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1 Overview

Air pollution is an environmental and social issue; air pollutants come from both anthropogenic and natural sources and give rise to health and climate change concerns. Accurate aerosol metrology i.e., dimensional quantification and chemical analysis, is a prerequisite for enforcing regulations, protecting human health, and supporting research on climate change and atmospheric processes. However, air pollution is a complex metrology challenge that currently lacks traceable measurement and characterisation of aerosols in the environment. To address this, this project provided methodological improvements, including calibration guides and reference materials, calibration of particle size spectrometers, pollen monitoring instruments and the chemical characterisation of ambient aerosols. New automated pollen monitors were traceably calibrated for the first time within this project and their performance was successfully validated against previously used Hirst-type impactors at test-sites in Switzerland and Norway.

The accuracy and performance of portable instruments for measuring particles and black carbon was validated during several outdoor intercomparison field campaigns across Europe under variable environmental conditions. These types of instruments are frequently used by non-experts in industry and the range of instruments undertaken by the project represented the currently commercially available instruments. Hence, the project responded to current challenges regarding aerosol instruments used by non-experts. by developing a guide to the use of these instruments for measuring ambient air quality.

2 Need

For regulatory bodies, air quality networks and atmospheric instrument manufacturers to be able to perform to the best of their abilities, they all require accurate and reliable data for air quality monitoring. However, there is currently a lack of traceable calibration standards, and harmonised calibration procedures for measuring both anthropogenic and natural aerosols.

Measurements of both the particle number size distribution and the particle mass concentration are a major part of the operations within the European Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS). For measurements of particle number size distributions using Mobility Particle Size Spectrometers (MPSS), understanding the behaviour of bipolar charge conditioners is of paramount importance. The recent years saw a growing interest into the deployment of non-radioactive alternatives to the standard radioactive charge conditioners. Here, a need for a more systematic examination arose, not at least with regard to the long-term performance. For measurements of the particle mass concentration using Optical Particle Size Spectrometer (OPSS) and Aerodynamic Particle Size Spectrometer (APSS), an improvement of the equivalency with gravimetrically derived mass concentrations is needed, particularly in the cases with a significant fraction of coarse mode particles (EU Air Quality Directive 2008/50/EC, Annex VI and CEN EN 12341).

The quantification of element mass concentrations in aerosols is part of existing EU Regulations and is necessary for understanding the origins, behaviour, environmental fate and impacts of ambient particulate air pollution. However, current methods lack flexibility in terms of sensitivity and both time and spatial resolution. This may (in terms of the generally decreasing air pollution levels in Europe) challenge the practice of monitoring the required and regulated air pollutants. Therefore, air quality monitoring networks need strengthening with additional options, such as offering high detection sensitivity and flexibility for monitoring the temporal and spatial variability of air pollution sources.

Currently, pollen monitoring relies on manual methods, which suffer from poor time resolution, are time consuming and hence incur high labour costs. As a result, meteorological institutes across Europe are now planning to equip their monitoring stations with automatic pollen monitors. However, to date very little is known about the specifications of these automatic monitors. Thus, there is an urgent need for a rigorous investigation of their performance in the laboratory and in the field in order to establish a metrological framework for this newly emerged field.

3 Objectives

The overall goal of this project was to focus on the traceable measurement and characterisation of aerosols in the environment. The specific objectives were to

1. Use test aerosol particles in test chambers, to develop traceable methods for the calibration of optical and aerodynamic particle size spectrometers to determine mass and number concentration, as well as size distribution. In addition to characterise aerosol bipolar charge conditioners in MPSS and based on this the pre-normative definition of their performance criteria.
2. Develop new calibration procedures for automatic pollen monitoring instruments based on fluorescent polystyrene latex and real pollen particles (at concentrations $< 1 /\text{cm}^3$, and particle sizes of $0.5 \mu\text{m}$ up to $> 20 \mu\text{m}$, with target uncertainties of $< 10 \%$). In addition, comparisons to reference instruments in the field will be used to characterise instrument responses to pollen.
3. Develop certified reference substrates and traceable measurement techniques for the quantification of regulated and unregulated substances in ambient air with cascade impactor sampling and X-Ray Fluorescence analysis (XRF).
4. Develop portable instruments and software applicable for the measurement of ambient aerosol particle concentrations under variable environmental conditions. Knowledge gained will be applied to other appropriate classes of portable instruments by producing suitably adapted procedures.
5. Facilitate the take up of the technology and measurement infrastructure developed in the project by: the measurement supply chain (accredited laboratories), standards developing organisations (ISO and CEN) and end users (e.g. the Network of European Air Quality Reference Laboratories (AQUILA), the European Monitoring and Evaluation Programme (EMEP), the European Research Infrastructure (Aerosol, Clouds and Trace Gases Infrastructure) and its future European Research Infrastructure Consortium (ERIC).

4 Results

Objective 1: Use test aerosol particles in test chambers, to develop traceable methods for the calibration of optical and aerodynamic particle size spectrometers to determine mass and number concentration, as well as size distribution. In addition to characterise aerosol bipolar charge conditioners in MPSS and based on this the pre-normative definition of their performance criteria. Knowing the particle charge distribution as generated by charge conditioners in MPSS is crucial for the accuracy of aerosol size spectroscopy. The knowledge about aerosol charging properties and long-term performance of new non-radioactive bipolar charge conditioners is not sufficient and therefore these devices need to be further investigated. This project has been addressing these two issues. The activities were directly related to the development of the working draft standard WD19996 “Charge conditioning of aerosol particles for particle characterization and the generation of calibration and test aerosols” into an ISO standard within ISO TC24/SC4 Working Group 12 (WG12). This project specifically aims at the development, for the first time, of a robust procedure for testing the aerosol charging performance of soft X-ray charger conditioners (SXR CC). The work has been beyond the current state-of-the art in particular because (i) experimental designs have been used by the project which haven’t ever been investigated before, (ii) the tests covered almost one year of operation, and (iii) competing manufacturers are jointly working with the project in an effort to finally provide public information on key performance features of X-ray-chargers in form of an international standard. Industrial stakeholders have provided 4 commercially available SXR CC to the project. The SXR CC were subjected to two rounds of a long-term performance test, from November 2021 to July 2022 (4300 operating hours), and from March to August 2023 (3200 operating hours), such comprehensive long-term tests over several months have not been conducted before. Before the first round and between the two rounds, the SXR CC were subjected to comparative performance experiments according to ISO 19996.

Currently the calibration of OPSS for particle mass concentration and number size distribution measurement is based on field-based empirical comparisons of aerosol gravimetry with the OPSS to be tested. This approach is both cost and time consuming and lacks the desired level of accuracy, versatility and comparability. This project intended to go beyond the current state-of-the art by developing a traceable and lab-based approach to calibrate OPSS and APSS, using a modified Condensation Particle Counter (CPC) for comparison.

The main part of the detailed works having addressed this objective focused on non-radioactive bipolar charge conditioners (e.g., soft X-ray, corona, plasma) in MPSS, more specifically on the long-term performance of soft X-ray conditioners. The partners conducted a literature review in this area; the compiled literature list was declared as official document N123 by ISO TC24/SC4 working group 12 (WG12). An extensive literature

research was performed to compile relevant scientific knowledge for project partners and stakeholders; the compiled literature list was declared as official document N123 by WG12. During the review, several studies were identified that described experiments using these types of non-radioactive bipolar charge conditioners, in comparison with radioactive charge conditioners. Most of the studies also compared the particle charge distributions produced by the non-radioactive bipolar charge conditioners with theoretically expected equilibrium charge distributions with the aim of demonstrating the practical applicability of non-radioactive bipolar charge conditioners. The results of the project's literature review showed that in general, soft X-ray charge conditioners could reliably and repeatedly achieve a well-defined charge equilibrium. However, the performance of corona charge conditioners is less consistent. These performance differences are mainly due to the physical working principles of the different types of non-radioactive bipolar charge conditioners i.e., both radioactive and soft X-ray charge conditioners expose particles to a true bipolar ion environment, whereas corona chargers alternate between generating positive and negative ions. Plasma charge conditioners are a much newer novel technology, and few studies on these are so far available. For all non-radioactive bipolar charge conditioners, the experiments from the literature review were of limited length (hours or days), only in one case was 15 months of continuous operation of a soft X-ray charge conditioner reported. Therefore, long-term performance studies on non-radioactive bipolar charge conditioners are still needed.

