieved at wavelengths of 532 nm and 1064 nm, respectively (Drinovec et al., 2022).

Filter-based photometers can be calibrated against in-situ reference methods for light absorption in the laboratory by generating fresh and aged soot particles in a wide SSA range as shown in Fig. 8 (see (Kalbermatter et al., 2022) and references therein). Fresh soot particles are typically generated by a combustion generator, e.g. a CAST-type burner (see (Ess et al., 2018) and references therein), and can be further processed by coating them with primary organic matter (Salo et al., 2024). Aged particles are generated by coating soot with secondary organic or inorganic matter (Ess, Bertò, Keller, Gysel-Beer, & Vasilatou, 2021; Kalbermatter et al., 2022; Salo et al., 2024). The calibration parameters have been shown to depend on particle size, coating thickness and wavelength of the light source (Drinovec et al., 2022; Kalbermatter et al., 2022).

5.2. EC mass concentration with TOA

TOA is the primary method used to quantify the mass of elemental carbon within a number of applications (i.e., aviation, as per above). The technique has been reviewed in detail elsewhere (Boparai, Lee, & Bond, 2008; Karanasiou et al., 2015; Massabò et al., 2021). Briefly, carbonaceous particles are collected on a filter. The filter is later subjected to a range of elevated temperatures (the profile of which is termed a thermogram) in inert then oxidizing environments. This causes the carbon to devolve from the filters, where it is subsequently converted to CO_2 and detected downstream of the filter. At the same time, optical measurements are made to monitor the material on the filter. Organic carbon (OC) is the carbonaceous material that devolves from the filter at high temperatures in an inert environment while elemental carbon (EC) is the carbonaceous material that subsequently devolves from the filter at high temperatures in an oxidizing environment. Since a fraction of the organic carbon will pyrolyze during heating in the inert environment, optical analysis of the filter is used during the test to determine the 'split point' – the point at which all previous carbon should be defined as organic carbon. These two quantities are operationally defined, based on thermal profiles rather than fundamental physical characteristics of the particles. Even so, EC is sometimes taken as a surrogate for BC and is used as the standard for optical methods measuring particle mass (as when measuring aviation emissions (SAE ARP6320B, 2024)). Some discussion of the differences between BC, EC, and other related terms is provided by (Michelsen et al., 2020). A sample standardized experimental protocol for TOA is provided in NIOSH/NMAM 5040. In Europe, measurements are standardized by the CEN - EN 16909:2017 standard (CSN EN 16909, 2017) using the EUSAAR2 protocol (Cavalli, Viana, Yttri, Genberg, & Putaud, 2010), which is applicable for the measurement of elemental carbon (EC) and organic carbon (OC) following the requirement for all EU member states to measure EC and OC in particulate matter from June 2010 at background sites according to the Council Directive 2008/50/EC on ambient air quality.

Interlaboratory and related comparisons of TOA measurements have been the leading method to establish metrology for the technique, given limitations in establishing bottom-up uncertainty budgets for an operationally-defined quantity and the lack of a comprehensive set of reference materials to cover EC, OC, and TC (Baumgardner et al., 2012; Chai, Birch, & Deye, 2012; Panteliadis et al., 2015). In practice, this has led organizations like the Aerosol, Clouds and Trace Gas Research Infrastructure (ACTRIS) network to

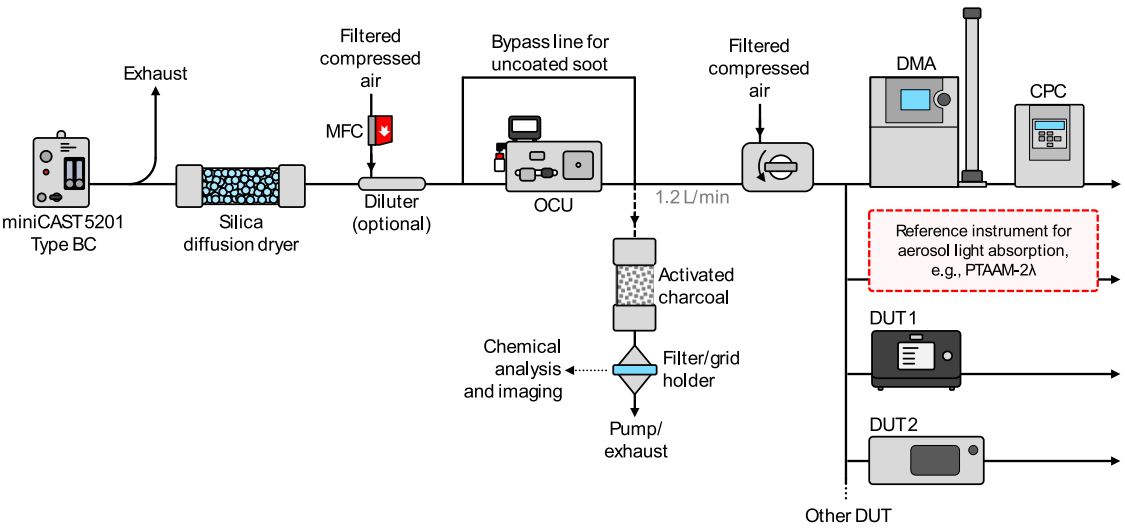

Fig. 8. Example of a setup that has been used for calibrating filter-based absorption photometers and in situ black-carbon-monitoring instruments against a reference method for light absorption (Kalbermatter et al., 2022). OCU stands for Organic Coating Unit, i.e. a type of oxidation flow reactor (Keller et al., 2022). Test aerosols exhibit an SSA in the range 0.03 to 0.7. PTAAM-2λ is a dual-wavelength photothermal aerosol absorption monitor developed by (Drinovec et al., 2022) and commercialized by Haze Instruments, Slovenia. Figure has been adapted from (Keller et al., 2022).

perform regular interlaboratory comparisons. A summary of the ILCs for TOA are provided in Table 5. Comparisons have also considered variations in the thermal protocol; char correction; the quantities defined by TOA (e.g., EC, OC, and TC); and have spanned a number of particle sources, with many of these details discussed in the associated reviews, e.g., (Karanasiou et al., 2015). Some such comparisons can be used to isolate other uncertainties that could be convolved with interlaboratory differences. For example, Conrad and Johnson (Conrad et al., 2019) isolate the effect of repeatability alone, while Zhang et al. (Zhang, Trzepla, White, Raffuse, & Hyslop, 2021) assesses the differences between instruments alone.

It is hopeful that new measurement standards and BC-like reference materials can be established to better align measures of BC to fundamental characteristics of the particles rather than operationally defined quantities. The use of catalytic strippers/thermal denuders to remove volatile coatings on the particles before measuring particle mass (e.g., gravimetric) in more traditional ways may provide a route to this outcome, though still having some operational effects (i.e., BC is taken as the amount of mass that survives the denuder).

5.3. rBC mass concentration measurement with LII

Laser-induced incandescence (LII) is increasingly being used for in situ rBC measurements of mature soot in combustion and ambient atmospheric environments. This technique involves heating soot particles with a high-power, pulsed or continuous-wave (CW) laser to temperatures high enough to emit measurable quasi-blackbody radiation. Instruments employing this technique include the LII 300 (Artium Technologies), which uses a pulsed laser, and the Single Particle Soot Photometer (SP2, Droplet Measurement Technologies, USA) which uses a continuous-wave laser instead of the pulsed laser, effectively sublimating absorbing particles. This emission is recorded either with gated (time-integrated) detection techniques, often for the spatially resolved visualization of particle-distribution or volume-fraction measurements, or with time-resolved detection methods, particularly for primary-particle size or single-particle measurements (see (Michelsen, Schulz, Smallwood, & Will, 2015) and references therein). One advantage of LII is that it is particularly sensitive to and highly selective for soot particles because they absorb strongly and can be heated to temperatures of ~4000 K before they sublime. While there are a number of limitations to the technique, including possible particle annealing effects and anomalies at both low (the volume fraction is underestimated for reasons that are not entirely understood) and high laser fluences (due to particle sublimation), careful selection of the laser fluence can mitigate these limitations.

Table 5
List of ILCs and closely aligned studies that are relevant to aerosol standards and uncertainty quantification. Many of these studies are summarized by (Karanasiou et al., 2015). AQUILA is the EU National Air Quality Reference Laboratories. TOR refers to a thermal–optical reflectance (TOR) method described in (Chow et al., 1993). Estimates of reproducibility coefficient of variation (CoV, or the relative standard deviation, which is computed by normalizing the corresponding uncertainty by the consensus value) are computed as averages (in quadrature) compiled across the tests in each work.

