April 10th, 2026

Revised manuscript “**An intercomparison study of optical particle size spectrometers for aerosol number size distribution measurements**”, by S. Bau et al. — Aerosol Research AR-2025-39

Dear Editor,

We were pleased to learn that the Reviewers have found the paper interesting. We would like to thank them for their constructive remarks and advices to improve our work.

We have substantially revised our manuscript accordingly. We fully agree that other parameters are also of interest in the description of size distributions. Therefore, we fully reviewed our dataset and further proceeded lognormal fitting to better interpret experimental results. The issue of total concentration has been discussed in the revised version, in line with the different dilution ratios which were dependent on the candidate OPSS involved by each partner.

Please find enclosed the revised version of the above-quoted manuscript for your review and consideration for publication in *Aerosol Research* as a full-length Paper.

Thank you for your consideration.

Sincerely yours,

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Comments made by Reviewer 1 :

This manuscript describes an inter-laboratory comparison of optical particle size spectrometers organized at the French national level. The focus was on the measurement of particle size. No reference method for number concentration measurements was available. According to the authors, this study aimed to reveal good laboratory practices when using commercially available optical particle counters. The authors, however, conclude that "Our database does not allow good laboratory practices to be proposed yet".

In my opinion, this study simply confirms what is already known, namely that the measurement of optical diameter with optical particle counters depends on the properties of the aerosols (especially the refractive index). Since little to no information is provided on the calibration of the particle counters under test by the manufacturers or end users, the study is simply descriptive and the main conclusion is that the particle counters do not report the same particle size. That was expected!

The calibration of optical particle counters (OPCs) designed for clean room monitoring has already been standardised within the ISO standard 21501-4. Likewise, the calibration of optical particle size spectrometers (OPSS) for ambient measurements is described in ISO 21501-1 (a revised version is soon to be published).

Size-certified spheres (e.g. silica or polystyrene spheres) are typically used for size calibration. Since ambient aerosols contain particles of different shapes and complex refractive index, the measured particle size distributions must be post-corrected by assuming a complex refractive index for the local ambient aerosol which was monitored. Do the monitoring stations in France apply such a correction? The authors seem to imply that all monitoring stations should calibrate their OPSS with the same size-certified particles, so that the measurements are "harmonised". This is not necessarily true. Monitoring stations should choose a "realistic" calibration aerosol, i.e. an aerosol with a refractive index as close as possible to that of the local ambient aerosols and then apply, if necessary, a correction. Alternatively, the method based on an aerodynamic aerosol classifier (Sang-Nourpour & Olfert, 2019; https://doi.org/10.1016/j.jaerosci.2019.105452) and atmospheric particles as calibration particles can be used.

In my opinion, this manuscript does not build on existing knowledge nor does it propose any measures for improving existing calibration procedures (as the authors themselves conclude). I therefore don't recommend publication.

Response :

We thank the reviewer for the time and attention devoted to evaluating our manuscript. We are fully aware of the points raised, including the fact that several observations made in our study are already known within the scientific community, particularly the dependence of optical diameter measurements on aerosol properties and the constraints associated with calibration procedures. On a first reading, we naturally agree with many of the reviewer's statements, which essentially recall well-established principles in aerosol metrology. However, the primary objective of our work was not to re-establish this existing knowledge. Instead, the study provided an opportunity to bring together a large number of partners at the national level on a topic that still requires further scientific investigation, especially regarding the access to an affordable and widely deployable calibration bench for multiple laboratories. Such approach has already been adopted in a previous work on Mobility Spectrometers (J Nanopart Res, 22, 2020). We acknowledge that coordinating the contributions of fifteen partners inevitably created challenges, including highlight more clearly the originality of our collective approach.

This originality lies in the fact that, despite using the same aerosol, the same generator, the same reference measurement device, the same experimental protocol and the OPS instruments from each participating laboratory, the reported results still differed in several cases. This outcome is scientifically meaningful: it shows that robust aerosol characterization is not as straightforward as might be assumed, and certainly not as easily guaranteed as some might suggest. It underlines the need for strengthened robustness in aerosol measurements and for a clearer understanding of the sources of variability when instruments are deployed under routine monitoring conditions.

From our perspective, this collaborative effort is still lacking in the current scientific literature: to our knowledge, no inter-laboratory comparison of such scale, involving the instruments commonly used by national monitoring networks, has been documented (not in France, but not elsewhere either). In this respect, the work carried out within ASFERA and its partners represents a unique and valuable contribution.

Although our database does not yet allow good laboratory practices to be proposed, it does provide the first systematic documentation on how these instruments perform under shared conditions, using resources that are realistically available for routine monitoring laboratories. In particular, elements to further interpret deviations related to particle optical properties are still required when OPS performance characterization is sought. The latter properties are particularly difficult to determine, especially when complex particles are involved (composition, shape, structure). Nonetheless, this study offers an essential starting point for future work that will need to address the more advanced calibration approaches mentioned by the reviewer and to further investigate how such methods could be implemented in practice by monitoring networks.

In this sense, we believe the study represents a necessary preliminary step, filling a current gap in the literature and supporting future methodological improvements.

