

Answer to Referee #4 comments on egusphere-2025-2571 under-review manuscript

We thank the referees for carefully evaluating our manuscript for a second time and for the constructive comments, which have helped us refine the manuscript. In response to the reviews, we have revised the manuscript and include their comments and suggestions.

The authors have made substantial revisions to the manuscript, significantly improving its clarity, methodological rigor, and transparency. Although the article is already quite comprehensive, there is one issue regarding field observations that the author could elaborate on in greater detail; this would be significant for advancing future research on dust microphysics.

In this study, PSD measurements are a fundamental prerequisite for retrieving the imaginary part of the complex refractive index. The authors acknowledge that, unlike controlled chamber experiments, field observations suffer from imperfect sampling between the OPC and optical instruments, and they adopt a tssa-based cost function to reduce sensitivity to absolute concentration mismatches. This approach is well justified if the inter-instrument discrepancy manifests primarily as a uniform scaling of total particle number while preserving the PSD spectral shape.

Yet, if the sampling mismatch leads to significantly different PSD shapes between instruments, rather than merely a constant offset in total concentration, the retrieval could suffer from substantial systematic errors that are not fully captured by the current sensitivity framework.

We thank the referee for the positive assessment of the revised manuscript and for raising this important point. We agree that the fidelity of the PSD used in the inversion is a central limitation for our field-based retrievals of the imaginary part of the refractive index. The revised manuscript already discusses the possible mismatch between the OPC-derived PSD and the optical measurements, but we agree that the distinction between an absolute number-concentration mismatch and a size-dependent PSD-shape distortion should be made more explicit.

We have therefore further revised the discussion to clarify that the use of the tssa-based cost function reduces sensitivity to absolute concentration mismatches, but does not fully remove the effect of size-dependent sampling differences. We now explicitly discuss the possible impact of PSD-shape distortion on retrieved k , and we provide recommendations for future field campaigns aimed at reducing this uncertainty.

I encourage the authors to address the following questions more explicitly:

1. Do the authors expect the PSD shape to remain approximately invariant across instruments, with only total number varying? Or is spectral shape distortion a realistic concern given the different inlets, sampling lines, and size-dependent transmission efficiencies involved?

We agree with the referee that PSD-shape distortion is a realistic concern in field observations. We do not assume that the PSD shape necessarily remains invariant across instruments, with only the total number concentration changing. The tssa-based cost function reduces sensitivity to a uniform scaling of the particle number concentration, but it cannot fully correct for size-dependent differences in the particle population sampled by the OPC and by the optical instruments.

In our campaign, this issue is particularly relevant because the Fidas measured the PSD through its own inlet and over a broad optical-diameter range, whereas the AE33 and nephelometer sampled through a common PM_{2.5} inlet during the period used for the retrieval. Although we applied an effective PM_{2.5} transmission to the converted Fidas PSD, this procedure may fully reproduce possible differences in inlet transmission, sampling-line losses, particle deposition, counting efficiency, or size-dependent sampling efficiency between instruments.

We have therefore revised the manuscript to state more clearly that both types of mismatch may occur: a uniform number-concentration mismatch and a size-dependent PSD-shape distortion. The latter is more problematic because it can change the relative contribution of different particle sizes to absorption and scattering, and therefore affect the retrieved k .

2. If PSD shape distortion does occur, what would be its impact on k retrievals?

If PSD-shape distortion occurs, it can introduce systematic biases in the retrieved k . This is because absorption, scattering, and truncated scattering do not respond equally to all particle sizes. A size-dependent under- or over-representation of particles in the PSD can therefore change the simulated optical properties and force the inversion to compensate through a different value of k .

The diagnostic closure test shown in Fig. R4.1 of the [previous response](#) supports this concern. In that test, the factor required to improve closure between the OPC-derived PSD and the optical measurements was not constant with wavelength. If the mismatch were only a uniform scaling of total particle number, and assuming perfect optical measurements, the optimal scaling factor would be approximately wavelength independent. Instead, its spectral behaviour suggests that part of the discrepancy may be size dependent. One possible interpretation is that the PSD may underestimate particles near the lower and upper ends of the PM_{2.5}-relevant size range, although the available field data do not allow us to prove this or derive a robust correction.