The loan of 4 soft X-ray bipolar charge conditioners (SXR CC) from 3 manufacturers (TSI, PALAS & GRIMM) was successfully negotiated. The instruments were delivered to partner BAM in May 2021 and the manufacturers provided valuable technical support for the experiments. BAM conducted the first series of lab-based performance tests until which resulted in the first round of the long-term test of SXR CC was conducted at partner TROPOS from November 2021 to July 2022, using ambient air and amassing about 4300 operating hours, the equivalent of 180 days. During those tests, the SXR CC and the reference (a state-of-the-art radioactive ⁸⁵Kr charge conditioner) were used in parallel to produce particle mobility distributions of the ambient aerosols; the distributions for negatively charged particles were compared.

Before and after the 1st round of the long-term tests at TROPOS, the SXR CC were subjected to comparative ISO 19996 tests at partner BAM, using monodisperse NaCl-, Sucrose- and Ag test aerosols. The following preliminary results emerged: (i) the chosen test designs are suitable for the comparative investigation of charger performances under load limits such as uncharged or extremely unipolarly charged aerosol particles, and standard loads such as aerosols with bipolar charge distributions do not differ much compared to the well-known equilibrium distribution. (ii) Slight differences in the particle charging characteristics were found amongst the tested devices, however the meaning of this for charging performances and the relevance for data evaluation software requires further discussions.

From March to August 2023, a second round of the long-term experiment was conducted at TROPOS, covering another 3200 operating hours, the equivalent of about 130 days. During the second round, the set-up was temporarily converted from monitoring negatively charged particles (the standard mode for MPSS measurements of particle size distributions) to monitoring positively charged particles. The main conclusion from both rounds of the long-term tests together were (i) that the behaviour is strongly model-dependent on the manufacturer, i.e. It will be necessary to develop individual standards for each SXR CC model. (ii) Significant alteration in performance over time became apparent; these alterations had however not the character of ageing, but rather of a burn-in period: also the 8 months interruption between the two rounds had apparently affected the performance, the alterations in performance were most pronounced during about the first 2000 operating hours of each test round, and the statuses reached at the end of each test round were rather similar.

Partner PTB conducted experiments on the equilibrium charge distribution of mono-disperse soot particles of various diameters within the range 50 to 300 nm, using either radioactive ⁸⁵Kr charge conditioners or SXR CC (a different instrument than the ones used for the long-term experiments at TROPOS was used here). The experiments with both types of charge conditioners showed significant discrepancies between findings and expectations, pointing to the need to review the current standards.

Partner NPL co-supervised a PhD student with the University of Liverpool to develop and test a novel plasma charging source. A Surface Dielectric Barrier Discharge (SDBD) source compared well under preliminary testing to current Kr-85 charging sources. Initial results were promising for a tunable plasma source in the future. The results have been published in the PhD thesis 'Low temperature atmospheric pressure plasma development and use for nanoparticle charging and related applications'.

The second part of the works addressing this objective focused on the development of the portable test aerosol generator that can be used for the calibration of optical and aerodynamic particle size spectrometers. Literature on the state-of-the-art-knowledge for the calibration of OPSS and APSS was collected, and relevant

instruments and characteristics were identified and collated. The findings were used to form a basis for the assessment and development of suitable calibration strategies for OPSS and APSS.

Partner TROPOS focused on the use of a Condensation Particle Counter (CPC) as a reference for OPSS and APSS calibration. The strategy consisted in a modification of a state-of-the-art CPC: by removing any bends in the airflow, particle losses due to impaction were to be reduced. The modified CPC was tested using test particles of various sizes. The desired effect could not be observed, a likely reason for this was however a contamination of the test particles by smaller particles (for which impaction losses are practically irrelevant). Such a contamination cannot be easily avoided and may render this calibration method non-viable as a compact and portable set-up, as envisaged.

Partner NPL has been developing an OPSS calibration service concentrating on high particle number concentrations for polluted ambient measurements and industrial emissions. The final design will use oil aerosols produced by a Vibrating Orifice Aerosol Generator to produce aerosols of variable size, number concentration and density. Delays in purchasing the sampling system and issues getting the required particle number concentrations from the first two particle generation methods tested has meant that the calibration service is still under development at the end of the project.

Partner LNE developed an own calibration set-up, comprising a vortex shaker (manufacturer: ADDAIR) as particle generator, and a mobile FIDAS as reference. In 2022, LNE carried out an interlaboratory comparison: the set-up was deployed at 12 French laboratories and the measurements by the FIDAS were compared to those of the OPSS of each laboratory for 3 different types of test powder samples. Generally, the comparison results were satisfactory. Using the FIDAS as reference only allowed comparing the modal diameter of the test powder particles (the original plan was to deploy also the modified TROPOS CPC within the set-up, this would have allowed comparing also intensities).

The project partners BAM, LNE, NPL, PTB and TROPOS developed new robust and improved calibration procedures for instrumentation such as OPSS and APSS, the instrument manufacturers of which directly benefit from these new capabilities. In addition, aerosol instrument manufacturers benefit from the project's improved knowledge of the performance of radioactive and non-radioactive bipolar charge conditioners including their long-term stability. To support the manufacturers, the project intends to include instrument manufacturers and their devices in the project's tests and comparisons.

4 soft X-ray bipolar charge conditioners from 3 leading manufacturers of aerosol particle size spectrometers (TSI Inc., Grimm GmbH, Palas KG) were subjected to performance tests at TROPOS (two long-term tests covering about 4300 and 3200 operating hours using ambient air) and BAM (test aerosols). The tests revealed important and partly unexpected results which have to be considered in current standardization works. The performance characteristics of non-radioactive charge conditioners turned out to be different to radioactive bipolar charge conditioners. Methods to measure and model the performance parameters were developed and thoroughly tested during the project and were proposed to the corresponding standardisation body. Meanwhile they are part of an international standard allowing manufacturers to adapt their evaluation algorithms. The project achieved the goal of supporting new commercial opportunities for the use of aerosol particle size spectrometers in situations where the use of conventional radioactive charge conditioners is preferred due to statutory regulations or practical considerations.

Objective 2: To develop new calibration procedures for automatic pollen monitoring instruments based on fluorescent polystyrene latex and real pollen particles (at concentrations < 1/cm³, and particle sizes of 0.5 µm up to > 20 µm, with target uncertainties of < 10 %). In addition, comparisons to reference instruments in the field will be used to characterise instrument responses to pollen.

At present, there is no standardisation or available calibration procedures for automatic pollen monitors. This is despite the fact that these instruments are about to be installed at meteorological stations across Europe. This project has been going beyond the state of the art by developing, for the first time, traceable calibration procedures for automatic pollen monitors in the laboratory. The project has also been producing a standardised pollen aerosolisation method and a transfer standard for instrument validation in the field. For the first time automated pollen monitors have been traceably calibrated and the extended calibration capabilities up to 20 µm particles have been validated. The automated pollen monitors have been trained to measure pollen taxa and have been used in the field since 2021. In spring 2022 in Payerne/Switzerland and Oslo/Norway two automated pollen monitors were evaluated by comparing their measured pollen taxa to the reference method, the Hirst-type sampler.