Reference	Source within work	Particle source	Protocol	Analyzers	Est. reproducibility CoV ($k = 2$)		
					EC	OC	TC
Countess (1990)	Table 3	Atmospheric, automotive exhaust, smog chamber	–	–	–	–	19 %
Birch (1998)	Table 5 (TOR, TOM)	Various	NIOSH 5040, TOR	–	36 %	11 %	–
Schmid et al. (2001)	Table 2	Atmospheric (Berlin, traffic)	Various	–	–	–	18 %
Schauer et al. (2003)	Tables 3 and 4	Atmospheric (ACE-Asia, including aviation, marine, USA)	Base case described in that work	Sunset analyzer	35 %[a]	16 %[a]	–
Emblico, Cavalli, Hafkenscheid, and Borowiak (2012)	Tables 4–6	Atmospheric (EU, AQUILA)	NIOSH-like, EUSAAR2	Sunset analyzer (including semicontinuous)	28 %	14 %[b]	10 %[b]
Cavalli, Douglas, and Borowiak (2012)	Table 4	Atmospheric (EU, AQUILA)	NIOSH-like, EUSAAR2	Sunset analyzer	–	–	15 %
Chai et al. (2012)	Table 2	Carbon black	Various	NIOSH 5040	16 %	16 %	13 %
Chiappini et al. (2014)	Tables 4–6	Atmospheric (France, CARA programme)	EUSAAR-2, NIOSH 5040, IMPROVE	NIOSH 5040, IMPROVE, EUSAAR2	27 %	16 %	10 %
Panteliadis et al. (2015)	Table S09 – S10	Atmospheric (5 European cities), sucrose	NIOSH870, EUSAAR2 (independently)	Sunset analyzers (including semicontinuous)	40 %, 52 %	–	24 %, 30 %
European Centre for Aerosol Calibration (2017)	Table 6	Atmospheric (2 European sites)	EUSAAR2[c]	Sunset analyzers (including semicontinuous)	–	–	15 %
Sipkens et al. (2024)	Table 3	Aviation (Gnome engine with catalytic stripper)	SAE protocol (SAE ARP6320B, 2024)	Sunset Model 5L	17 %	15 %	13 %

[a] Average excludes the aircraft test, which had anomalously large uncertainties and low loadings.
[b] Average excludes S14 test, which gave anomalously high uncertainties.
[c] Except two participants who used either the QUARTZ and NIOSH870 protocols.

Table 6
Technologies used to compute the particle size distribution alongside the corresponding standards that make use of those technologies.

Technology	Examples	Typical size range	Typical number concentration range	Typical flow range	Relevant standards
Optical particle counting (OPC)	OPCs for cleanrooms	0.3 μm–5 μm [a]	<10 cm^{-3}	up to 100 L min^{-1}	ISO 14644, ISO 21501-4
Optical particle sizing (OPSS)	11-D, OPS 3330	0.25 μm–10 μm [b]	up to a few thousand particles per cm^3	1 L min^{-1} to 5 L min^{-1}	ISO 21501-1
Aerodynamic particle sizing (APSS)	APS 3321	0.5 μm–15 μm	<3000 cm^{-3}	5 L min^{-1}	–
Aerodynamic aerosol classifier (with a particle counter)	AAC	0.025 μm–5 μm	- (classifier only)	0.3 L min^{-1} to 1.5 L min^{-1}	–
Bioaerosol monitors	Poleno, Rapid-E, POMO BAA500	1 μm–100 μm	down to a few tens of particles per m^3	from a few L min^{-1} to a few hundred L min^{-1}	A technical specification is being developed by CEN/TC 264/WG39
Differential mobility analysis (MPSS)		<0.01 μm–1 μm	10 cm^{-3} to 10^6 cm^{-3}	0.2 L min^{-1} to 5 L min^{-1}	ISO 15900 (DMA), ISO 27891 (CPC), CEN/TS 17434:2020

[a] Some instruments detect particle sizes in the range 0.1 μm–10 μm.
[b] Some OPSS models claim particle detection up to about 40 μm.

Measurements are implicit in that the technique is sensitive to volume fraction rather than mass, requiring some knowledge of the density, or calibration to a reference mass.

The SP2 can only detect particles with an rBC component equivalent to a spherical (geometric or mobility) diameter larger than about 70 nm (Yu et al., 2020). The most common calibration materials for the incandescence channel of the SP2 are fullerene soot and Aquadag (see (Gysel, Laborde, Olfert, Subramanian, & Gröhn, 2011; Liu et al., 2023) and references therein). The calibration curve is dependent on the chemical structure of the calibration material and, for instance, the SP2 is about ~23 %−29 % less sensitive to fullerene soot than to Aquadag; nevertheless, Aquadag-based SP2 broadband incandescence calibration curves can be recalculated to fullerene soot equivalent SP2 calibration following a simple equation (Baumgardner et al., 2012). It has been shown that fullerene soot behaves most similarly to ambient BC in terms of SP2 response per unit mass of BC, refractive index and effective density (see (Gysel et al., 2011) and references therein). Ideally, the SP2 would be calibrated with an appropriate particle source and a particle mass analyzer (such as an APM or CPMA) to determine the relationship between the incandescence signal and single particle mass (Moteki et al., 2010), and a condensation particle counter to determine the transmission efficiency of the SP2 (Schwarz et al., 2010).

According to a recent study, the observed correlation between rBC mass measured with SP2 and EC mass measured with TOA reveals a linear relationship with a constant ratio, providing clear evidence that both methods essentially quantify the same property of atmospheric aerosols (Pileci et al., 2021). However, significant systematic discrepancies in measured absolute values by up to a factor of 2 remain, highlighting the need for traceable reference methods or reference aerosols combined with a robust uncertainty budget for each method to clearly quantify the sources of discrepancies (Pileci et al., 2021).

6. Particle size distribution

6.1. Measurement methods

Airborne particle size can range from a few nanometers (e.g. nucleation particles) to several tens of micrometers (e.g. pollen particles). Moreover, particle number concentration can vary from < 1 cm^{-3} in clean rooms to a few thousand particles per cm^3 in ambient air, and even to several hundred thousand particles per cm^3 in industrial settings or in vehicle tailpipes. Inevitably, no single instrument can cover the entire particle size and number concentration range. Commercial instruments target specific applications, for instance:

i) many optical particle counters (OPC) are optimized for clean-room monitoring, with particle sizing for 0.3 μm to 5 μm (or sometimes 100 nm to 10 μm) and concentrations typically below 10 cm^{-3} (to avoid coincidence);
ii) optical and aerodynamic particle size spectrometers (OPSS and APSS, respectively) are suitable for ambient monitoring up to and beyond 10 μm and concentrations up to a few thousand particles per cm^3;
iii) bioaerosol monitors are developed for the detection of fungal spores and pollen particles at low concentrations;
iv) mobility particle size spectrometers (i.e., DMA combined with a CPC) can efficiently measure sub-micrometer particles.

Particle size is often expressed as radius or diameter for spherical particles, while equivalent sizes (e.g., mobility diameter, aerodynamic diameter, optical diameter, etc.) are often used for particles with irregular shape. Fig. 9 and Table 6 provide a short summary of common technologies for sizing aerosol particles. Previous studies have shown that each instrument type has a different size-dependent CE based on the measurement principle and particular design, and that the actual CE profile often deviates from the technical specifications defined by the manufacturer (Vasilatou et al., 2021; Vasilatou, Wälchli, et al., 2022). To be able to analyze data collected from different instruments and draw reliable trends of aerosol properties over long periods of time, instruments must be regularly calibrated against primary standards with small and well-defined measurement uncertainties.

6.2. Optical particle counters for clean rooms

6.2.1. Format of measured particle size distributions

Optical particle counters (OPCs) used in cleanroom applications exhibit a reduced number of size channels compared with other aerosol instruments dedicated to measuring particle size distributions. Typically, the allocation of size channels per decade in particle diameter is limited to fewer than five. The constrained number of size channels in OPCs can be attributed to two primary factors. First, OPCs derive their particle size information from the cleanroom classification table outlined in (ISO 14644, 2015). The particle diameters listed in the classification tables are 0.1 μm, 0.2 μm, 0.3 μm, 0.5 μm, 1.0 μm, and 5.0 μm, and these six sizes are sufficient for OPC users in various industries to promptly assess and confirm the adequacy of their manufacturing environment in terms of particle cleanliness. The second rationale pertains to the nonmonotonic relationship between light scattering intensity and particle size, which is primarily influenced by the use of a laser as the incident light source in OPCs. As illustrated in Fig. 10, which depicts light scattering intensity versus particle diameter for different particle materials, these values are computed using Mie scattering theory—a complete solution for the scattering of electromagnetic waves by homogeneous spherical particles (Dave, 1968; Mie, 1908). The incident light wavelength in this context is set at 800 nm. The intensity-versus-size curve tends to plateau within the particle diameter range of approximately 500 nm to 1 μm, diminishing the sensitivity concerning particle diameter in this specific size range. To counteract this, the incorporation of size channels with a relatively broader size width proves beneficial in sustaining a monotonic relationship between intensity and size. The size distribution determined by an OPC is presented as a cumulative particle number-based size distribution. This representation encapsulates the total number of particles with diameters greater than the specified size within a designated volume of air. This format enables OPC users to directly compare the results with the established classification table.