For this reason, we avoid interpreting the retrieved k as a unique, unconstrained value. Instead, we present it as a conditional retrieval under prescribed assumptions for particle shape, density, n , AE33 absorption correction, and the PSD used in the inversion. We have further clarified in the manuscript that the current sensitivity framework addresses several

important sources of uncertainty, but cannot fully quantify all possible PSD-shape distortions caused by size-dependent sampling differences.

3. How should future field campaigns be designed to minimize this uncertainty? Specifically, what instrumental co-location strategies, inlet harmonization, or cross-instrument closure protocols would be most effective?

We agree that this is an important outcome of the study. Future field campaigns aiming to retrieve dust refractive indices from ambient measurements should prioritize PSD–optical closure from the design stage. Based on our experience, the most effective strategy would be to ensure that the PSD and optical instruments sample as similar a particle population as possible.

Ideally, the OPC, nephelometer, and absorption instrument should sample from a common, well-characterized inlet and manifold, with short sampling lines, minimized bends, controlled flow conditions, and similar transmission efficiencies across instruments. Where a common inlet is not feasible, parallel inlets should be harmonized as much as possible in terms of inlet type, cut-off, height, flow rate, tubing length, and orientation relative to the dominant wind direction. The use of conductive tubing, careful control of flow rates, and minimization of particle residence time would also help reduce size-dependent particle losses.

In addition, future campaigns should include explicit closure protocols. These could include: side-by-side intercomparison of PSD instruments before and during the campaign; periodic switching of instruments between inlets; laboratory or field characterization of inlet transmission using representative dust; comparison between OPC-derived mass and independent filter or impactor measurements; and optical closure tests in which measured PSDs are used to simulate scattering and absorption under controlled assumptions. Size-resolved filter or impactor samples, microscopy-based particle-shape information, and independent density measurements would also help constrain the PSD conversion and reduce the degrees of freedom in the retrieval.

We have added a discussion of these recommendations to the manuscript. We also note that the optimal configuration will depend on the field site, expected dust loading, available power and maintenance capacity, and the scientific objective of the campaign. Nevertheless, the central requirement is to design the sampling system so that PSD and optical measurements are as co-located and size-consistent as possible.

4. What is the scientific value of PSD measurements when their absolute accuracy and spectral shape fidelity are uncertain? Should future efforts prioritize perfecting PSD sampling in the field, or should the community invest more heavily in chamber-based retrievals with well characterized PSDs, using field data primarily for validation and context?

We thank the referee for this broader question. In our view, field and chamber measurements provide complementary constraints, and neither can replace the other.

Chamber experiments are essential because they allow better control of the sampled particle population, repeated measurements under known conditions, and more rigorous closure

between PSD, scattering, absorption, and retrieved refractive index. They are therefore particularly valuable for testing retrieval methods, quantifying instrumental biases, and isolating the role of particle size, shape, composition, and mixing state.

Field measurements, however, remain indispensable because they sample dust under natural emission, transport, turbulence, and mixing conditions. These conditions are difficult to reproduce fully in the laboratory. In particular, near-source field observations provide information on freshly emitted dust, natural PSD variability, local emission mechanisms, and the possible interaction between mineral dust and other aerosol types. Even when PSD measurements have uncertainties, they provide essential constraints that would otherwise be unavailable. Their scientific value lies not only in providing absolute PSDs, but also in documenting the variability and representativeness of dust optical properties under real atmospheric conditions.

Therefore, future efforts should not choose between field and chamber approaches. Instead, the community should combine them more tightly. Chamber experiments can be used to characterize instruments, evaluate retrieval algorithms, and quantify the sensitivity to PSD, shape, and composition under controlled conditions. Field campaigns can then test how these relationships hold under natural dust-emission conditions. Conversely, field observations can identify which particle sizes, shapes, and source conditions should be reproduced in the laboratory. This combined strategy is likely the most effective path towards reducing uncertainty in dust absorption and refractive-index retrievals.

These points have been emphasized in the conclusion section.

A more thorough discussion of these points would greatly enhance the paper's contribution to the broader community's efforts to constrain dust absorption properties, and would help clarify the unique value proposition of field-based versus laboratory-based approaches to refractive index retrieval.