The partner METAS has established methods to produce polystyrene latex (PSL) particles with sizes between 10 µm and 20 µm. The aerosol spatial homogeneity at the sampling location was found to be within 4 % for the PSL particles produced. This was measured using calibrated optical particle size spectrometers, for the larger particle size in collaboration with NMIJ (the Japanese metrology institute).

The automated pollen monitors (Rapid-E, Poleno, WIBS and KH3000) have been calibrated with non-fluorescent and fluorescent PSL particles up to 20 µm at METAS with support by partner EDI. The calibration was successfully performed in the range 0.5-2 cm⁻³. The calibration shows that the instruments could detect particles in the design size range and that the particle counting efficiency was strongly size dependent and, therefore, calibration of the instrument is needed. The new capabilities of the calibration facility at METAS have been successfully validated by comparison with NMIJ. In addition, six more Poleno instruments have been calibrated, which showed considerable unit-to-unit variability.

The partner EDI has developed algorithms to measure the different pollen types and started using the algorithms with automated monitors in the field since summer 2021 and published the data for seven pollen species in almost real time on their web page. EDI together with the collaboration partner NILU have also prepared and run the pollen measurement campaigns in Switzerland and Norway, which run in spring 2022. First results show that the Hirst type sampler and the automated monitors reported similar levels of alder, hazel, and birch pollen. Re-evaluation of the pollen samples by the partner NTUA allowed this comparison at even higher time resolution, which also showed good agreement. The partner PTB assessed different high-resolution techniques of vibrational and x-ray spectroscopy with respect to their discrimination capabilities of different pollen types in order to identify independent validation procedures.

A transfer standard for calibrating pollen monitors in the field has been designed and the components for building it have been selected. The design is based on the commercially available Swisens atomiser, with a custom-made downward flow mixing tube and includes a TSI optical particle size spectrometer or a Grimm 11D, which have been calibrated at METAS. The design has been tested with limited resources at METAS and uncertainties were around 50 %.

The project's work on pollen monitoring developed new metrological tools for manufacturers to (i) characterise the performance of such monitors, (ii) detect malfunctions and (iii) improve the measurement efficiency of their instruments. This supports the availability of new improved pollen monitoring products in the European market, particularly as three of the major players in this field are based in Europe. Three automated pollen monitors (Rapid-E, Plair SA, Switzerland, Poleno, Swisens, Switzerland, WIBS, Droplet Measurement Tech., US) were calibrated with non-fluorescent and fluorescent PSL particles up to 20 µm at the partner site of METAS with support by partner EDI. The manufacturers have shown interest in the project's results and discussions with them have begun. Meteoswiss has ordered more calibrations of their monitors for 2024. Swisen, Switzerland, has launched a project together with METAS to extend calibration capabilities for particles up to 80 µm. EDI together with the collaborator NILU have also prepared and run the pollen measurement campaigns in Switzerland and Norway, which run in spring 2022. The results show that the Hirst-type sampler and the automated monitors reported similar levels of alder, hazel and birch pollen and that the laboratory calibration of the automated pollen monitors performed at METAS are in line with previous published collection efficiencies of the Hirst-type impactor.

Objective 3: To develop certified reference substrates and traceable measurement techniques for the quantification of regulated and unregulated substances in ambient air with cascade impactor sampling and XRF

Benchtop-Total Reflection X-ray Fluorescence (TXRF) spectrometers have been calibrated using reference standards. 11 sets of reference samples for TXRF analysis, using lithography-based techniques (parylene mask, direct laser writing, lift-off), diblock-copolymer, nk jet techniques and microspotter printing of a 5-element standard ink by partners EK, INRIM, POLITO and BAM. Full traceability to primary standards, such as a synchrotron based GIXRF setup could be achieved. Sets of reference calibration samples have been produced and initial laboratory- and synchrotron-radiation-based characterization measurements have been performed. The effect of calibration on accuracy and comparability to reference-free SR-TXRF was demonstrated by measurements at facilities of PTB and CEA, using reference sample sets produced during the project. An agreement in the order of 20% could be achieved. This is a beyond state-of-the-art progress as now tools can be provided to correct raw data from TXRF/GIXRF measurements based on a new methodology not yet implemented in the original manufacturer's software. In this context, scanning XRF (GIXRF, i.e., variation of the incident beam angle) also in benchtop instruments was considered necessary in order to better adapt

quantification methods to sample characteristics. The realization of the GIXRF method in benchtop spectrometers could be achieved and is beyond state of the art because this measurement option was originally not available. While the bench top instruments provide automated positioning optimization along the beam axis experiments revealed the necessity to optimize the sample position of cascade impactor samples in two dimensions in order to improve accuracy and repeatability and a specialized SOP along with a specialized set of reference samples with perfect circular deposition geometry was developed for this purpose using existing maintenance options of the spectrometers.

Reference samples should have properties of real environmental samples with respect to elemental composition, concentration, and spatial structure of the deposit. This issue was intensively discussed in the AEROMET II project. This led to the production of a set of reference samples which contain the test candidate element (Ti) diffused at different quantities into a 3D structure made of organic material. The samples have already been successfully used for comparative measurements.

Three promising deposition techniques (i. e. parylene mask, atomic layer and lift-off) have been tested successfully with Ti and Cr as deposited elements. These 3 techniques are based on lithography and physical vapor deposition and were chosen as candidates for the preparation of cascade impactor reference samples due to their suitability for (i) good control of deposited material quantities, (ii) flexibility in deposited materials, and (iii) flexibility in lateral distribution and shape of deposited material layers or dots arrangements. The sets of reference samples manufactured to date are:

- Set 1. Thin Ti film samples with varying thicknesses on square Si wafers, prepared with atomic layer deposition
- Set 2. 13 Dekati stage patterns (80 nm Ti dots arrangements) using parylene mask/atomic layer deposition
- Set 3. Sample production completely failed due to a technical malfunction.
- Set 4. Random Cr dots arrangements with varying thicknesses) on Si wafers using lift-off deposition forming a May-type deposition pattern (stripe). s
- Set 5. (Random Cr dots arrangements with varying thicknesses) on Si wafers using lift-off deposition and forming Dekati-type deposition patterns
- Set 6. Dekati stage 5 patterns (Ti dots arrangement, 3 thicknesses) using lift-off deposition on acrylic discs
- Set 7. Ti patterns (Dekati stages 2 and 4) and films on organic spacers on quartz substrates
- Set 8. Al₂O₃ and TiO₂ infiltration-in-block-copolymers nanoscale templates using atomic layer deposition on square Si wafers for GIXRF. Dekati stage 5 patterns and block structure
- Set 9. Dekati stage 5 patterns (Cr, dots arrangement, two thicknesses) using lift-off deposition on round Si wafers
- Set 10. Cr-circular patterns (rings) on round Si wafers, prepared with lift-off technique
- Set 11. Inkjet printing of Y on acrylic discs / Dekati patterns 2x stage 5, 1x stage 4
- Set 11_2. This set consists of reference samples printed with BAM's micro spotter array using a 5-element standard aqueous ink, which was analysed by NTUA.
- Set 12. Cr dots arranged as first stage pattern of newly developed Bruker impactor

Benchtop TXRF- and GIXRF investigations on the sample sets led to the production of an SOP for the proper adjustment and positioning of Dekati cascade impactor samples when using a S4-TStar TXRF spectrometer. Initial results have also been published in: <https://doi.org/10.1016/j.aca.2021.339367> 'Reliable compositional analysis of airborne particulate matter beyond the quantification limits of total reflection X-ray fluorescence', Anal. Chem Act. 2022. A preliminary method for the calibration of benchtop TXRF instruments was developed and applied. Based on standard film reference samples for SR-TXRF and Set 7 reference samples the calibration of BAM's desktop TXRF spectrometer resulted in an uncertainty of 20 %. Reference-free measurements of Set 9 at the CEA facility corroborated the quantities predicted from preparation by partner EK within less than 15%. Project partners intend to continue comparative measurement of different sets of reference samples with SR- and benchtop TXRF and GIXRF setups. The project results motivated partner

Bruker to develop a novel commercial cascade impactor with deposition patterns optimized for benchtop TXRF instruments. Preliminary laboratory and field tests of this impactor and TXRF measurements were successful. Using the technique developed by EK for Set 9 a first set of calibration reference samples were prepared for this new impactor.

