Referee Report on egusphere-2026-424

General assessment

This manuscript presents a multi-platform characterization of a polluted dust episode using in situ optical measurements, particle size distributions, single-particle composition, and an inverse Mie-based retrieval of aerosol complex refractive index (ACRI).

The dataset is valuable and scientifically relevant. The combination of optical properties, size distributions, and compositional information is particularly useful for understanding aerosol processes in a dust-influenced environment. The manuscript is clearly within the scope of ACP and represents a meaningful contribution.

The inversion framework used to retrieve refractive index is also appropriate and follows a class of optical-closure approaches that has precedent in the literature. The main concern raised in this review is therefore not related to the existence of the inversion itself, but to the strength of the interpretation attached to the retrieved refractive index.

Central scientific issue

(a) Inverse formulation and what is actually solved

The study measures the bulk optical coefficients

$$\sigma_{\mathrm{sca,obs}}, \qquad \sigma_{\mathrm{abs,obs}}.$$

The aerosol complex refractive index is written as

$$m = n + ik,$$

where n is the real part and k is the imaginary part.

At the most fundamental level, the modeled optical coefficients are obtained by integrating the optical contribution of particles over the particle size distribution. If D_p is particle diameter and $N(D_p)$ is the number size distribution, then for spherical particles the modeled scattering and absorption coefficients may be written as

$$\sigma_{\mathrm{sca,model}}(m,\lambda) = \int_0^\infty Q_{\mathrm{sca}}(m,x)\,\frac{\pi D_p^2}{4}\,N(D_p)\,\mathrm{d}D_p,$$
$$\sigma_{\mathrm{abs,model}}(m,\lambda) = \int_0^\infty Q_{\mathrm{abs}}(m,x)\,\frac{\pi D_p^2}{4}\,N(D_p)\,\mathrm{d}D_p,$$

where

$$x = \frac{\pi D_p}{\lambda}$$

is the size parameter, and Q_{sca} and Q_{abs} are the Mie efficiencies for scattering and absorption.

It is useful to unpack these expressions explicitly. Each particle contributes to the bulk optical coefficient through three linked terms:

- the particle abundance at that size, $N(D_p)$,
- the geometric cross-section, $\pi D_p^2/4$,
- the optical efficiency, $Q(m,x)$, which depends on refractive index and size parameter.

Thus, in compact conceptual form,

$$\sigma \sim \int (\text{number}) \times (\text{area}) \times (\text{efficiency})\, \mathrm{d}D_p.$$

This makes clear that the optical coefficients depend simultaneously on particle size and refractive index:

$$\text{optical coefficient} = \text{function of size distribution, particle shape, and } m.$$

In the spherical Mie formulation used by the manuscript, this dependence is approximated as

$$\sigma = f_{\text{sphere}}(\text{size distribution, } m).$$

To compare model and measurements, the problem can be written in vector form as

$$\mathbf{y}_{\text{obs}} = \begin{bmatrix} \sigma_{\text{sca,obs}} \\ \sigma_{\text{abs,obs}} \end{bmatrix}, \qquad \mathbf{y}_{\text{model}}(n,k) = \begin{bmatrix} \sigma_{\text{sca,model}}(n,k) \\ \sigma_{\text{abs,model}}(n,k) \end{bmatrix}.$$

The residual vector is then

$$\mathbf{r}(n,k) = \mathbf{y}_{\text{model}}(n,k) - \mathbf{y}_{\text{obs}}.$$

The inversion seeks the pair (n,k) that makes the residual as small as possible. In its simplest least-squares form, this means minimizing

$$J(n,k) = \|\mathbf{r}(n,k)\|_2^2 = \mathbf{r}(n,k)^{\mathrm{T}}\mathbf{r}(n,k),$$

that is,

$$J(n,k) = \left(\sigma_{\text{sca,model}} - \sigma_{\text{sca,obs}}\right)^2 + \left(\sigma_{\text{abs,model}} - \sigma_{\text{abs,obs}}\right)^2.$$

A normalized form, which is often closer to what is implemented in practice, is

$$J(n,k) = \left(\frac{\sigma_{\text{sca,model}} - \sigma_{\text{sca,obs}}}{\sigma_{\text{sca,obs}}}\right)^2 + \left(\frac{\sigma_{\text{abs,model}} - \sigma_{\text{abs,obs}}}{\sigma_{\text{abs,obs}}}\right)^2.$$