We agree with the referee. We have added a paragraph to the manuscript discussing the complementary value of field and chamber approaches, and the specific role of PSD uncertainty in field-based refractive-index retrievals. The new text emphasizes that field observations provide unique representativeness for natural dust-emission conditions, while chamber studies provide stronger control over PSD, sampling, and closure. We also clarify that improving future field retrievals will require better inlet harmonization, co-located PSD and optical measurements, independent constraints on density and shape, and explicit PSD–optical closure protocols.

Answer to Referee #3 comments on egusphere-2025-2571 under-review manuscript

General comments: First of all, I must appreciate the amount of work authors did to improve the article. It is a rare sight to see such dedication to keep pushing and publish almost unsalvageable work, and to meddle and reshape it into something of actual use to the community. I apologize for being late on my review, remembering what was provided almost a year ago, and meaningfully ingesting a supplementary material that itself worth a separate publication takes some effort. I do see a significantly stronger methodology and much more robust conclusion justification in the resubmitted article. However, I still see minor issues, that have to be addressed prior to publishing, I provide the details below.

We thank the referee for the careful second evaluation of the manuscript and for the positive assessment of the revised version. We are particularly grateful for the comments on the OSSE section, which helped us make the connection between the Supplement and the main manuscript clearer and to describe the synthetic perturbations more explicitly. We have revised the manuscript and Supplement accordingly, as detailed below.

Minor comments:

Once again, I could not stress enough what an impressive analysis authors did in the OSSE section of the supplement. Provided clarifications addressed all of my concerns that were raised in the previous review. Technically I should have nothing to complain, however, the OSSE study is provided in an additional supplement which is barely related to the document itself. The fact that it is referenced in document only twice gives an impression that authors what to swipe some dirt under the carpet. Indeed, the OSSE provided is very detailed, and analyses robustly the error propagation of the methodology of k retrievals. I would suggest integrating it better with the main document discussion.

We thank the referee for this positive assessment of the OSSE analysis. We agree that the OSSEs are central for supporting the retrieval methodology and should be more clearly connected to the main manuscript. Our intention in placing the detailed OSSEs in the Supplement was to keep the main text focused on the field observations and retrieval results, while providing the full technical analysis separately. However, we agree that the main manuscript should better explain why the OSSEs were performed and how they support the methodological choices.

We have therefore revised Sect. 2.6.3 to introduce the OSSEs more explicitly. The new text explains that the OSSEs test the inversion method, support the choice of the (tssa)-based cost function and truncated-scattering observation operator, and quantify the sensitivity of retrieved (k) to perturbations in absorption, truncated scattering, PSD, particle density, particle shape, and (n). We also now state more clearly that the OSSEs distinguish between approximately uniform particle-number perturbations and size-dependent PSD-shape perturbations. This directly supports our interpretation of the retrieved values as conditional estimates of (k), rather than as a unique unconstrained retrieval of dust shape, PSD, (n), and (k).

We have also revised the Supplementary OSSE section to clarify the figure captions, and better connect the interpretation of Figs. S11–S15 with the methodological choices in the main manuscript.

The OSSE uses synthetic perturbations of unspecified/generic magnitude I would suggest include detailed descriptions of these, ideally link to to the realistic estimations of the observation noise from the main document. Also it is not clear how PSD were perturbed, S11 figure shows some perturbations but it is not clear are they related to shape assumption, refractive indices, FIDAS uncertainty (or all three combined), please provide these details.

We agree with the referee that the perturbations used in the OSSEs should be described more explicitly. We have revised the Supplement to specify both the magnitude of the perturbations and how they were generated.

In the OSSEs, the synthetic truncated scattering coefficient, (t_{sca}), and absorption coefficient, (abs), were independently perturbed using a uniform distribution within ($\pm 10\%$) of the prescribed true value. This range was chosen as a controlled sensitivity test of measurement-level uncertainty in the optical variables, rather than as a full probabilistic uncertainty model.

The PSD perturbations were designed to explore both changes in the shape of the PSD and changes in the absolute particle number concentration. First, the campaign-average PSD was approximated as the sum of eight lognormal modes. The total number concentration of each lognormal mode was then independently perturbed using a uniform distribution within ($\pm 50\%$) of its prescribed value. The resulting PSD was first normalized to preserve the true total particle number; this step isolates PSD-shape distortions while keeping the integrated particle number constant. In a second step, the total number concentration of the full PSD was also perturbed using a uniform distribution within ($\pm 50\%$) of the true total number. This procedure allows the OSSEs to explore both size-dependent PSD-shape distortions and approximately uniform particle-number changes.