A method for the calibrate benchtop TXRF spectrometers for the analysis of cascade impactor aerosol samples was developed and applied successfully. The method was cross-validated by parallel measurements of reference samples at two SR-TXRF/GIXRF facilities of the partners CEA and PTB. The project provided methods for the harmonised and traceable calibration of TXRF spectrometers with reference samples produced by using the techniques developed in the project by the partners INRiM and EK. The methods are fully adoptable to new cascade impactor designs and hence are providing high flexibility in the combination of elements as well as deposition patterns in reference samples. In effect, individual end user requirements, e.g., for the measurement of specific industrial air pollutants or background aerosols can be met. Repeated measurements of reference samples with a mobile S4-TStar TXRF device using a Dekati cascade impactor deposition patterns demonstrated the need for an SOP on the proper positioning of samples with laterally extended deposition. Using the project's SOP, positioning with low tolerances could be achieved which led to acceptable repeatability of TXRF measurements. The SOP has been integrated into the project's quality assurance scheme and is made available to end users on the project's website. The SI traceable elemental analysis of aerosols by means of X-ray spectrometry under grazing incidence has been demonstrated by the partner PTB. Based on standard film reference samples for SR-TXRF and Set 7 reference samples the calibration of BAM's desktop TXRF spectrometer resulted in an uncertainty of 20 %. Additionally reference sample sets have been successfully used for a standardized sample positioning method in benchtop TXRF devices as well as for the determination of correctional factors for different deposition patterns.

Objective 4: To develop portable instruments and software for the measurement of ambient aerosol particle concentrations under variable environmental conditions. Knowledge gained is to be applied to other appropriate classes of portable instruments by the production of suitably adapted procedures.

Portable, commercially available instruments for the measurement of ambient aerosol particle concentrations are popular because of their potential easy handling and rather low acquisition costs. However, they are associated with less defined uncertainties as well as signal instabilities compared to more sophisticated laboratory-based instrumentation. The project will undertake a comparison of the portable, commercially available instruments to calibrated reference instruments under laboratory conditions, as well as field studies under significant different environmental conditions. This will be the first step in quantifying the effects of a variety of environmental conditions, e.g., high humidity, on the accuracy of the portable, commercially available instruments. The project is also going beyond the current state of the art by developing novel compensation algorithm procedures for the use of the portable, commercially available instruments, that consider the effects of the environmental conditions and apply relevant measurement principles.

Real-time online measurements of black carbon (BC) with portable optical instruments are also facing similar measurement quality issues related to inappropriate guidelines and the lack of traceable calibration methods. These BC instruments are currently not a mandatory component of air quality monitoring stations, therefore periodic calibration does not happen on a regular basis. This project is going beyond the current state of the art by improving the current limited knowledge about reliability and accuracy in BC measurements in different environmental scenarios by researching and developing compensation algorithms that can cancel out measurement artefacts and enhance the instrument's reproducibility.

Both portable particle counters and BC instruments have been characterised with respect to relevant key parameters such as counting efficiency, precision, and long-term reliability. The first two intercomparison campaigns have been performed in laboratory settings and are the most complete intercomparison studies conducted so far, as they involve all commercially available types of portable instruments. In addition, the most comprehensive outdoor intercomparison campaign have been carried out during 2022 with all available types of portable particle counters and BC instruments. This intercomparison campaign includes co-location of portable instruments at three different outdoor reference air quality measurement stations, as well as measurement campaigns at state-of-the-art test facilities, more specifically wind-tunnel at IRSN and wood-stove laboratory at DTI. The environmental conditions campaigns during these intercomparison measurement are representative for the majority of European climate, e.g., from sub-zero temperatures in Sweden during

winter to high temperatures in Greece during the summer. The large range of environmental conditions entails progress beyond the current state of the art.

The laboratory intercomparison: Seven types of portable particle counters were fully tested according to their counting efficiency, precision and long-term reliability against ambient aerosols with respect to state-of-the-art reference particle counting equipment at TROPOS. The 7 types of particle counters were (i) TSI P-Trak 8525, (ii) TSI CPC 3007, (iii) Brechtel MCPC 1720, (iv) Aerosol Devices MAGIC CPC 210, (v) Testo DiscMini, (vi) Nanoes Partector, and (vii) Oxilix NanoTracer and thus represent the current commercially available portable counters, which are used in both industrial and end-user communities. The test included multiple units of most of the different types of portable particle counters. The portable particle counters were provided by project partners LUND, RISE, AU, DTI, UNICAS and CIEMAT and delivered to TROPOS for their testing.

The results from the testing of the portable particle counters showed a linearity (value of slope in linear fit) in particle counts in the range from 0.4 and 1.1 when comparing with a TSI 3772 CPC which was chosen as reference device by the project partners, as this instrument is considered state-of-the-art, and currently there is no standardised reference instrument for particle counting instruments. The results from the testing also showed a large variety in counting efficiency between similar types of instruments, and thus highlighting the need for moving beyond the current state of these types of portable particle counters.

In addition, the correlation (R^2 value – square of the Pearson correlation coefficient) in particle counts between the 7 tested types of portable particle counters devices and the reference instrument was in the range 0.579 to 0.998. This result shows that the only a few types of the tested portable particle counters exhibit a test performance that are within the goals of the project, i.e., to reach the overall uncertainty target of 15 % when compared to reference equipment.

In similar work, 4 different types of portable BC instruments were tested in a laboratory intercomparison at LNE according to their efficiency, precision and reliability measured. The 4 types of portable BC instruments were tested in a dedicated calibration setup and compared to several state-of-the-art reference BC equipment (Magee Scientific AE 33), which was provided by partner DTI and stakeholders Airparif and Ecomesure. The intercomparison was performed in various environmental conditions (temperature and %RH). The 4 types of portable BC particle counters were: (i) Aehtlabs AE51, (ii) Aethlabs MA200, (iii) Aethlabs MA300 and, (iv) DSTech Observair and thus represent the majority of commercially available BC instruments used in both industrial and end-user communities. The intercomparison testing included multiple units of the different types of portable BC particle counters (9 portable instruments in total). The BC instruments were provided by partners LUND, NCSR Demokritos, DTI, UNICAS and CIEMAT and delivered to LNE for testing.

The results from the testing of the portable BC instruments were grouped into two groups: AE51's and MA's. The AE51's showed a linearity (value of slope in linear fit) in BC concentration in the range from 0.28 and 0.82 when comparing with an AE33 (Magee Scientific) which was chosen as a state-of-the-art reference device by the project partners. Hence, there was an underestimation of BC concentration for all AE51 when compared to AE33 in all environmental conditions. For the MA's the linearity when compared to AE33 were in the range from 0.78 to 1.18, thus showing a better agreement with the reference instruments than the AE51s. However, the data from the MAs were significantly noisier. These results show a large variety in precision and efficiency, and thus highlight the need for improvements to current portable BC instruments.