Therefore, the inversion solves

$$(n^*, k^*) = \arg\min_{n,k} J(n,k).$$

This ensures that

$$\mathbf{y}_{\text{model}}(n^*, k^*) \approx \mathbf{y}_{\text{obs}},$$

or in component form,

$$\sigma_{\text{sca,model}} \approx \sigma_{\text{sca,obs}}, \qquad \sigma_{\text{abs,model}} \approx \sigma_{\text{abs,obs}}.$$

However, this is a statement about agreement, not about uniqueness:

$$\text{good agreement in } \sigma \not\Rightarrow \text{ unique or intrinsically correct } (n,k).$$

This distinction is central. Optical closure only shows that, within the chosen forward model, one can find a refractive-index pair that reproduces the observed coefficients well. It does not by itself prove that the retrieved (n,k) is the only physically correct solution, nor that it is free from compensation for missing physics.

The reason is that the true forward problem is more general:

$$\mathbf{y}_{\text{true}} = F(\text{size distribution, particle shape, mixing state, } n,\ k),$$

whereas the inversion effectively uses

$$\mathbf{y}_{\text{model}} = F_{\text{sphere}}(\text{size distribution, } n,\ k).$$

For mineral dust,

$$\text{real particles are irregular and nonspherical} \neq \text{spherical particles.}$$

Hence the structural form of the model is incomplete:

$$F_{\text{true}} \neq F_{\text{sphere}}.$$

A useful way to write this is

$$\mathbf{y}_{\text{obs}} = F_{\text{sphere}}(\text{size}, n, k) + \boldsymbol{\varepsilon}_{\text{shape}} + \boldsymbol{\varepsilon}_{\text{size}} + \boldsymbol{\varepsilon}_{\text{mix}} + \boldsymbol{\varepsilon}_{\text{meas}},$$

where the extra terms represent, respectively, shape error, size-distribution error, mixing-state error, and measurement error.

Under this decomposition, the retrieved refractive index does not only fit the aerosol refractive index itself. It may also compensate for part of the missing physics:

$$\text{missing physics} \rightarrow \text{partly absorbed into } (n, k).$$

This leads directly to non-uniqueness. It is entirely possible that

$$J(n_1, k_1) \approx J(n_2, k_2), \qquad (n_1, k_1) \neq (n_2, k_2).$$

In other words, multiple refractive-index pairs may produce similarly good agreement with the observations.

The same point can be written in differential form:

$$\delta\sigma = \frac{\partial\sigma}{\partial n}\,\delta n + \frac{\partial\sigma}{\partial k}\,\delta k + \frac{\partial\sigma}{\partial(\text{size})}\,\delta(\text{size}) + \frac{\partial\sigma}{\partial(\text{shape})}\,\delta(\text{shape}) + \cdots$$

This expression shows, in a local linear sense, that the optical coefficient is sensitive not only to n and k, but also to errors in size distribution and particle shape. Consequently, a perturbation in size or shape can, in practice, be offset by a perturbation in the retrieved refractive index:

$$\text{change in size or shape} \Rightarrow \text{compensating change in } (n, k).$$

That is precisely why agreement in optical closure does not automatically imply a unique physical refractive index.

Conclusion of (a): the inversion is mathematically valid, but it is not strictly unique, even if the fit appears excellent.

(b) Consequence for interpretation

A good fit does not imply a uniquely determined or directly physical refractive index.

If the retrieved refractive index is interpreted as an intrinsic material property of the aerosol, then the interpretation becomes too strong relative to what the inversion actually demonstrates.

A more accurate formulation is:

> The retrieved refractive index represents an *effective parameter* that reproduces the observed optical properties under the assumptions of the model.

This interpretation is consistent with previous work. Abo Riziq et al. (2007) showed reduced agreement for mixed and absorbing aerosols, and Kong et al. (2024) showed that particle nonsphericity and size-distribution uncertainty are important sources of uncertainty for refractive-index retrieval in mineral dust.

This does not invalidate the results, but it does require a more cautious interpretation.

Major comments

1. **Missing reference in the introduction (Line 16)**
   The introduction should include Adebiyi et al. (2023, *Aeolian Research*), which provides a comprehensive synthesis of mineral dust in the Earth system.

2. **Method context (Section 2.5)**
   The manuscript should clarify that the inversion follows an optical-closure framework with precedent in earlier studies. This is important because the central concern is not the use of the framework itself, but the interpretation attached to its output.