6.2.2. Optical diameter

ISO 21501-4 (ISO 21501-4, 2018) establishes performance requirements for commercial OPCs and outlines the calibration methodology for these instruments. These requirements include parameters such as size setting error, CE, size resolution, false count, maximum particle number concentration, and errors in sampling flowrate and sampling time. The evaluation of size setting error involves the use of size standard polystyrene latex (PS) spheres as calibration particles. It is crucial to recognize that the particle size measured by an OPC is the PS-equivalent optical diameter, denoted as d_o. This represents the diameter of the PS particles at which the intensity of the scattered light matches that of the detected particles. Fig. 11 qualitatively illustrates the measured value of d_o by the OPC when the real part of the refractive index, n, of a detected particle is lower than that of PS particles (Yabuki, 2018). Depending on the particle size range and the optical configuration (i.e., incident wavelength and collection angles) of a given OPC, the relative error between d_o and the geometric diameter can exceed 10 % if the n-value of the detected particle differs significantly from that of the PS spheres. In cleanroom environments, where particles are expected to have refractive indices close to those of polymers such as polyethylene, polypropylene, and nitrile butadiene rubber—materials comprising cleanroom suits and personal protective equipment—their n-values hover around 1.5 (Mitchell, 2003). Tryptic soy broth, which is the collection medium for airborne microbiological particles, and fluorinated polymers exhibit even lower n-values. In addition, geogenic ceramic particles such as silica and aluminosilicates, present in aerosols sampled in cleanrooms (Mohan et al., 2019), possess n-values greater than those of PS particles (Mitchell, 2003).

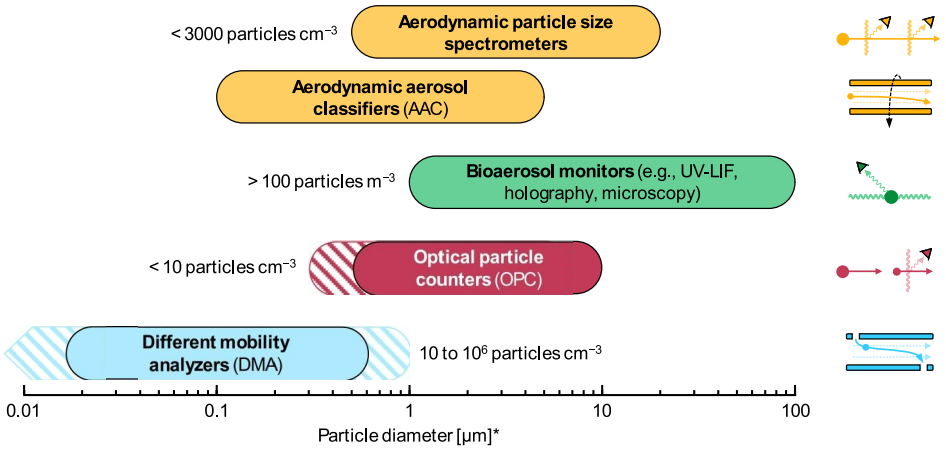

Fig. 9. Size ranges of common technologies for sizing aerosol particles. Note that the type of particle diameters varies between the different techniques (e.g., mobility diameter, aerodynamic diameter, optical diameter) (DeCarlo, Slowik, Worsnop, Davidovits, & Jimenez, 2004). Schematics on the right very roughly demonstrate the detection schemes, with smooth lines/arrows depicting particle trajectories; wavy lines depicting light interactions, such as scattering; and dashed lines indicating gas streamlines.

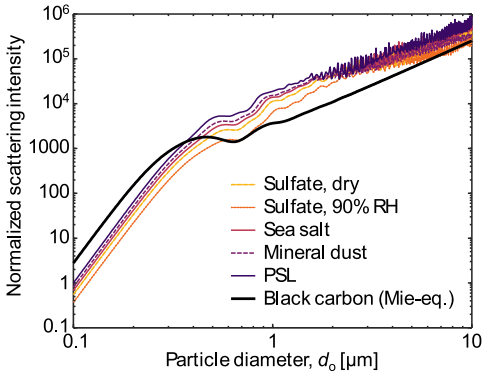

Fig. 10. Light scattering intensity from OPC, I_{OPC}, as a function of particle diameter based on Mie scattering theory for spheres of different particle materials. I_{OPC} was normalized by I_{OPC} of PS particles with 0.1 μm particle diameter (Yabuki, 2018). The incident wavelength was set to 800 nm. The center and width of the scattering angle were set to 90° and 40°, respectively. The complex refractive indices of mineral dust, dry sulfate, and sulfate droplets under 90 % relative humidity and BC (soot) were obtained from the optical properties of aerosols and clouds (OPAC) model (Hess, Koepke, & Schult, 1998). For simplicity, the BC curve corresponds to Mie-equivalent scattering, with aggregate structure causing the actual scattering to deviate from this trend. This plot is sourced from a review article on OPCs. The original graph, which was initially in Japanese, has been adapted with the consent from the Japan Association of Aerosol Science and Technology.

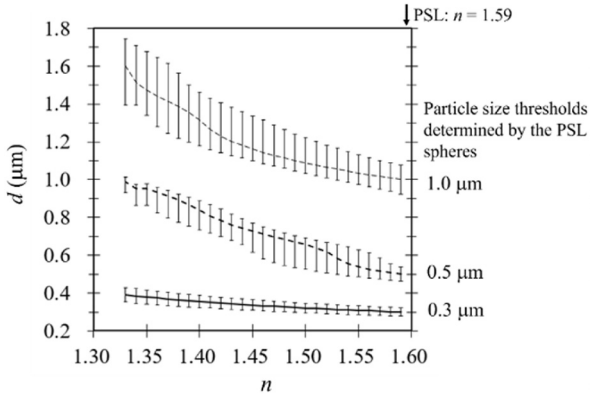

Fig. 11. PS equivalent optical diameter, d_o, measured by an OPC as a function of the real part of the refractive index n (Yabuki, 2018). The input wavelength and collection angles of the OPC align with those employed for the results depicted in Fig. 10. The error bar range signifies the size resolution of the OPC, which meets the stipulated requirement of 15 % in accordance with ISO 21501-4 (ISO 21501-4, 2018). As the n-value decreases from 1.59, the n-value of PSL, the value of d_o generally increases, leading to an overestimation of the particle diameter. The original graph, initially in Japanese, has been adapted with the consent from the Japan Association of Aerosol Science and Technology.

6.2.3. *Counting efficiency*

The CE of OPCs at a specific particle diameter is defined as the ratio of the number concentration measured by an OPC to that measured by a reference instrument for the same test aerosol. Alternatively, it is expressed as the ratio of the particle number measured by an OPC to that introduced to the OPC within a given sampling time (ISO 21501-4, 2018). Recent advancements in techniques have facilitated the evaluation of CE for particle diameters in the micrometer range. Pharmaceutical guidelines mandate the monitoring of particle number concentrations at sizes up to at least 5 μm as an indicator of microbiological particles (EU GMP, 2022). Consequently, OPC users in pharmaceutical industries are required to determine CE of their optical particle counters at 5 μm and above. ISO 21501-4 delineates two methods for calibrating CE across the micrometer range: the parallel comparison method and the generator method. Figs. 12 and 13 and Table 7 depict schematic diagrams and compare the characteristics of these two methods. Fig. 12 depicts the parallel comparison method, which uses size-certified PS spheres as calibration particles to evaluate the CE of OPCs. To minimize wall losses during particle transport to the instruments, this method employs a long mixing tube (Horender, Auderset, & Vasilatou, 2019). The number concentration at the inlet of a tested OPC, denoted as C_{inlet}, is preferably measured by a reference OPC. A reference CPC can also be used as secondary reference standard, but only in the submicrometer particle size range. Conversely, the generator method shown in Fig. 13 employs an inkjet aerosol generator (IAG) to generate monodisperse aerosol particles at a constant rate (Iida et al., 2014, 2021). The particle material typically used is lactose monohydrate, with a reported refractive index (n-value) of 1.53 (Bushill, Wright, Fuller, & Bell, 1965). The value of C_{inlet} is determined by the particle generation rate of the IAG divided by the measured sampling flowrate of the tested OPC (Iida et al., 2018). Inter-comparison studies conducted between the Swiss and Japanese NMIs