In addition, the correlation (R^2 value – square of the Pearson correlation coefficient) in BC concentration between the 4 tested types of portable BC devices and the reference instrument was in the range 0.61 to 0.99. This result shows that only a few of the tested portable BC instruments under some environmental conditions exhibit a test performance that are within the goals of the project, i.e., to reach the overall uncertainty target of 15 % when compared to reference equipment. Further data analysis is ongoing, in addition to developing compensation algorithms for parameters like filter loading effect correction, flowrate correction due to temperature and mass adsorption cross section.

Outdoor Ambient Intercomparison: In January 2022, the first outdoor intercomparison campaign was conducted; outdoors in wintery conditions in Southern Sweden (Hyltemossa Research Station, Perstorp, Skåne). Specialised weatherproofed box was designed by partner DTI in order to house the instruments during the different campaigns. 12 portable particle counters, 2 reference particle counters, 8 portable BC counters and 8 reference BC instruments were provided by the partners DTI, LNE, RISE, AU, CIEMAT, LUND, NCSR Demokritos, TROPOS and UNICAS. Data was collected for 14 days in total during this campaign.

In April 2022, the second outdoor intercomparison campaign was conducted in mild conditions in Leipzig Germany (German GUAN monitoring site "TROPOS"). A total of 20 portable instruments (13 particle counter

and 7 BC instruments) were tested during this field study. Instruments were provided by the partners DTI, LUND, RISE, NCSR, UNICAS, CIEMAT, AU, LNE.

All portable instruments were placed in two weatherproof boxes on the roof of the building, next to the inlets of the GUAN station. Exception: MCPC and Magic CPC (which both require connection to an external vacuum pump) were located inside the building and supplied by air from the GUAN inlets. Data was collected for 12 days in total during this campaign.

In July 2022, the third outdoor intercomparison campaign was conducted in summery conditions in Athens Greece (Greek GAW monitoring site). A total of 16 portable instruments (9 particle counter and 7 BC instruments) were tested during this field study. Instruments were provided by the partners DTI, LUND, RISE, NCSR, UNICAS, CIEMAT, AU, LNE. All portable instruments were placed in two weatherproof boxes on the roof of the building, next to the inlets of the GAW station. Data was collected for 17 days in total during this campaign.

In addition, a study of temperature and wind influence, as well as humidity, in an insulated controlled wind tunnel using outdoor atmosphere was conducted at IRSN in May 2022. A total of 14 portable instruments (6 BC instruments and 8 particle counters) were tested during this campaign. Instruments were provided by DTI, LUND, NCSR Demokritos, UNICAS, CIEMAT, and AU. Reference instruments were provided by AirParif (the observatory of air quality for Paris region) and TROPOS and IRSN. All portable instruments, except Magic CPC, have been put inside the wind tunnel and compared to the reference instruments, which was placed outside. The experimental protocols of the wind tunnel condition have been decided by Tropos, IRSN and IJS. For each test, the wind tunnel has been filtered from any initial particle and the filters of the AE51 have been changed.

Concerning the influence of the wind velocity, which is not expected to play an important role for the typical sub-micronic sizes of the atmospheric aerosols, five velocities have been considered at constant humidity and temperature. Concerning the influence of the couple ‘temperature-humidity’ at a given constant wind velocity, efforts have been put to reach conditions so that one of both parameters remains constant (3 to 8 different temperatures for one range of relative humidities). Steady-states of at least two hours for four ranges of conditions have been obtained: between 10-20°C at very high relative humidity RH, between 10-20°C at high RH, between 17-30°C at intermediate RH, and between 27-37°C under lower RH.

The last intercomparison study was conducted for portable BC instruments in a state-of-the-art wood stove test setup at DTI. The study was conducted with 2 types of portable BC instruments and a reference aethalometer. All instruments were provided by DTI.

The effect of temperature and relative humidity on portable particle counters was analysed considering the campaigns at outdoor sampling sites and in the wind tunnel. To this end, an intensive data preparation was conducted (including raw files collection, reading into the common database, removing “bad data”, applying flow and coincidence corrections, considering the time shifting, and, finally, applying a time resampling to the common time-scale). Then, per each instrument, the output was compared to the reference instrument output for all campaigns in the same conditions, i.e. for narrow T/RH range, and the calibration slopes were determined. Temperature and RH conditions covered in the experimental campaigns carried out at Hyltemossa, Leipzig, Athens and IRSN campaigns represents a broad range of temperatures, 0 - 40°C and RH, 20 - 95 % and even more a few data points at extreme T, sub-zero and above 50 °C, and RH close to 100% values were collected. The results of this analysis showed that only few instruments show consistent T/RH dependence, in other words, we recognized that the same instrument model may present different behaviors in different campaigns even for the same T/RH range. The analysis clearly show that no clear trends of the correlation (slope) between portable and reference instruments as a function of T, RH and water content can be obtained; thus, the effect of meteo-climatic conditions is maybe less important than particle composition, size distribution, counting efficiency ones.

Compensation algorithms for BC monitors were developed considering portable aethalometer data from Athens, Paris and Hyltemossa campaigns. In these campaigns the AE33 aethalometer was the reference instrument. First of all, outliers were removed and flow correction was applied from AE33, AE51, and MA200/MA300 collected data including the following steps:

1. we calculated the 120 minutes running mean value for black carbon concentration for the instruments AE33 (BC_{AE33}), AE51 (BC_{AE51}), and MA200/MA300 (BC_{MA200}) and the standard deviation for BC_{AE33} , BC_{AE51} , BC_{MA200} within these 120 minutes. Then, we removed the values laying outside the interval of the running mean ± 3 standard deviations;

2. when consecutive 1-min BC concentrations (BC_{AE33} , BC_{AE51} , BC_{MA200}) had a difference larger than 500 ng/m^3 , it was considered a spike value and it was removed;
3. when negative RH values were encountered, they were removed. For AE33, MA200/MA300, negative attenuation values were considered as invalid. For AE51 attenuation values below 1 were not considered;
4. black carbon values below 1 ng/m^3 were removed (BC_{AE33} , BC_{AE51} , $BC_{MA200} < 1 \text{ ng/m}^3$);
5. black carbon values above 5000 ng/m^3 were removed (BC_{AE33} , BC_{AE51} , $BC_{MA200} > 5000 \text{ ng/m}^3$);
6. visual inspection (very few values removed).

To those data the Virkkula correction with a k factor equal to 0.005 was applied (Alas et al., 2020; Virkkula et al., 2007) according to the following equations:

$$BC_{AE51-COR} = BC_{AE51} \times (1 + k \times Atn_{AE51}) \quad (1)$$

$$BC_{MA200-COR} = BC_{MA200} \times (1 + k \times Atn_{MA200}) \quad (2)$$

In order to assess if including ambient RH or temperature would improve the slope even more, the dataset was divided in quartiles. The first quartile contained all values with RH or Temperature up to the 25th percentile, the second quartile contained values with RH or Temperature from the 25th to the 50th percentile, etc. The slopes of the relation between BC_{AE33} and BC_{MA200} as well as BC_{AE33} and BC_{AE51} did not increase or decrease monotonically as we pass from one quartile to the next, probably indicating that there is no simple relation connecting the slope and RH and/or Temperature.

The project aimed at verifying the potential improvements of the selected portable particle counters and BC portable instruments carried out in previous tasks of the project with a larger field campaign conducted at an European monitoring site with high occurrence of UFP and BC.

The partners agreed to conduct the campaign in Cassino (Italy) in December 2022 for one week. The sampling site proposed by UNICAS is the Department of Civil and Mechanical Engineering which is located in the urban area of Cassino (FR, Italy). In particular, the sampling site was a covered balcony at the 2nd floor of the building (Figure 13). It is quite close to a road with a traffic density of approximately 24 vehicles/min with a mean velocity of about 30–40 km/h.