Comments made by Reviewer 2 :

The manuscript written by Bau et al. deals with comparative measurements of a series of OPCs. The sheer number of measuring devices to be compared and the number of participating institutes are noteworthy. A considerable amount of time, 18 months, was spent on this project so that all partners could carry out the same experiment protocol using the same measuring instruments.

The motivation given was that good laboratory practices would be established and that no inter-laboratory measurements of OPC were done before. Unfortunately, the manuscript stopped being convincing at this point. Images from a scanning electron microscope were presented as the sole reference measurement method. An OPC was specified as the reference instrument which was referenced against all other OPCs, but its individual calibration points were not examined in detail. Three different aerosols were used as samples, which are only suitable to a limited extent and whose repeatability has not been investigated. The evaluations of the measurements and instruments were not considered individually but rather as a block. Information on the last service date or calibration was generalised and dismissed in the running text as inconclusive. Unfortunately, there was a lack of in-depth analysis such as of the optics, or pulse heights. In summary, the manuscript consists of a preliminary experiment, which many measurement partners have repeated, and the results are roughly the same.

The importance of inter-laboratory comparison is emphasised in the manuscript, but in-depth analysis considering differences in measuring instruments (light intensity, angles, calibrations, monodisperse aerosols, multimodal…..) is not taken into account. In its current form, I do not recommend publication of this manuscript in Aerosol Research.

In addition, I recommend that authors read the following articles and familiarise themselves with the approaches described therein.:

An intercomparison study of optical particle counters by van der Meulen et al. [1980] for the lower particle sizes, since the manuscript did not analysis lower detection size limits. As well as other Intercomparison studies such as: Rosenberg et al. 2012, which focused on pulse high analysis. Rosenberg et al. and Sang-Nourpour et al. 2019 (mentioned in this manuscript) have also examined measurement artefacts that arise with large particles. Namely, that with large particles, small

particles are also reported even though they are not actually present. This can distort measurements, such as those made in this manuscript, but this was not examined.
A review study by Vasilatou et al. 2025 or by McMurry 2000 on aerosol calibrations, since no SMPS/CPC number concentration validations and no PSL standards were used in this manuscript by Bau et al.
Dependencies on optics, such as the viewing/integration angles of the measuring instruments as in Szymanski and Liu 1986 (mentioned in manuscript) or Liu 1985.
Comparison with Mie theory regarding multiply OPC such as in Heim et al 2008 or Whitby et al 1967.
It would also have been interesting to investigate other, more atmospheric aerosols like in Crilley et al 2020, or using Nigrosin as an light absorbing aerosol with known refractive index as in Foster et al 2019.

References

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Rosenberg, P. D., Dean, A. R., Williams, P. I., Dorsey, J. R., Minikin, A., Pickering, M. A., and Petzold, A.: Particle sizing calibration with refractive index correction for light scattering optical particle counters and impacts upon PCASP and CDP data collected during the Fennec campaign, Atmos. Meas. Tech., 5, 1147–1163, https://doi.org/10.5194/amt-5-1147-2012, 2012.
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Konstantina Vasilatou, Kenjiro Iida, Mohsen Kazemimanesh, Jason Olfert, Hiromu Sakurai, Timothy A. Sipkens, Gregory J. Smallwood,Aerosol physical characterization: A review on the current state of aerosol documentary standards and calibration strategies,Journal of Aerosol Science,Volume 183,2025,106483,ISSN 0021-8502, https://doi.org/10.1016/j.jaerosci.2024.106483.
Peter H McMurry,A review of atmospheric aerosol measurements,Atmospheric environment,Volume 34, Issues 12–14,2000,Pages 1959-1999,ISSN 1352-2310, https://doi.org/10.1016/S1352-2310(99)00455-0.
Szymanski, W.W. and Liu, B.Y.H. (1986), On the Sizing Accuracy of Laser Optical Particle Counters. Part. Part. Syst. Charact., 3: 1-7. https://doi.org/10.1002/ppsc.19860030102
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Kenneth T. Whitby and Richard A. Vomela *Environmental Science & Technology* **1967** *1* (10), 801-814 DOI: 10.1021/es60010a002.
Crilley, L. R., Singh, A., Kramer, L. J., Shaw, M. D., Alam, M. S., Apte, J. S., Bloss, W. J., Hildebrandt Ruiz, L., Fu, P., Fu, W., Gani, S., Gatari, M., Ilyinskaya, E., Lewis, A. C., Ng'ang'a, D., Sun, Y., Whitty, R. C. W., Yue, S., Young, S., and Pope, F. D.: Effect of aerosol composition on the performance of low-cost optical particle counter correction factors, Atmos. Meas. Tech., 13, 1181–1193, https://doi.org/10.5194/amt-13-1181-2020, 2020.
Foster, K., Pokhrel, R., Burkhart, M., and Murphy, S.: A novel approach to calibrating a photoacoustic absorption spectrometer using polydisperse absorbing aerosol, Atmos. Meas. Tech., 12, 3351–3363, https://doi.org/10.5194/amt-12-3351-2019, 2019.