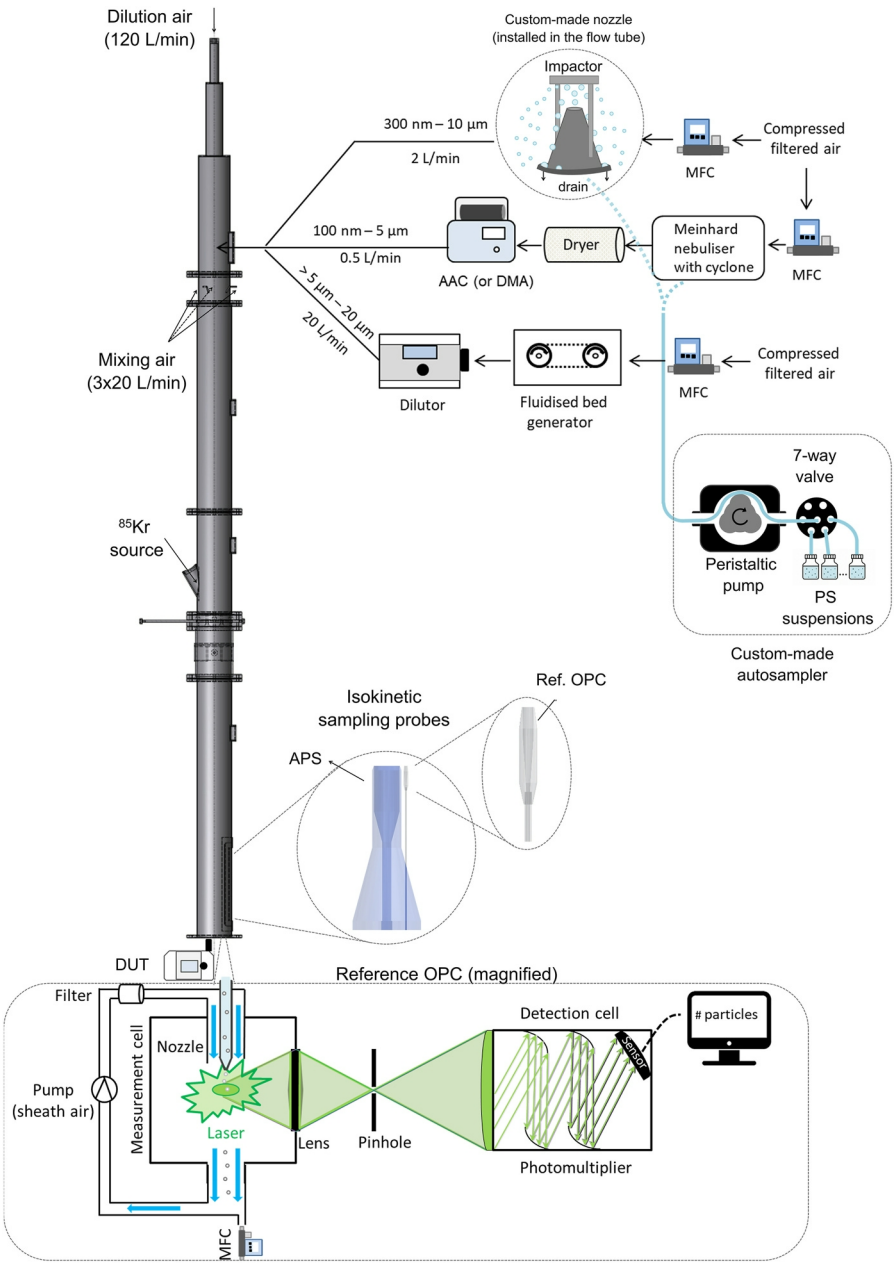

Fig. 12. Schematic illustration of the experimental setup for the parallel comparison method with a distribution tube. The parallel comparison method uses size-certified PSL particles as calibration aerosols and a custom-made reference OPC to determine the CE of a device under test (DUT) along with other parameters, such as size resolution and size accuracy. The setup has been used for calibrating different aerosol monitoring instruments, such as CPC, OPC, APS, bioaerosol monitors and aerosol diluters. Figure taken from (Vasilatou, Wälchli, et al., 2022); licensed under a Creative Commons Attribution (CCBY) license.

demonstrate that the CE obtained by both methods agrees within the stated uncertainties, typically within less than 10 %, across a particle diameter range from 0.5 µm to 15 µm (Vasilatou et al., 2020, 2021; Vasilatou, Wälchli, et al., 2022).

The Aerodynamic Aerosol Classifier (AAC) (Tavakoli, Symonds, & Olfert, 2014)) is a valuable instrument for generating test aerosols when assessing the CE of OPCs (Sang-Nourpour et al., 2019). This is particularly evident in accurately evaluating the CE curve near the minimum detectable size (d_{50}) of an OPC, as defined by ISO 21501-4 – the diameter at which the CE reaches 50 %, typically denoted as d_{50} with values of 0.1 µm or 0.3 µm (ISO 21501-4, 2018). A notable limitation of ISO 21501-4 in CE determination is its reliance on PS particles, which are available only at specific sizes, preventing an accurate characterization of the exact shape of the CE curve. CE curves often exhibit a sharp drop near the d_{50} value. Although a differential mobility analyzer theoretically can classify particles up to 1 µm based on particle electrical mobility, it introduces a population of multiply charged particles with larger mean

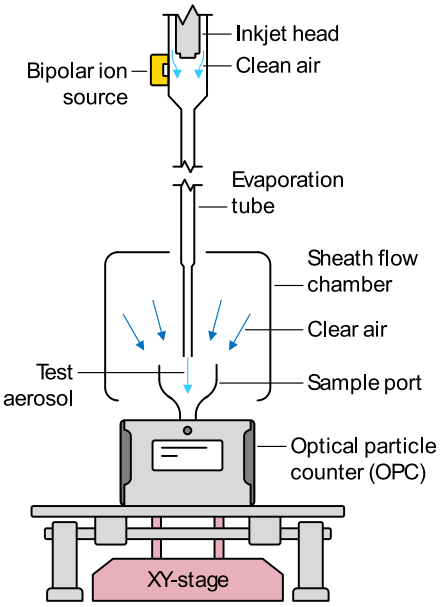

Fig. 13. Schematic illustration of the setup for the generator method. This method relies on an Inkjet to produce monodisperse aerosol particles at a constant rate. Figure has been adapted from (Vasilatou et al., 2020); licensed under a Creative Commons Attribution (CCBY) license.

Table 7
Characteristics of the parallel comparison method and generator method.

Parallel comparison method	Generator method (IAG)
Liquid or solid particles are nebulized from solutions/particle suspensions or dispersed from dry powder; PS spheres can be used as test particles.	Monodisperse solid or liquid particles are generated from aqueous solutions.
Particle size range: typically, from 100 nm to 20 μm.	Particle size range: typically, from 0.5 μm to 30 μm.
Since the method can select the particle size using classification devices such as DMA or AAC, the lower cut-off region of the CE curve can be evaluated.	Not an appropriate method to evaluate the lower cut-off diameter of the CE curve.
PS geometric diameter directly traceable to the SI unit of length.	Particle volume equivalent diameter is traceable to SI through mass and density
The number of particles delivered to a tested OPC is measured with a reference instrument (e.g., a reference OPC or CPC).	The number of particles delivered to a tested OPC is known.

diameters (Wiedensohler, 1988). Approximately 4% and 17% of particles at 0.1 μm and 0.3 μm, respectively, are multiply charged under steady-state conditions. AAC distinguishes itself by classifying aerosol particles based on their relaxation time, ensuring that the classified particles do not include a population of multiply charged particles. By adjusting the rotation speed of the classifying region under a constant flowrate, the mean diameter of monodisperse particles can be varied nearly continuously as a function of particle diameter, especially in the region where CE can sharply drop from its plateau to 0 %. The CE curve near d_{50} has been successfully evaluated using AAC-classified oil droplets for two OPCs with d_{50} values of 80 nm and 300 nm (Tran et al., 2020). As depicted in Fig. 12, an AAC is integrated into the parallel comparison method, where it classifies PS particles generated by aerosolizing an aqueous suspension of PS particles (Vasilatou et al., 2021). Remarkably, the particle number concentration of PS particles does not significantly decrease after being classified by AAC. The transmission efficiency of AAC is approximately 80% at $d = 1$ μm to 2 μm and remains above 60% at $d = 5$ μm (Payne, Johnson, & Symonds, 2023).

6.3. *Optical and aerodynamic particle size spectrometers*

Optical particle size spectrometers (OPSS), also known as aerosol spectrometers, work in a similar fashion to OPCs. The main differences are the following: OPSS have a) more size channels and b) a much lower sampling flow rate than OPCs, typically 1 L min^{-1} to 3 L min^{-1}, which means that they can typically measure particle number concentrations of up to at least 3000 cm^{-3} without coincidence losses. Technical specifications and calibration procedures are described in the ISO 21501-1 standard.