On the basis of the results obtained in the different experimental campaign the following conclusions can be summarized:

- Portable particle counters hardly were able to provide measurement data within the $\pm 15\%$ range of the reference CPC ones, with diffusion chargers, and in particular Nanotracers, showing the worst metrological performances.
- Tests performed in the wind tunnel demonstrated that the wind velocity does not have an influence on portable particle counters.
- Tests performed at different (controlled) temperature and relative humidity conditions revealed that most particle counters generally yielded slopes < 1 and the slopes appeared to increase linearly with increasing RH (and decreasing temperature) then approaching 1 for high RH (& low temperature). Moreover, such an effect (if any) was clearly recognized in tests performed in a wind tunnel where temperature and RH conditions were controlled and remained constant for a long period, on the contrary, this effect was not recognized in tests conducted at outdoor sampling sites. Summarizing, the effect of temperature and relative humidity on portable particle counters is not easily recognizable and, anyway, less evident than other influencing parameters (measurement range, instrument drift, particle size distribution).
- Most of the BC monitors yielded slopes < 1 , nonetheless, the slopes improved if the loading correction of the aethalometer (Virkkula correction) is applied.
- The effect of temperature and relative humidity on BC monitors is not easily recognizable as well, and, anyway, less evident than loading correction of the aethalometer (Virkkula correction). Nonetheless, for very high RH values ($> 70\%$) a worsening of the correlation between MA200s and AE33 was detected; on the contrary, no significant effect was recognized for AE51s.
- The highest uncertainty for both AE51s and MA200s measurements was observed at Hyltemossa winter campaign, where the lowest temperatures and highest RH values were observed. It should be noted that Hyltemossa is a remote station in a forest exhibiting very low BC values.

- The uncertainty of both portable BC monitors and particle counters, even considering just the Type A contributions and a coverage factor $k=1$, resulted well above the $\pm 15\%$ range.

The use, by non-experts, of portable, commercial instruments such as monitors for BC and particle number concentration for ambient aerosol measurements, has meant that manufacturers of such instruments urgently needed to develop robust and easy to follow technical user guidance. This project supported the development of such guidance with the interpretation of data from non-expert users. The analysis is to be used to provide end users and manufacturers with a comprehensive description of application limits such as environmental conditions and margins of uncertainty. The project aimed to highlight the potential use of portable particle counters for ambient aerosol measurements under varying environmental conditions (from indoor conditions to extreme summer and winter conditions). The tested devices comprised all types of portable particle counters that are currently commercially available. Already the results of the first intercomparison campaign allowed clear statements regarding the achievable accuracy. These results may strongly influence the user's choice (which for portable devices usually is a compromise between accuracy and simplicity & price). It will also assist users in the correct interpretation of results obtained using these types of instruments.

5 Impact

The impact activities of the AEROMET II project involved the dissemination of information to end user communities via stakeholder meetings, conference contributions, dedicated sessions and workshop organisations, training activities as well as input provision to standardisation committees. The project has put on display the respective information on its dissemination progress to end users via a website at <http://www.aerometprojectii.com/>.

In addition, three presentations were given at the E-MRS 2021 Spring Meeting together with the organisation of the symposium Analytical techniques for precise characterisation of nano materials (ALTECH 2021) by the partners PTB, CEA, INRiM and NPL. Two presentations were also given at the online conference Open Alt 2021 in November 2021 on 'Calibration of a DIY Low-cost Aerosol Detector and metrology of aerosol particles' and on progress of the AEROMET II project. In addition, partner NTUA organised the 12th International Conference on Instrument Analysis: Modern Trends and Applications (IMA) 2021 which included a dedicated session to AEROMET II containing 8 presentations and a poster from partners.

The partners CEA and PTB organised two workshops on X-ray fundamental parameters <https://www.exsa.hu/fpi.php>. As well as one-to-one web-conferencing training for the consortium on pollen counting method according to CEN EN 16868:2019. Partner CMI also organised a workshop in September 2021 on, 'Numerical modelling for electromagnetic field interaction with aerosol particles and on real-world optical measurements of aerosol particles'.

A Masters thesis at partner EK (Budapest University of Technology and Economics) was produced on 'Application of X-ray spectrometric methods for characterisation of aerosol particles from urban environment

In 2022 the main impact event was the International Aerosol Conference (IAC2022) in Athens. The findings from and the impact of the AEROMET II project were disseminated by:

- A training session (hosted by N.C.S.R. Demokritos) in six different fields, for example aerosol analysis by traceable x-ray spectrometry - by B. Beckhoff (PTB); Benefits and issues of aerosol analysis by TXRF – by Armin Gross (Bruker Nano)
- A dedicated IAC2022 session on AEROMET II containing six oral presentations by different project partners on topics ranging from traceable calibration of real-time bioaerosol particle counters (K. Vasilatou (METAS)) to potential reference samples for TXRF elemental analysis of aerosol particles (J. Osan (EK))
- Nine poster presentations at IAC2022 on, for example, qualification of portable instruments for ambient aerosol measurements (S. Koust, DTI) and intercomparison of stationary and portable soot instruments (M. Spanne (Malmö Stad))

Other than this flagship event, the project gave four presentations at the Hungarian Aerosol Conference ; “Capital of Knowledge” (EAPE 2022), “*Traceable chemical analysis of aerosols by reference-free X-ray spectrometry*” and “*TXRF to trace quick and seasonal changes in elemental size distribution of airborne particulate matter*” (EXRS 2022), “Identification of urban aerosol sources using combination of size-fractionated sampling and X-ray spectrometry methods”

A separate training event “*Apprenticeship - building a movable sensor for PM profile determination at DIESE facility*” in July 2022 was a 3 weeks one to one training of an IRSN student (M. Radosavljevic) at JSI (K. Bucar).

Finally in 2022, a poster was presented at the Air Quality and Emissions Show (Telford, UK): “*An Investigation by NPL into the Airborne Particle Removal Efficiency of a Commercial Extraction System in a Dental Surgery*”

The project organised 12 events in 2023, including a training webinar on the realization of reference materials and on the *X-ray spectrometry methods relevant for aerosol metrology* which was organised by PTB, CEA-LNHB and INRiM and a series of lectures and hands-on training on the *theory and practice of aerosol chemistry and engineering for climate, air quality, emissions and health effects* which was organised by NCSR and TROPOS. A training session for municipalities and public bodies on *Black Carbon (Soot) Air Pollution – Measurement, Sources, Fate, and Regulation* was organised by RISE and LUND and TROPOS organised a training course under the ACTRIS framework on *ACTRIS CAIS-ECAC Aerosol In-Situ Course: Fundamentals & Methods for Aerosol Sampling and Physical & Chemical In-Situ Measurements*. CEA organised a training session in French titled *Caractérisation d'échantillons de référence pour la mesure des aérosols par fluorescence X en réflexion totale* and a poster on the *validation of a voltammetric approach for determination of mercury in airborne particulate matter using screen- printed electrodes* was presented by NTUA. The experimental work from this project was used to support the ISO standard ISO 19996 (ISO/TC 24/SC 4/WG 12) and the findings were presented at the 26th ETH Nanoparticle Conference, multiple AEROMET II partners presented the findings from WP1 at EAC 2023 in a presentation titled *1-year performance test of Soft X-ray charge conditioners*. TSI GMBH carried out a stakeholder meeting and workshop called *Challenges in Aerosol Metrology- Perspectives from Manufacturers and Application* and a workshop titled *Aerosol instrument calibration procedures (CPC, MPSS, OPSS, APSS, BC-monitors)* was run by TROPOS. NPL delivered a presentation *Perspectives for Aerosol Metrology* to AEROMET II stakeholders and finally NPL also conducted a TEAMS meeting to inform the Environment Agency and the Source Testing Association in the UK of the *OPSS Calibration Facility for Industrial Emissions*.