Nevertheless, I would urge the authors to possibly change the focus of these study and, with regard to the literature references presented, to supplement the manuscript with further methods and theoretical approaches.
With a stronger focus on the generation of the measured aerosol and calibration data, the data obtained could still be relevant. As a great number of OPC instrument did not differ significantly from each other regardless the service status.

Response :
We thank the reviewer for evaluating our manuscript.
We agree that optical equivalent diameter provided by OPC can be significantly different to the geometric diameter of particles observed from EM images. It is forth noting that OPC are typically calibrated with PSL particles with a refractive index of 1.59. Therefore, for a given light intensity, the size classification of OPC strongly depend on that parameter. Determining or measuring optical properties of the test aerosols is a complex question, which is largely beyond the scope of this paper. An in-depth study of the impact of particle refractive index on the response of the OPC would be feasible through the softwares with a post-analysis. However, this parameter remains unknown for the Holi powder sample, and at least its imaginary part for the two other test aerosols.
We would like to thank the reviewer for drawing our attention to specific technical points. Concerning the issue of particle coincidence, each participant was asked to make sure that the OPC involved were used under "good laboratory pratices" conditions, i.e. by ensuring the absence of internal errors reported by the devices, including coincidence errors. Concerning the choice of the test aerosols, we would like to mention that this inter-laboratory exercise was organised without funding, each participant performed the experiments on its own budget. There was no intention to generate test aerosols that would mimic specific scenarios, e.g. a typical urban atmospheric aerosol. Besides, regarding another of your comments, we ensured that the size range covered by the test aerosols was in line with the size specifications of the OPC involved to avoid potential biases due to detection limits.
Nonetheless, we would like to emphasize that this study establishes both a methodology and a reference dataset for improving OPC reliability at national and international scales. As stated in the title, this work is an inter-laboratory comparison and not a calibration study. For that reason, no reference devices (eg SMPS, APS, AAC-CPC) were used for validation, and no PSL standards were used.

Comments made by Reviewer 3 :
The authors aimed to establish good laboratory practice for the use of optical particle counters. Inter-laboratory comparative tests were conducted according to a common protocol at the national level in France. Three types of test aerosols generated with the same equipment were used, and the same OPC as the reference instrument was used to retrieve the modal diameters. In the end, the conclusion is that, based on their measurements, it is not possible to demonstrate good laboratory practice. While the stated goal is welcome and forward-looking, many questions remain unanswered. I highlight here a few of these, which definitely need to be addressed in the manuscript.
It remains unclear what authors mean by good laboratory practice when using standard OPCs. Optical particle counters measure the size distribution and number concentration of the particles being examined. When analysing systematic deviations and dispersion in the measured data, coincidence error is mentioned as a possible explanation. If this data is available, why was it not taken into account during the measurements and data processing?
If the stated goal is to reveal a good laboratory practice, why was only one protocol followed during the measurements?

Three types of test aerosols were selected by the authors: why these in particular? In the introduction, the authors start from the premise of improving the temporal resolution of measurements relating to ambient air quality. What is the relevance of the listed test aerosols in this context? Why is the aerosol used for calibration not among them?
The article mentions the involvement of 16 partners in several places, but ultimately only 12 are included in the study.
The authors state that: This article presents the measured PNSD using 35 OPCs – where are these results presented?

Response :
We are grateful for the remarks formulated by the Reviewer.
As already mentioned in our response to a previous comment regarding the issue of particle coincidence, each participant was asked to make sure that the OPC involved were used under « good laboratory pratices » conditions, i.e. by ensuring the absence of internal errors reported by the devices, including coincidence errors. This point has been stressed in the revised version of the manuscript.
A common generation setup and measurement protocol has been shared and implemented by all participants. This study did not aim at comparing experimental results stemming from multiple approaches ; rather, the idea was to elaborate both a methodology and a reference dataset for improving OPC reliability at national and international scales.
Concerning the choice of the test aerosols, there was no intention to generate test aerosols that would be representative of specific ones, e.g. a typical urban atmospheric aerosol. Instead, one of our objectives was to investigate the possibility to produce the same aerosols in the different laboratories involved by using a straightforward setup. In this context, we decided to implement a dry-based method, as it seemed easier to provide the products (powders) to all partners. Because this inter-laboratory exercise was organised without funding, the powders selected were provided by three partners to all participants without seeking specific characteristics. We believe that the three test aerosols allow the OPC involved to be challenged over a large range of sizes, with varying complexity.
As pointed out by the Reviewer, 16 partners were involved by only 12 provided experimental data. Indeed, technical issues (instruments with zero drift, with inappropriate flowrate, etc.) have arisen during some of the experiments, that could not be repeated for organizational reasons (each participant had the overall setup for only few days).
Finally, we decided to not show the particle size distributions obtained from the 35 instruments involved. This choice was made after a graphical representation of the 35 spectras, which was extremely difficult to read, taking into account the relative scale of each device / dilution levels and various channel widths. Insted, these data were further digested and expressed in terms of modal optical diameter, which was supposed to be more appropriate for inter-instrument comparison. Instead, a data processing involving a lognormal fitting procedure was applied to all data.