Aerodynamic particle sizers (APS), such as the Model 3321 spectrometer (TSI Inc., USA), are compact and portable devices, which provide real-time aerodynamic measurements of particles from ~0.5 μm to ~15 μm. The Model 3321 APS uses an optical system with two partially overlapping laser beams. The time-of-flight of the particle between the two laser beams can be related to the aerodynamic particle size. For the conversion of the time-of-flight to aerodynamic particle size, spherical PS spheres of known geometric diameter and density are used. In the past, the size-dependent CE of APS up to 10 μm had been calibrated by collecting test aerosol particles onto

filters or inertial impactors (Armendariz et al., 2002; Peters et al., 2003; Volckens et al., 2005). Aerodynamic particle sizers, though not yet standardized, can be calibrated following the procedures developed for OPSS and OPCs, i.e. by comparison with a reference particle counter in the size range 100 nm to about 15 μm. The size-dependent CE can be determined with expanded relative uncertainties of 2% to 9 %, depending on particle size and number concentration (Vasilatou, Wälchli, et al., 2022). Experiments show that the performance of optical and aerodynamic particle sizers deteriorates with time, necessitating calibration at regular intervals, e.g., annually.

The aforementioned Aerodynamic Aerosol Classifier (AAC, Cambustion Ltd., UK) and a CPC (Tavakoli & Olfert, 2014; Tavakoli et al., 2014) can also be used to calibrate APS and OPSS. The AAC classifies particles between <100 nm and ~5 μm according to their aerodynamic diameter while the CPC measures the total PNC. Since the CE of the CPC drops above ~1 μm, care must be taken to correct for this bias. Also, losses within the AAC are not insignificant and the actual width of the AAC transfer function can differ from theory, so these issues must be accounted for in the data inversion process to make accurate size distribution measurements (Johnson, Irwin, Symonds, Olfert, & Boies, 2018).

6.4. Automated bioaerosol monitors

Recently, optical particle size spectrometers have been enhanced with various analytical methods, such as UV-LIF, holography, and machine learning to distinguish between viable and non-viable particles or even identify different pollen taxa and fungal spores (Buters et al., 2022; Maya-Manzano et al., 2023; Sauvageat et al., 2020; Šaulienė et al., 2019). Most of these automatic bioaerosol monitors have been designed to measure particle size distribution in the range ~1 μm up to ~100 μm, though particle losses due to impaction can be expected at sizes larger than about 10 μm to 20 μm. At the moment, bioaerosol monitors have only been calibrated with respect to the total particle number concentration for particle sizes up to 10 μm, with the methods described above for conventional optical particle counters (Iida, Ikeda, Minakami, & Sakurai, 2024; Lieberherr et al., 2021). Efforts to standardize methods for quantifying the uncertainties related to classification algorithms used for particle identification are made within CEN/TC 264/WG39 (Tummon et al., 2022). If necessary, fluorescent particles can be used to test the performance of the UV-LIF detector at the same time (Iida et al., 2024; Lieberherr et al., 2021).

6.5. Mobility particle size spectrometers

The international standard ISO 15900 (ISO 15900, 2020) describes procedures for the periodic tests and calibration of a mobility particle size spectrometer (MPSS). For the calibration of sizing by the MPSS, two methods are provided. One is a method for the evaluation of the sizing accuracy of the whole MPSS system including the software for the measurement and analysis of particle size distribution, as the MPSS is operated in a way it is usually used. The other is a method for the evaluation of the accuracy of mobility classification by the DMA and the determination of the mobility correction factor. For both methods, the use of particle size standards is required. ISO 15900 refers to ISO 27891 (ISO 15900, 2020; ISO 27891, 2015) for the calibration of the CPC, which is a component of the MPSS system. For the characterization of charge conditioners (i.e., bipolar/unipolar chargers and neutralizers), which is another important component of an MPSS system, a new international standard, ISO 19996 (ISO/DIS 19996), is under development to supplement ISO 15900. Methods for testing the accuracy of the total number concentration for the overall MPSS system, which reflects the performance of the CPC and the charge conditioner, the algorithm for data inversion and corrections for charging and diffusion loss, through parallel measurements with a reference CPC with laboratory-generated and ambient aerosols are described in an annex. The work by (Wiedensohler et al., 2017) serves as a basis for some of these calibrations and tests. Traceability diagrams in ISO 15900 explain how these calibrations and tests support establishment of SI traceability for both particle size and number concentration. In addition to the calibration procedures, ISO 15900 specifies the slip correction factor and the charge distribution function to be used in the derivation of the particle size distribution function (ISO 15900, 2020).

In Europe, the size distribution of sub-micrometer particles is monitored by various atmospheric observatories. The technical specification CEN/TS 17434:2020 (CEN/TS 17434, 2020) describes a standard method for determining particle number distributions in ambient air in the size range 10 nm to 800 nm at total concentrations up to 10^5 cm^{-3} based on MPSS. The document outlines the performance characteristics and minimum requirements of the instruments and other equipment to be used and makes recommendations for aerosol sampling, data processing and QA/QC procedures. Regarding instrument calibration, the requirements are well aligned with those laid out in the ISO 15900 and ISO 27891 standards, and the CEN/TS 16976 technical specification (CEN/TS 16976, 2016). The particle sizing accuracy of the DMA must be checked experimentally using size-certified PS spheres with a diameter of >80 nm (recommended particle size between 100 nm and 300 nm). The accuracy of the total particle number concentration, i.e. the number

Table 8
Documentary standards related to particle size distributions.

Document number	Originating organization	Originating region	Application	Technology/ Instrumentation	Particle type
ISO 21501-4	ISO	International	Clean rooms	OPC, calibration of	Typically, PS or lactose monohydrate
ISO 21501-1	ISO	International	Ambient air	OPSS, calibration of	Typically, PS or lactose monohydrate
ISO 15900	ISO	International	All applications	MPSS and DMA, calibration of	PS (or other size-certified spherical particles)
CEN/TS 17434	CEN	Europe	Ambient air	MPSS and DMA, calibration of	PS

concentration integrated over the MPSS particle size range, must be determined experimentally by comparison with the data of a reference CPC which fulfils the requirements of CEN/TS 16976. The measurement is performed with ambient atmospheric aerosols, which are dried, split in two parts and fed simultaneously to the MPSS under test and reference CPC over a period of at least 8 h.

Table 8 provides an overview of standards related to particle size distribution measurements in clean rooms and ambient air.

7. Geometric particle size

7.1. Measurement methods

Techniques to characterize the geometric particle size of aerosols largely rely on measurement standards external to aerosol science. Microscopy methods provide direct measurement of particle size, in contrast to indirect methods such as mobility analysis, light scattering, electro-gravitational balance, and impaction, which estimate particle size from the measurement of a property related to size. Another advantage of microscopy methods is that the shape (morphology) of particles can be observed in addition to particle size. Microscopy often serves as the primary measurement for the calibration of other aerosol sizing methods thanks to the high accuracy of microscopy techniques for linear measurements. Several microscopy methods exist for measuring particle size: optical microscopy, electron microscopy (EM), and scanning probe microscopy (SPM). While optical microscopy has a resolution limit of ~0.2 μm and is suitable for sizing solid particles having diameters of 0.3 μm to 20 μm, EM and SPM techniques have much lower resolution (in the nanometer range) and are suitable for sizing nanoparticles. Scanning electron microscopy (SEM; typical resolution of ~10 nm) and transmission electron microscopy (TEM; typical resolution of <1 nm) are the two common EM types, while atomic force microscopy (AFM) and scanning tunnelling microscopy (STM) are the two common methods in SPM family. In section 7.2, we focus on AFM and TEM as the two most-commonly used primary methods for particle size measurement with traceability to SI.

Another way to determine the geometric particle size for monodispersed size standard particles is the electro-gravitational aerosol balance (EAB) method (Ehara, Takahata, & Koike, 2006a, Ehara et al., 2006a) developed by the Japanese NMI (NMIJ). This method will be described in detail in section 7.3.

7.2. AFM and TEM

AFM is a technique for imaging surfaces by the atomic interaction between the final atoms of a sharp tip and the atoms of the sample surface. The tip is placed on a cantilever and topography images can be obtained by scanning the tip along the surface. AFM can be operated in two distinct modes: the static mode (also known as contact mode) and the dynamic mode (tapping and non-contact modes). AFM has provided a means to dimensional nanometrology for national metrology institutes (NMIs) since its transition from a purely qualitative to a quantitative instrument. Since 1990s, NMIs have developed metrological AFMs that have the highest accuracy for dimensional measurements along with traceability to the SI unit of length (Bienias, Gao, Hasche, Seemann, & Thiele, 1998; Gonda et al., 1999; Haycocks et al., 2005; Klapetek, Valtr, & Matula, 2011; Meli, 2005; Picotto et al., 2001). Metrological AFMs use optical and/or X-ray interferometers to measure the position of a high precision lateral scanning stage and the relative displacement between the AFM tip and the sample in the vertical axis and the traceability is derived from the wavelength of light used to produce interference (Yacoot et al., 2011). Metrological AFMs are then used to calibrate a range of transfer standards such as one- or two-dimensional grating structures (pitch of ~100 nm) and step height standards (<10 nm). These transfer standards are used in turn to calibrate other AFMs used in industry or research institutes and have been used in international intercomparisons between NMIs (Garnaes et al., 2008; Koenders et al., 2003; Misumi et al., 2010). Research work has also been performed on calibration standards based on atomic lattices, such as graphite (Aketagawa, Honda, Ishige, & Patamaporn, 2007) and silicon (Yacoot, Koenders, & Wolff, 2007), although these standards are not used routinely yet.