Impact on industrial and other user communities

The project provided input to industrial and related communities by means of a direct involvement of instrument manufacturers in the characterisation and calibration works performed. The industrial interest was reflected in joint discussions with project partners on the results achieved as well as in general discussions at stakeholder events. The characterisation and calibration results are relevant for companies and their customers to identify the respective ranges of reliable applicability of different instruments for the purpose of aerosol metrology.

This project developed new robust and improved calibration procedures for instrumentation such as OPSS and APSS. OPSS and APSS instrument manufacturers will directly benefit from these new capabilities. In addition, aerosol instrument manufactures benefited from the project's improved knowledge of the performance of radioactive and non-radioactive bipolar charge conditioners including their long-term stability. To support the impact on manufacturers, the project included instrument manufacturers and their devices in the project's tests and comparisons. Manufacturers were included in the project's stakeholder committee, so that feedback from them can be included in the project.

Important industrial partners, such as TSI Inc., Grimm GmbH, Palas KG, Airmodus Ltd., who are leading manufacturers of aerosol particle size spectrometers, joined the project's stakeholder committee. The performance characteristics of non-radioactive charge conditioners turned out to be different to radioactive bipolar charge conditioners. Methods to measure and model the performance parameters were developed and thoroughly tested during the project and were proposed to the ISO TC24/SC4 standardisation body. Meanwhile they are part of an international standard (ISO/DIS 19996:2023). With this standard, manufacturers are able to adapt their evaluation algorithms. The project achieved the goal of supporting new commercial opportunities for the use of aerosol particle size spectrometers in situations where the use of conventional radioactive charge conditioners is preferred due to statutory regulations or practical considerations.

The project's work on pollen monitoring developed new metrological tools for manufacturers to (i) characterise the performance of such monitors, (ii) detect malfunctions and (iii) improve the measurement efficiency of their instruments. This will support the availability of new improved pollen monitoring products in the European

market, particularly as three of the major players in this field are based in Europe. So far 3 automated pollen monitors (Rapid-E, Plair SA, Switzerland, Poleno, Swisens, Switzerland, WIBS, Droplet Measurement Tech., US) were independently calibrated for the first time ever (objective 2). The manufacturers showed interest in the project's results and discussions with them have begun. Meteoswiss has ordered more calibrations of their monitors for 2024. Swisen, Switzerland, has launched a project together with METAS to extend calibration capabilities for particles up to 80 μm .

A method for the calibration of benchtop TXRF spectrometers for the analysis of cascade impactor aerosol samples was developed and applied successfully. The method was cross-validated by parallel measurements of reference samples at two SR-TXRF/GIXRF facilities. The project results motivated one manufacturer of benchtop TXRF spectrometers to start development of an impactor design with optimized adoption to the TXRF requirements. The project provided methods for the harmonised and traceable calibration of TXRF spectrometers with reference samples produced by using the techniques developed in the project. The methods are fully adoptable to new cascade impactor designs and hence are providing high flexibility in the combination of elements as well as deposition patterns in reference samples. In effect, individual end user requirements, e.g., for the measurement of specific industrial air pollutants or background aerosols can be met. Repeated measurements of reference samples with a mobile S4-TStar TXRF device using Dekati cascade impactor deposition patterns demonstrated the need for an SOP on the proper positioning of samples with laterally extended deposition. Using the project's SOP, positioning with low tolerances could be achieved which led to acceptable repeatability of TXRF measurements. The SOP has been integrated into the project's quality assurance scheme and is available to end users on the project's website.

The use, by non-experts, of portable, commercial instruments such as monitors for BC and particle number concentration for ambient aerosol measurements, has meant that manufacturers of such instruments urgently need to develop robust and easy to follow technical user guidance. This project supported the development of such guidance with the interpretation of data from non-expert users. The analysis was used to provide end users and manufacturers with a comprehensive description of application limits such as environmental conditions and margins of uncertainty. The project also aims to highlight the potential use of portable particle counters for ambient aerosol measurements under varying environmental conditions (from indoor conditions to extreme summer and winter conditions). The tested devices comprised all types of portable particle counters that are currently commercially available. Already the results of the first intercomparison campaign allow clear statements regarding the achievable accuracy. These results may strongly influence the user's choice (which for portable devices usually is a compromise between accuracy and simplicity & price). It will also assist users in the correct interpretation of results obtained using these types of instruments (objective 4).

Impact on the metrology and scientific communities

The project closely coworked with academic and industrial collaborators or stakeholders multiplying the project's impact to the metrology and scientific communities. Here the extension of calibration ranges as well as the validation of applicability regimes is equally interesting for academic and industrial purposes. The project's new and improved calibration procedures for OPSS and APSS: with (i) an extended traceable size range, (ii) knowledge of the performance of non-radioactive bipolar charge conditioners and (iii) their long-term stability for MPSS measurements will be disseminated to the European Research Infrastructure ACTRIS. Thus, the project's results was promoted to ACTRIS' stakeholders and supported ACTRIS and its central facility (i.e., Topical Centre for Aerosol In-Situ Measurements) within the European Research Infrastructure Consortium (ERIC). Overall, a better understanding of the performance characteristics of non-radioactive charge conditioners supported new scientific opportunities and accurate measurements of particle number size distributions.

The project's development of calibration procedures for automatic pollen monitors enabled meteorological stations to automate their monitoring stations and in turn improved data comparability in Europe. This resulted in accurate, real-time data becoming readily available while at the same time reducing operational costs (compared to the current time-consuming, manual methods for monitoring pollen). Moreover, the methods developed for pollen monitors could be used in future applications for the detection of other bioaerosols, such as bacterial and fungal spores, many of which are human and plant pathogens. Traceable calibration of these new pollen monitors also helped to continue long term data series and supports their relevance for climate sciences.

In atmospheric research and air quality networks the project supported the availability of traceable reference samples. This supported traceable and validated measurements and add value to the use of cascade impactor

sampling and TXRF spectroscopy for element mass concentration measurements in ambient air. Indeed, the project's first results supported the development of methods for the quantification of air pollutant by cascade impactor sampling and TXRF as an alternative method. The project reached the goal to demonstrate production, characterization and application of fully traceable standards/references for the combination of aerosol sampling by cascade impactors and TXRF/GIXRF analysis in the same way as the already established methods for chemical aerosol analysis (i.e. filter sampling and AAR or ICP-MS) (objective 3).

In the scientific aerosol community, portable instruments are widely used as an important supplement to laboratory-based reference equipment. The project's qualification of portable instruments for measurements of the particle number concentration and BC will, therefore, be of great importance for researchers in the air quality network community (objective 4). The devices tested by the project include all currently commercially available portable instrument types (i.e. both particle counters and BC). The results of the intercomparison campaigns have been used to determine the ability of each device to achieve an overall uncertainty < 15 %, which is generally considered as a minimum requirement by the scientific community.

Impact on relevant standards

Project partners are active in several standardisation committees where also many industrial collaborators or stakeholders are present. The methodological developments aiming at new standards are being driven from NMIs and companies. Partners of the consortium create impact on relevant standardisation bodies such as: ISO TC201 SC10 X-ray Reflectometry (XRR) and X-ray Fluorescence (XRF) Analysis including its German mirror committee DIN NA062-08-16AA, EURAMET TC-MC SC Gas Analysis, BIPM CCQM WG Gas Analysis and WG Surface Analysis, CEN/TC 264 WG 32 - Ambient air - Particle number concentration, CEN/TC 264 WG 39 - Ambient air - Airborne pollen grains and fungal spores, CEN/TC 264 WG 42 - Low-cost sensors, ISO/TC 24 SC4 WG 12 - Electrical mobility and number concentration analysis for aerosol particles and ISO/TC 24 SC4 WG 9 - Single particle light interaction methods.