3. **Interpretation of refractive index (Section 2.5)**
   The retrieved refractive index should be described as model dependent. In particular, it should not be presented as a uniquely determined intrinsic material property unless that stronger claim is justified.

4. **Dust particle shape (Section 2.5)**
   The manuscript should explicitly note that Mie theory assumes spherical particles, whereas mineral dust particles are nonspherical and often inhomogeneous. This is not a minor limitation. It is central to the inversion.

5. **Elevated refractive index values (Results/Discussion)**
   Higher values than literature should be interpreted cautiously. Such differences may arise from inversion degeneracy, particle size-distribution uncertainty, particle nonsphericity, mixing-state assumptions, or measurement uncertainty, rather than from a true physical enhancement alone.

6. **Size-distribution sensitivity (Section 2.5)**
   The dependence of the retrieval on particle size should be stated more explicitly. In compact form,

   $$\Delta(\text{size distribution}) \Rightarrow \Delta m_{\text{retrieved}}.$$

   More fundamentally, because

   $$\sigma_{\text{sca,model}} = \int_0^\infty Q_{\text{sca}}(m,x)\,\frac{\pi D_p^2}{4}\,N(D_p)\,\mathrm{d}D_p,$$

   and

   $$\sigma_{\text{abs,model}} = \int_0^\infty Q_{\text{abs}}(m,x)\,\frac{\pi D_p^2}{4}\,N(D_p)\,\mathrm{d}D_p,$$

a perturbation in $N(D_p)$ propagates directly into the retrieved m. This is especially important for coarse-mode dust particles and in the context of SMPS–OPC merging.

Specific comments

7. **Instrument maintenance (Line 86)**
   Please clarify whether a regular daily maintenance or instrument-check period was applied between 11:45 and 12:15 (UTC +05:30), including filter changes at 12:00. If so, it would be helpful to explicitly state how these periods were treated in the dataset (e.g., excluded from analysis or flagged) and whether they influence the reported averages.
8. **Inlet description (Section A4)**
   Line 3 of Section A4 needs a citation for the statement that the sampling lines were kept below 1 m for all instruments.
9. **Figure 1 caption (Line 4)**
   A clearer wording would be:

   > The real part of the refractive index (RRI) primarily influences scattering, whereas the imaginary part (IRI) primarily influences absorption.

Reviewer assessment

The manuscript presents a high-quality dataset and a sound methodological framework. The main issue identified relates to the interpretation of the retrieved refractive index rather than to the underlying analysis.

The requested revisions mainly concern clarification of assumptions and a more precise framing of the results. These changes do not alter the overall value of the dataset or the main observational findings.

This point can be addressed through clarification of assumptions and a more precise formulation of the conclusions, without requiring substantial additional analysis.

Accordingly, the manuscript is suitable for publication after **minor revision**, provided that the interpretation of the refractive index is adjusted to reflect its model-dependent nature.

Summary evaluation

A structured evaluation of the manuscript is provided below.

Principal criteria	**Low (1)**	**Fair (2)**	**Good (3)**	**Excellent (4)**	**Outstanding (5)**
Scientific significance: Does the manuscript represent a substantial contribution to scientific progress within the scope of Atmospheric Chemistry and Physics?				X	
Scientific quality: Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way, including appropriate references?			X		
Presentation quality: Are the scientific results and conclusions presented in a clear, concise, and well-structured way?				X	

References

Abo Riziq, A., Erlick, C., Dinar, E., and Rudich, Y.: Optical properties of absorbing and non-absorbing aerosols retrieved by cavity ring down spectroscopy, *Atmos. Chem. Phys.*, **7**, 1523–1536, 2007.

Adebiyi, A., Kok, J. F., Murray, B. J., et al.: A review of coarse mineral dust in the Earth system, *Aeolian Research*, **60**, 100849, 2023.

Kong, S., Wang, Z., and Bi, L.: Uncertainties in laboratory-measured shortwave refractive indices of mineral dust aerosols and derived optical properties: a theoretical assessment, *Atmos. Chem. Phys.*, **24**, 6911–6935, 2024.

Womack, C. C., Washenfelder, R. A., Wagner, N. L., et al.: Complex refractive indices in the ultraviolet and visible spectral region for highly absorbing biomass burning aerosol, *Atmos. Chem. Phys.*, **21**, 7235–7252, 2021.