When AFM is used for measuring particle size, often the height of the particle from the substrate is considered as particle diameter. For this measurement to be valid, the particles should be spherical and individually isolated (not agglomerated), and the substrate on which the particles are deposited must be as smooth as possible. Freshly cleaved mica substrate is commonly used for this purpose due to its very low roughness. If two or more spherical particles are adjacent (attached) to each other on the substrate, the pitch distance between highest points of the particles can be considered as particle diameter. The accuracy of particle size measurement by AFMs depends on several factors which should be considered in the uncertainty budget (Delvallée, Feltin, Ducourtieux, Trabelsi, & Hochepied, 2016; Misumi, Sugawara, Takahata, Takahashi, & Ehara, 2018; Yacoot et al., 2011): (i) accounting for interactions between AFM tip and sample; e.g., elastic deformation of particles due to contact from AFM tip; (ii) uncertainty in the position of the scanning stage; (iii) uncertainty in the position of tip and cantilever; (iv) influence of scanning speed; (v) uncertainty in the transfer standard used for AFM calibration; (vi) repeatability and noise level in the vertical direction; and (vii) temperature effects and particle dilatation. There is one standard for AFM-based measurement of nanoparticle size, ASTM E2859-11 (ASTM E2859-11, 2023), which provides a standard practical and metrological guideline for nanoparticle size measurement using AFM based on maximum displacement of the probe (i.e., particle height) (Hackley et al., 2023).

TEM is a technique that uses an electron beam which passes through the sample and produces images and diffraction patterns with atomic-scale resolution on a detector. Accurate size measurements with TEM depends on the calibration of magnification used for particle sizing. Since the early 2000s, NMIs (most notably, NIST and the Physikalisch-Technische Bundesanstalt PTB) performed experiments using the known value of the bulk silicon lattice spacing as a natural ruler to establish traceability to the meter for dimensional nanometrology applications. In 2019, the International Committee for Weights and Measures (CIPM) published a *Mise en Pratique* for the SI definition of the meter, which adopted the primary realization of meter based on the fixed value of speed of light in

vacuum as well as a secondary realization of meter for dimensional nanometrology based on silicon lattice spacing. This secondary realization of meter provides a pathway to traceability for TEM measurements. Calibration of TEM magnification by reference to a single crystal silicon artifact, where the crystal lattice is visible in the field of view of the TEM and by counting the number of lattice planes in the nanostructure. By using this method, expanded uncertainties below 1 nm ($k = 2$, confidence interval of approximately 95%) could be achieved for linear measurements smaller than 200 nm (CIPM, 2019).

Commercial calibration standards for TEMs, such as MAG*I*CAL®, are used by industry and research institutes to calibrate TEM magnification. This calibration standard consists of four sets of five SiGe alloy layers (each ~10 nm thick), alternating with ~13 nm thick pure silicon layers. The four sets of alternating layers (superlattices) provide dark and light contrast in the TEM and were directly calibrated by high-resolution TEM against Si (111) lattice spacing. Using this calibration standard, it is possible to calibrate the entire range of magnification in a TEM from about 1000 × up to 1 000 000 × . Other types of calibration standards for TEM, specifically for life science applications, have also been developed in recent years, which are described in ISO/AWI/TS 20510 and are ultimately traceable to silicon lattice spacing (ISO/TS 20510, in preparation).

Several international standards exist on measurement and characterization using TEM. ISO 29301 (ISO 29301, 2023) specifies calibration methods of TEM magnification over a wide range by using reference materials with periodic structures, such as a diffraction grating replica, a super-lattice structure of semiconductor, and a crystal lattice image of carbon, gold, or silicon. ISO 21363 (ISO 21363, 2020) describes how to prepare particle samples and capture, measure, and analyze TEM images to obtain particle size and shape distributions. In addition to calibration and particle size measurement, standard methods for characterization of certain nanomaterials such as carbon nanotubes using TEM are described in (ISO/TS 10797, 2012) and (ISO/TS 11888, 2017). ISO 22292 (ISO/TS 22292, 2021) also describes 3D image reconstruction of nano-objects using TEM.

Several ILCs on particle size measurement have been performed in the past two decades. The studied range of particle nominal size in these ILCs were typically from about 10 nm to a few hundred nm, making the results of such ILCs of interest to the aerosol community. These ILCs often include a wide variety of analytical methods for particle size measurements, including electron microscopy (SEM and TEM), AFM, dynamic light scattering (DLS), small-angle X-ray scattering (SAXS), differential mobility analysis (DMA), particle tracking analysis, X-ray diffraction (XRD), and field flow fractionation. These methods include direct and indirect sizing methods and are based on different measurands; thus, care should be taken when a comparison between methods is made. It should be noted that most nanoparticle samples used in these ILCs were in the form of liquid suspensions and various sample preparation protocols suitable for each analytical method were used. For methods such as DMA, nanoparticles were aerosolized from liquid suspensions using atomization or electrospray prior to measurement. Here, we briefly describe the main findings of a recent comprehensive ILC on particle size measurement (Lin et al., 2019), organized by Asia Pacific Metrology Programme (APMP) where several NMIs participated in the study. Interested readers are referred to more particle size ILCs available in the literature, such as

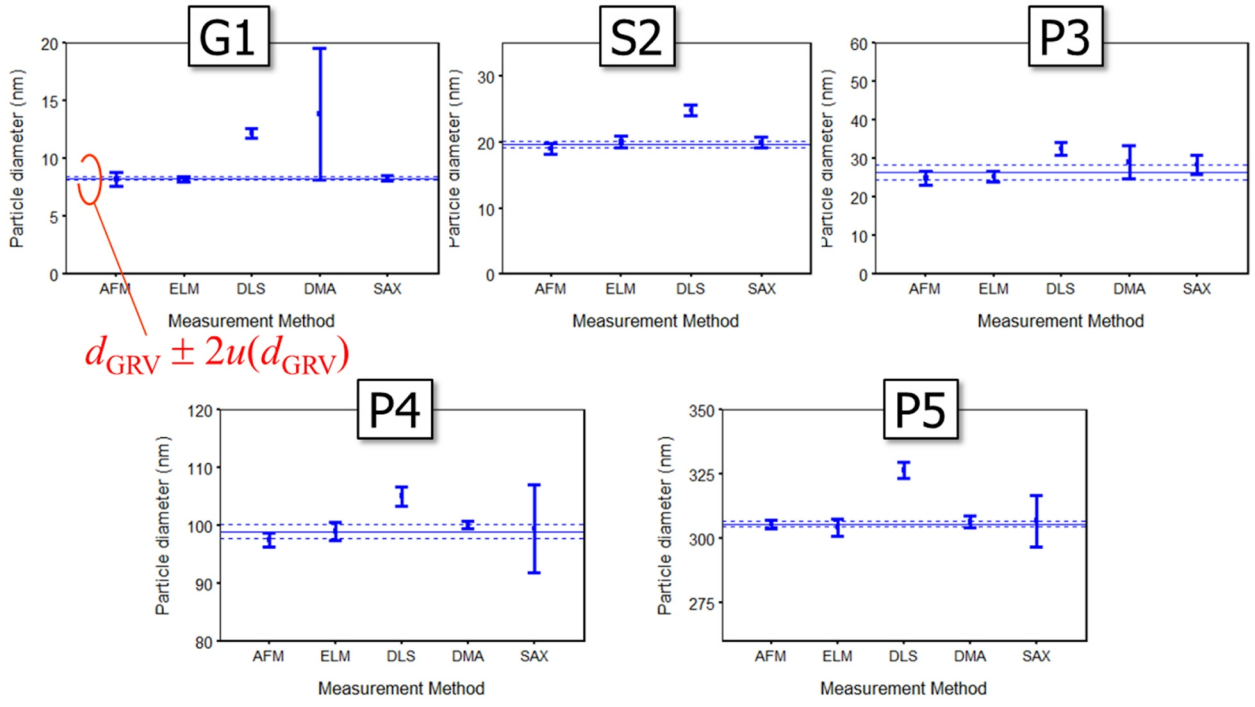

Fig. 14. A summary of measurement results from the ILC study by Lin et al. (2019). The method-dependent reference values (MRV) and global reference values (GRV) for particle diameter, d, for all 5 particle samples and different measurement methods are shown. MRVs ± $2u$ (expanded uncertainty at $k = 2$) are shown as data points and associated error bars and GRV ± $2u$ (expanded uncertainty at $k = 2$) is shown as solid line and associated dashed lines. Figure taken from (Lin et al., 2019) as a public domain report available from BIPM website.