The project's work on the calibration of OPSS and APSS will impact standardisation activities within CEN/TC 264 WGs 32, 39 and 42. The performance characteristics of non-radioactive charge conditioners was disseminated to ISO/TC 24 SC4 WG 12 for inclusion into the meanwhile published standard ISO/DIS 19996:2023 - Charge conditioning of aerosol particles for particle characterisation and the generation of calibration and test aerosols. The project researched and provided a literature review on the performance of non-radioactive bipolar charge conditioners (soft X-ray, corona, plasma) in MPSS (Objective 1) as well as performance characterisation methods for soft-X-ray charge conditioners in Annex B of the above standard. .

In addition, partners METAS and EDI are working within CEN TC 264/WG39 on the development of a new standard on automatic bioaerosol monitors entitled: "Ambient air - near real time measurement methods for monitoring airborne pollen and fungal spores concentrations". Results generated within (Objective 2) have already been incorporated into the draft standard. The project's harmonised calibration guides for TXRF spectrometers may also lead to a new work item proposal within ISO/TC 24 SC4 or ISO/TC 201. Currently, at ISO TC201 SC10 a recently revised German national norm for TXRF and a draft technical specification for XRF under grazing incidence conditions relevant for reliable aerosol compositional analysis are being discussed. Furthermore, the work on portable aerosol particle instrumentation will support a future CEN Technical Specification (TS) on the performance evaluation of sensors for the determination of concentrations of particulate matter (PM) in ambient air (CEN TC 264).

Longer-term economic, social and environmental impacts

This project will make significant improvements to air quality data by reducing the measurement uncertainties for key particulate air pollutants, such as ultrafine particles (UFP), BC, and metals, and allergens such as pollen. This will be achieved by developing new calibration procedures for (bio)aerosol monitoring systems, reference materials as well as portable devices that can serve as transfer standards for instrument calibration in the field.

Improving the monitoring of airborne particles is indispensable for developing effective public health measures and reducing the exposure of the population to the most harmful components of ambient PM. The outputs from this project will enable a more targeted policy on health-relevant components/properties and promote the revision of air quality legislation by extending the current legal framework to include metrics beyond PM mass concentration. Considering the enormous costs arising from hospitalisation and premature deaths due to particulate air pollution, the protection of public health should also lead to considerable financial benefits.

6 List of publications

The project achieved its publications goals with respect to all its technical objectives ahead of schedule reflecting the high level of collaborative works among the project partners as well as the external interest in the aerosol metrology topics addressed:

1. Method for Preparation of a Candidate Reference Material of PM₁₀ and PM_{2.5} Airborne Particulate Filters Loaded with Incineration Ash-Inter Comparison Results for Metal Concentrations. Bescond, A.; Oster, C.; Fiscaro, P.; Goddard, S.; Quincey, P.; Tsakanika, L.-A.; Lymperopoulou, T.; Ochsenkuehn-Petropoulou, M. *Atmosphere* 2021, 12(1), 67; <https://doi.org/10.3390/atmos12010067>
2. Quantification of Element Mass Concentrations in Ambient Aerosols by Combination of Cascade Impactor Sampling and Mobile Total Reflection X-ray Fluorescence Spectroscopy. Seeger, S.; Osan, J.; Czömpöly, O.; Gross, A.; Stosnach, H.; Stabile, L.; Ochsenkuehn-Petropoulou, M.; Tsakanika, L.A.; Lymperopoulou, T.; Goddard, S.; Fiebig, M.; Gaie-Levrel, F.; Kayser, Y.; Beckhoff, B. *Atmosphere* 2021, 12(3), 309; <https://doi.org/10.3390/atmos12030309>
3. Statistics of a Sharp GP2Y Low-Cost Aerosol PM Sensor Output Signals. Bučar, K.; Malet, J.; Stabile, L.; Pražnikar, J.; Seeger, S.; Žitnik, M. *Sensors* 2020, 20(23), 6707; <https://doi.org/10.3390/s20236707>
4. Calibration of optical particle size spectrometers against a primary standard: Counting efficiency profile of the TSI Model 3330 OPS and Grimm 11-D monitor in the particle size range from 300 nm to 10 µm. Vasilatou, K.; Walchli, C.; Koust, S.; Horender, S.; Lida, K.; Sakurai, H.; Schneider, F.; Spielvogel, J.; Wu, T. Y.; Auderset, K. *Journal of Aerosol Science*, Volume 157, September 2021, 105818 <https://doi.org/10.1016/j.jaerosci.2021.105818>
5. Characterization of unique aerosol pollution episodes in urban areas using TXRF and TXRF-XANES. Ottó Czömpöly, Endre Börcsök, Veronika Groma, Simone Pollastri, János Osán; *Atmospheric Pollution Research*, Volume 12, Issue 11, November 2021, 101214 <https://doi.org/10.1016/j.apr.2021.101214>
6. Assessment of real-time bioaerosol particle counters using reference chamber experiments. Gian Lieberherr, Kevin Auderset, Bertrand Calpini, Bernard Clot, Benoît Crouzy, Martin Gysel-Beer, Thomas Konzelmann, José Manzano, Andrea Mihajlovic, Alireza Moallemi, David O'Connor, Branko Sikoparija, Eric Sauvageat, Fiona Tummon, Konstantina Vasilatou; *Atmos. Meas. Tech.*, 14, 7693–7706, <https://doi.org/10.5194/amt-14-7693-2021>, 2021 <https://doi.org/10.5194/amt-14-7693-2021>
7. Reliable compositional analysis of airborne particulate matter beyond the quantification limits of total reflection X-ray fluorescence. Yves Kayser, János Osán, Philipp Hönicke, Burkhard Beckhoff; *Analytica Chimica Acta*, Volume 1192, 1 February 2022, 339367 <https://doi.org/10.1016/j.aca.2021.339367>
8. Towards standardisation of automatic pollen and fungal spore monitoring: Best practises and guidelines. Fiona Tummon, Nicolas Bruffaerts, Sevcen Celenk, Marie Choël, Bernard Clot, Benoît Crouzy, Carmen Galán, Stefan Gilge, Lenka Hajkova, Vitalii Mokin, David O'Connor, Victoria Rodinkova, Ingrida Sauliene, Branko Sikoparija, Mikhail Sofiev, Olga Sozinova, Danijela Tesendic, Konstantina Vasilatou; *Aerobiologia*, 2022, <https://doi.org/10.1007/s10453-022-09755-6>
9. Extending traceability in airborne particle size distribution measurements beyond 10 µm: Counting efficiency and unit-to-unit variability of four aerodynamic particle size spectrometers. Konstantina Vasilatou, Christian Wälchli, Kenjiro Iida, Stefan Horender, Torsten Tritscher, Tobias Hammer, Jenny Risslerd, François Gaie-Levrel, Kevin Auderset; *Aerosol Science and Technology*, 2022 <https://doi.org/10.1080/02786826.2022.2139659>
10. Flexible and reusable parylene C masks technology for applications in cascade impactor air quality monitoring systems: L. Vigna, M. Gottschalk, N. Cacocciola, A. Vernbusra. afsar@euramet.org, S.L. Marasso, S. Seeger, *Micro and Nano Engineering*, <https://doi.org/10.1016/j.mne.2022.100108>
11. Experimental determination of the gadolinium L subshells fluorescence yields and Coster-Kronig transition probabilities in X-ray spectrometry, Yves Kayser, Philipp Hönicke, Malte Wansleben, André Wählich, Burkhard Beckhoff. <https://doi.org/10.1002/xrs.3313>

12. Missed evaporation from atmospherically relevant inorganic mixtures confound experimental aerosol studies, J. Rissler, C. Preger, A. Eriksson, J. Lin, N. Prisle, B. Svenningsson, Environmental Science & Technology, <https://doi.org/10.1021/acs.est.2c06545>

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