We have clarified that these PSD perturbations are not intended to represent one single source of uncertainty, such as particle shape, refractive index, or Fidas counting uncertainty alone. Rather, they are designed to mimic the combined effect of plausible size-dependent sampling and sizing uncertainties that could arise from inlet transmission, particle losses, counting efficiency, and the optical-to-geometric diameter conversion.

We have also expanded the interpretation of the OSSE results in the Supplement, particularly the comparison between the (t_{ssa})-based cost function used in the manuscript and the alternative cost function based on extensive optical properties, closer to the approach of Di Biagio et al. (2019). The expanded discussion explains that, when an extensive-property cost function is used, biases in the PSD or in the relative sampling of the OPC and optical instruments can be translated directly into biases in the retrieved k . In the OSSE, when the PSD underestimates the true particle number, the inversion compensates by retrieving a larger k , fitting absorption but underestimating t_{sca} and t_{ssa} . Conversely, when the PSD overestimates the true particle number, the inversion retrieves a lower k , fitting absorption but overestimating t_{sca} and t_{ssa} . This behaviour illustrates why the t_{ssa} -based cost function is more appropriate for the present field dataset, where the OPC and optical instruments

sampled through different inlet systems and robust absolute PSD–optical closure cannot be guaranteed.

We also clarify that a Di Biagio et al. (2019)-type cost function can be appropriate when PSD and optical properties are measured from the same aerosol population and no relevant PSD–optical sampling bias is present. Under such controlled conditions, the independent information in absorption and scattering may help constrain both n and k . In the present field campaign, however, possible PSD–optical sampling differences cannot be robustly corrected with the available data, so we retain the tssa-based cost function and interpret the retrieved (k) values as conditional estimates.

We have added these details to the Supplement, and Fig. S11 now illustrates the resulting perturbed PSDs, abs , $tsca$, and $tssa$.

Table 1. AAE_370-950: Why values so different for full period as compared to others? Is this a typo or a dataset bias? Please discuss.

We thank the referee for identifying this issue. It was a typo in Table 1. The correct AAE value for the full PM_{2.5} period is 3.02, not 13.02. We have corrected the table accordingly.

I do believe mentioning Lin et al., 2018 is well justified in the scope of this article (authors ignored this suggestion from the previous review for some reason). This team made amazing job in tackling non-sphericity of dust

Lin, W., Bi, L., & Dubovik, O. (2018). Assessing superspheroids in modeling the scattering matrices of dust aerosols. Journal of Geophysical Research: Atmospheres, 123, 13,917–13,943. <https://doi.org/10.1029/2018JD029464>

We agree with the referee. We have added Lin et al. (2018) to the Introduction in the paragraph discussing the effect of dust non-sphericity on optical properties. This reference is highly relevant because it assesses the use of superspheroids for modelling dust scattering matrices and therefore complements the discussion of spheroids, irregular particles, and other non-spherical shape models used in the present study.

The revised text now acknowledges that several approaches have been developed to represent dust non-sphericity, including spheroids, superspheroids, and more complex irregular-particle models.

Technical comments:

Line 295: It is not clear if any assumed filter convolution was used in the conversions, or instrument was assumed to be monochromatic, does this choice affect the conversions? Please specify and discuss.

We thank the referee for pointing out that this needed clarification. The Fidas light source was not treated as monochromatic. Instead, we assumed an effective Gaussian light-source spectrum centred at 389 nm, with a standard deviation of 77 nm. This spectrum was inferred by fitting the manufacturer-provided conversion tables, which convert PSL-calibrated PSDs into PSDs for spherical particles with different refractive indices.

In practice, for each aerosol type, particle shape, refractive index, and size, the sideward scattered intensity was computed by integrating over this effective light-source spectrum. Thus, the conversion from PSL optical diameter to dust geometric diameter includes a spectral convolution over the inferred Fidas light-source distribution.

We agree that the assumed light-source spectrum can affect the conversion. However, because the manufacturer does not provide the full spectral response of the instrument, we constrained the effective spectrum using the manufacturer conversion software itself. This approach makes the conversion internally consistent with the available Fidas information, although uncertainty in the exact light-source spectrum remains a limitation. We have clarified this point in Sect. 2.6.1.