(Hole et al., 2013; Lamberty et al., 2011; Langevin et al., 2018; Motzkus et al., 2013; Mulholland et al., 2024; Pauw, Kastner, & Thunemann, 2017; Rice et al., 2013; Roebben et al., 2011).

The APMP-organized ILC (Lin et al., 2019) involved 37 international laboratories from NMIs and other research organizations and studied the size of particles over a wide range of sizes and compositions: 10-nm gold particles (G1); 20-nm silver particles (S2); and 30-nm, 100-nm, and 300-nm polystyrene latex particles (P3, P4, and P5, respectively), all samples in liquid suspension form. The analytical methods for particle size measurement included AFM, EM (TEM and SEM), DMA, and SAXS. The method-dependent reference values (MRV) and global reference values (GRV) for particle diameter, d, were determined by statistical analysis of measurement results and associated uncertainties for each analytical method and for all analytical methods, respectively. Fig. 14 shows the summary of MRVs ± expanded uncertainty (data points and associated error bars) and GRV ± expanded uncertainty (solid line and associated dashed lines) for all 5 particle samples and various measurement methods. This figure shows the consistency results of the MRVs for all 5 nanoparticles based on the largest consistent subset method. It also shows that the MRVs were found inconsistent for all 5 nanoparticles. The MRV of the DLS is consistently larger than the other 4 MRVs for all nanoparticle sizes. As a result, the MRV of DLS was always excluded when constructing GRV. Finally, these results show that AFM and EM (TEM and SEM) methods were in good agreement with each other considering their uncertainties for all studied particles sizes, highlighting that the use of these techniques as primary methods for particle sizing is appropriate.

7.3. Electro-gravitational aerosol balance method

This method has been applied for the assignment of particle size in the range from 100 nm to 1 μm with traceability to the SI through size standard particles, such as NMIJ CRM 5722-a (NMIJ, 2024). In the most recent study, the relative expanded uncertainties (coverage factor of 2) in the particle diameter measurement were about 0.4 % and 0.2 % for polystyrene particles of nominal diameters of 100 nm and 300 nm, respectively (Takahata, Sakurai, & Ehara, 2020). The method enables the determination of the number-weighted average of volume-equivalent diameter of monodispersed particles by combining it with a separate measurement of the density of the particles and assuming that the particles are spherical. The measurement of the EAB method is performed with a Millikan-cell type instrument, which has two parallel, horizontally oriented electrodes with a gap of a precisely known distance and utilizes the balance of electrostatic and gravitational forces to measure the average mass of particles (Fig. 3 in (Takahata et al., 2020)). Monodispersed particles suspended in water are aerosolized, charge conditioned, classified with a DMA to remove residual particles formed during the aerosolization, and introduced into the gap between the electrodes in an aerosol form. Particles in the gap feel upward electrostatic force, in addition to the downward gravitational force, by applying a voltage of an appropriate polarity between the electrodes since all particles from the DMA are charged. Depending on the relative magnitudes of the two forces, particles drift either upward if the electrostatic force is greater, or downward if the gravitational force is greater, and are eventually 'lost' by attaching to the surface of either electrode. In a measurement of the EAB method, a constant voltage is applied between the electrodes after particles are introduced into the gap for a pre-determined holding time. The number of particles initially introduced into the gap and the number of remaining particles in the gap after the holding time are counted with a CPC. The ratio of the concentration after the holding time to the initial concentration gives the 'survival rate'. Repeating this measurement at different voltages with the same holding time creates a monomodal triangular (non-diffusive) or bell-shaped (diffusive) curve in a spectrum of the survival rate against applied voltages (Figs. 4 and 6 in (Takahata et al., 2020)). The mode voltage of the curve corresponds to the case that the gravitational and electrostatic forces on the particles are balanced, which is expressed with Eq. (2):

$$m_{\mathrm{p}}\left(1-\frac{\rho_{\mathrm{air}}}{\rho_{\mathrm{p}}}\right)g=\frac{eV}{H} \qquad \text{(Eq. 2)}$$

where m_{p} is the average particle mass, ρ_{air} the density of air, ρ_{p} the density of the particles, g the gravitational acceleration, e the elementary charge, V the voltage applied between the electrodes, and H the gap distance between the electrodes. The left-hand term is for the gravitational force with the buoyancy correction while the right-hand term is for the electrostatic force. In Eq. (1), it is assumed that all particles have one net charge, which can be realized for monodispersed particles by passing them through a DMA with an appropriate transfer function setting. The particle mass can be calculated by substituting the mode voltage into Eq. (2). The measurement of the density of particles, ρ_{p}, is achieved by putting the particles into a series of bottles filled with aqueous sodium chloride solutions with slightly different concentrations and hence slightly different densities at a given temperature. Depending on the density of the particles relative to the density of the aqueous solution in a bottle, the particles move up to the solution surface, move down to the bottom of the bottle, or remain suspended (Fig. 3 in (Ehara et al., 2006b)). The density of the solution for the bottle in which the particles remain suspended corresponds to the density of the particles. After these mass and density measurements, the average volume-equivalent diameter, d_{ve}, is calculated with Eq. (3):

$$d_{\mathrm{ve}}=\left(\frac{6m_{\mathrm{p}}}{\pi\rho_{\mathrm{p}}}\right)^{1/3} \qquad \text{(Eq. 3)}$$

The accuracy of the EAB method has been investigated by comparison of measured particle diameter with other techniques. A recent study conducted by the NMIs in the U.S. (NIST) and Japan (AIST) compared the diameter of 100 nm PS spheres measured by DMA, AFM, and SEM by NIST, and by EAB by AIST (Mulholland et al., 2024). The measured average particle diameters agreed within 3 % for all the methods, in line with the uncertainty of the electrical mobility measurements (Kinney, Pui, Mulholland, & Bryner, 1991). However, the difference between the diameter by the EAB, which had the smallest reported uncertainty (0.4 %), and those by the other

methods, which had larger uncertainties (1 % to 4 %), was notable. Further precise investigation is necessary to elucidate the source of the difference.

Finally, it is worth noting that the EAB method can also be used to certify PS particles with respect to mass, which can serve to calibrate particle mass analyzers, such as the APM and CPMA.

8. Summary and outlook

Fig. 15 presents an overview of the aerosol physical properties (metrics) which are relevant to a range of applications, including air quality, emission control and atmospheric measurements. Table 9 summarizes the current state-of-the-art in the calibration of common aerosol instruments, such as particle counters and sizers, PM low-cost sensors and BC monitors. Typically, relative expanded measurement uncertainties lie in the range of 1 % to 5 % when the calibration is performed against a primary measurement standard, with the exception of aerosol light absorption, where the measurement uncertainties are higher.

Despite the remarkable progress in measurement science over the past decade, several challenges still need to be addressed. For instance, there is a need for size-certified particles, particularly those below 10 nm, to cover the full-range of DMA classification sizes. Silver particles, generated directly in the gas phase without any additives/surfactants, appear to be the best candidate material for this purpose. It is therefore crucial to investigate the measurement agreement between SEM, TEM and AFM for 10 nm silver particles through international inter-laboratory comparisons.

Furthermore, the expansion of current laboratory standards for number concentration to encompass particle sizes >15 μm, which are pertinent for pollen monitoring, could be a game-changer. The introduction of traceable fluorescence standards (e.g. fluorescent PS or other particles) with a fluorescence spectrum that mimics the properties of various bioaerosols (bacteria, fungal spores and pollen) could be instrumental in validating/calibrating the UV-LIF detectors of bioaerosol monitors. Lastly, the establishment of standardized procedures for assessing the performance of machine learning algorithms, particularly a robust uncertainty budget for particle taxon/type identification, could significantly enhance bioaerosol monitoring networks that employ automated bioaerosol instruments.

In the realm of emission control for on-road vehicles, there are active deliberations in several European countries regarding the potential implementation of PN-based procedures for PTI of gasoline direct injection (GDI) engines. Given that these engines tend to generate aerosols with smaller particle size and a higher amount of semi (volatile) compounds compared to diesel engines, dedicated field and laboratory studies should be carried out to define field measurement protocols of GDI engine exhaust as well technical specifications for the PN-PTI counters.

The primary issue for aviation emissions is the high levels of uncertainty associated with the measurement of nvPM number and mass. For mass, this is being addressed with consideration of improved calibration techniques, such as CERMS, and calibration particle

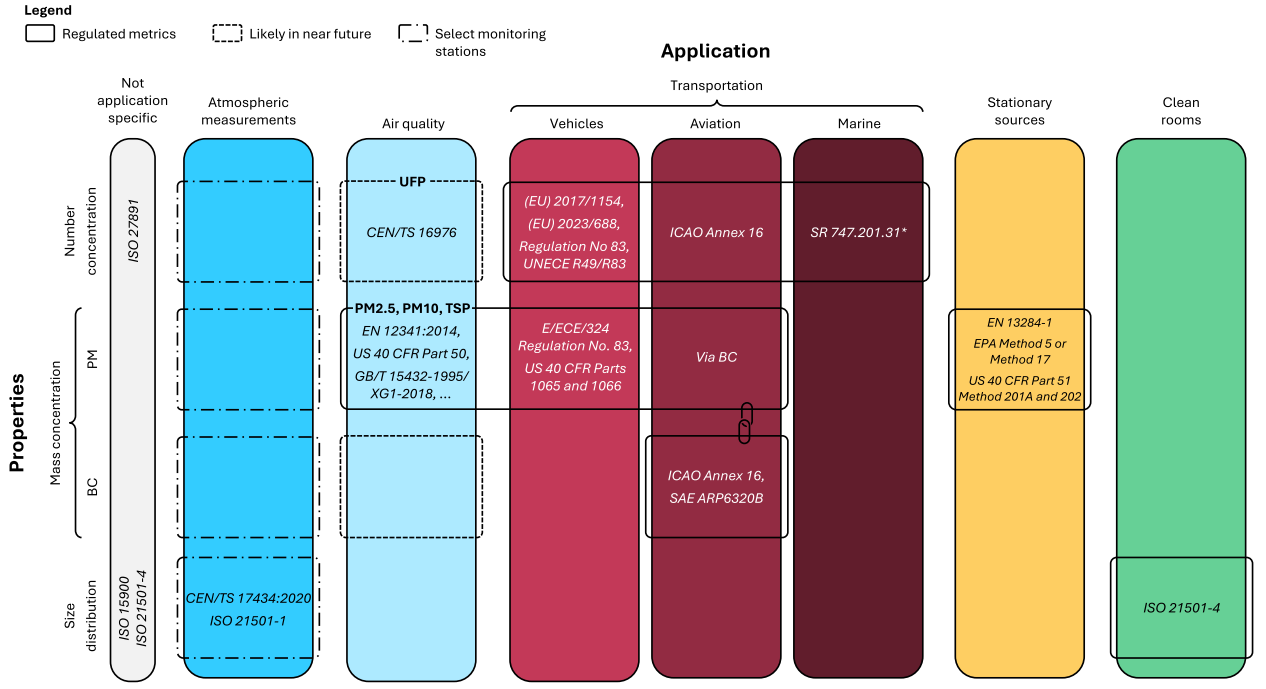

Fig. 15. Overview of aerosol physical properties (metrics) relevant to air quality, emission control, cleanroom and atmospheric measurements. Solid lines designate regulated metrics, while dashed lines designate metrics that might be regulated in Europe in the near future. All metrics are relevant to atmospheric observations and are being measured at selected monitoring stations for research purposes (dashed-dotted line). (*) Note that marine emissions are only regulated in Switzerland.

Table 9
Current state-of-the-art in the calibration of different aerosol instruments. U_{rel} denotes relative expanded uncertainty (coverage factor $k = 2$; 95 % confidence interval).

Instrument type	Typical particle size range	Traceable calibration range	Test/calibration aerosol	Primary standard	U_{rel} (%)	References
CPC/PN-PEMS/ PN-PTI	5 nm–200 nm	1 cm^{-3} to 1000 cm^{-3}	Various	Electrometer equipped with diluters or reference OPC	≥2	Sakurai et al. (2023)
		1000 cm^{-3} to >100 000 cm^{-3}		Electrometer or reference CPC	≥1.1	(Brown et al., 2023; Sakurai et al., 2023)
DMA	5 nm–1000 nm	>80 nm	PS	Microscopy methods (e.g. TEM, SEM and AFM)	>2 [a]	
		~10 nm	Silver particles (sintered, spherical)			
		100 nm to 1 µm	PS	Electro-gravitational aerosol balance (EAB) method	>0.2	(Ehara et al., 2006a, Ehara et al., 2006a; Takahata et al., 2020)
OPC/OPSS/APS	100 nm–15 µm	0.5 cm^{-3} to 800 cm^{-3}	Typically, PS	Reference OPC	2 to 9	(Horender et al., 2019; Iida and Sakurai, 2018; Vasilatou et al., 2021; Vasilatou, Wälchli, et al., 2022)
	500 nm - 15 µm	0.02 cm^{-3} to 2 cm^{-3} [b]	Typically, LM particles	IAG	1 to 6	
PM monitors and LCS	defined by PM impactor	≥10 µg m^{-3}	Synthetic ambient-like	Manual gravimetric method	≥4	(Horender, Auderset, et al., 2021; Horender, Tancev, et al., 2021)
nvPM mass	<1 µm (defined by very sharp cut cyclone)	50 µg m^{-3} to 500 µg m^{-3}	Diffusion flame combustion aerosol source (soot)	EC from TOA	17	Sipkens et al. (2024)
Filter-based photometers	Defined by size-selective inlet	Absorption coefficient ≤40 000 Mm^{-1}	Fresh and aged soot	PTAAM-2λ	8 to 12 [c]	(Drinovec et al., 2022; Kalbermatter et al., 2022)

[a] Depends on microscopy method. The agreement between AFM, SEM and TEM for 10 nm silver particles has not yet been investigated.
[b] The particle generation rate of the IAG is usually set at 10 particles s^{-1} or 30 particles s^{-1}. The concentration range depends on the sampling flowrate of DUT. Here, we assumed a sampling flow rate of 28.3 L min^{-1} and 1.0 L min^{-1} to calculate the lower and upper number concentration limit, respectively.
[c] Depending on wavelength of light source (see section 5.1 for more details).

sources, such a nebulized BC particles. For number, the major issue is the large losses in the lengthy sampling lines. Further research is needed to explore methods to ameliorate this challenge, such as placing instruments in close proximity to the engine (requiring robust instruments) or performing in situ measurements (requiring new non-intrusive measurement technology). Further challenges that are emerging for aviation include consideration of non-CO_2 climate effects, such as the formation of contrail cirrus clouds, which appear to be connected to particulate emissions at altitude, such as nvPM and volatile particulate matter (vPM), which includes sulphates and organic matter.

9. Conclusions

In this work, we have reviewed developments in aerosol standards that are well-known to metrologists, the industry and regulators, but might be overlooked by scientists performing basic aerosol/atmospheric research. We have aimed to provide a comprehensive overview of both laboratory and documentary standards that might be useful to research students, experts running monitoring stations or established researchers in neighboring fields wishing to improve the accuracy of their measurements.

Significant progress has been made in the physical charcaterization of ambient aerosols in the last few years. Primary measurement standards have been developed and validated for key metrics, such as particle number and mass concentration, size/size distribution, and light absorption. The expanded measurement uncertainties (95 % confidence interval) can be as low as 1.1 % for particle number concentration under ideal conditions. Even for aerosol light absorption, one of the most challenging metrics, recent advances in instrumentation have led to expanded measurement uncertainties of $<$15 %. Meanwhile, ISO and CEN documentary standards have been established, outlining methods to determine the measurement efficiency of common aerosol instruments, such as condensation, aerodynamic and optical particle counters and size spectrometers. In Europe, two new CEN documentary standards are in the pipeline, focusing on automated bioaerosol monitors and low-cost PM sensors, to standardize measurements in these rapidly expanding fields. The need for further standardization is evident, particularly for BC-related metrics, such as light absorption and BC mass concentration.

As new measurement techniques emerge, existing aerosol instruments are being miniaturized, and new emission control or air quality regulations are enforced, new aerosol standards must be designed accordingly. With emissions regulations becoming more stringent, there is a pressing need to characterize nanometer-sized particles at high concentrations, particularly soot. On the other hand, as climate change leads to longer pollen seasons and worsened allergy symptoms, very low concentrations of micrometer-sized bioaerosol particles – though not regulated – must also be monitored and identified. The complexity of ambient aerosols in terms of size, concentration, chemical and optical properties presents continuous challenges to scientists. To tackle these challenges, it is crucial to foster interdisciplinary collaboration. Similarly, it would be advantageous to address metrology requirements early in a process, whether it's the development of a new instrument or the design of a monitoring network. By establishing a suitable measurement framework and proper quality assurance procedures from the outset, scientists can significanlty enhance data quality and comparability.

CRediT authorship contribution statement

Konstantina Vasilatou: Writing – original draft, Supervision, Project administration, Investigation, Conceptualization. **Kenjiro Iida:** Writing – original draft, Investigation, Conceptualization. **Mohsen Kazemimanesh:** Writing – original draft, Investigation, Conceptualization. **Jason Olfert:** Writing – original draft, Investigation, Conceptualization. **Hiromu Sakurai:** Writing – original draft, Investigation, Conceptualization. **Timothy A. Sipkens:** Writing – original draft, Investigation, Conceptualization, Visualization. **Gregory J. Smallwood:** Writing – original draft, Investigation, Conceptualization.

About this review

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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