Remarks

Original comments by the editor are printed in bold.

Our answers are printed in regular font.

Changes in the manuscript are printed in italic font.

General comments

We thank the editor for their additional assessment and comments. We performed some additional calculations, in particular regarding the possible nitrate coatings effect.

In this context, we would like to point out that increasing the complexity of the aerosol model is not necessarily beneficial for our application, because the typically available measurements do not provide sufficient constraints for the additional parameters. A more complex model would therefore increase the number of poorly constrained parameters, the mathematical complexity, and the computational cost without providing additional meaningful information.

As clearly explained in the manuscript, the retrieved refractive index should be interpreted as a Mie-equivalent quantity. Although it may not correspond exactly to the physical refractive index of the actual, morphologically complex particles, it provides an optically consistent representation of the aerosol for radiative transfer calculations, which is the principal focus of this study. The underlying assumptions, such as spherical particles, idealized size distributions, and internally or externally mixed aerosol components, are commonly employed in current chemical transport and climate models. Expressing our results within this established framework therefore facilitates their interpretation and implementation in such models.

Following this approach, we adopted the simplifying assumptions described in the manuscript, but devoted substantial effort to developing an error-propagation framework that accounts for their associated uncertainties. Within the resulting uncertainties the retrieved properties remain meaningful and useful within these stated bounds, as also acknowledged by the reviewers.

To address the editor's concerns, we performed targeted sensitivity studies, which are described in detail in our responses to the specific comments below. We also added Figure S2 to our supplement and at the end of this response, and added the appropriate references in the manuscript.

1. Particle size distribution (PSD) is missing larger particles

Your retrieval uses two log normal distributions to fit the in situ PSD measurements. Yet the observed PSD (Figure S1) shows clear additional features, including substantial bumps around 160-180 nm and again around 200-250 nm. However, your fitted distribution peaks at ~135-150 nm and has negligible mass above 200 nm. Particles in the 200-400 nm range strongly affect UV scatter and absorption. By omitting these slightly larger particles, you are possibly introducing structural model error.

Required action:

You must perform one or more of the following: (a) refit the PSD to include these larger modes (e.g., a trimodal distribution) and re run the retrieval to quantify how your imaginary refractive index, Angstrom exponent, and derived half lives change; or (b) perform a sensitivity study where you artificially add a plausible third mode (using literature values for wildfire aerosol) and estimate any bias in your retrieved absorption properties. The results must be reported, and the limitations of the current PSD fit must be clearly stated.

We believe the editor is referring to the PSDs in Figure S1 A. The gray lines show PSDs measured over very short time intervals (1 s) and are intended to illustrate measurement noise. In the 10-second (0.1 Hz) averaged PSDs, the measurement noise is considerably less, thereby allowing assessment of in-transect variability of the general PSD shape. The retrieval uses the transect-averaged PSDs shown in Figure S1 B. Because a down-plume series of transect-averaged PSDs is plotted together in Figure S1 B, their individual shapes (dots) and fitted modes (lines) may be difficult to distinguish.

To ensure that the representation of the PSD using a bimodal fit is appropriate, we performed an additional sensitivity calculation using the complete measured PSDs directly, without representing them by bimodal log-normal fits. This test is more directly constrained by the observations than adding a hypothetical third log-normal mode, as proposed by the editor.

The first column of Figure S2 (see below) compares the measured PSD (grey symbols) with the bimodal fit (black line). Blue dashed lines illustrate the shift assumed by our error propagation. The remaining columns of the figure compare normalized extinction and absorption coefficients as well as asymmetry parameter calculated from the measured bins (grey dashed lines) and the bimodal fit (black solid lines). For context, the blue envelope illustrates the effect of the radius perturbation used in our error propagation.

The optical curves obtained from the measured bins and from the bimodal fit are nearly coincident and the differences are at most a few percent and are about an order of magnitude smaller than the effect of the already assumed size uncertainty (blue envelope). We conclude that the observed shoulders and larger-particle bins will not

have a significant impact on our retrieval results. We added a corresponding sentence to the manuscript:

"The bimodal distribution describes the observed PSDs accurately (see Supplement S2)."

2. Neglect of nitrate coated BC and systematic residual structures

Your Figures S4 and S5 show high uncertainty, systematic, wavelength dependent residuals. The upwelling residual changes sign across the spectrum (negative at some wavelengths, positive at others), while the downwelling residual is persistently negative. This pattern is not consistent with a simple BrC/BC mixture and a unimodal or bimodal log normal PSD. It strongly suggests missing physical processes. One such known process is the formation of nitrate aerosols from co-emitted NO2 (which are highly scattering and have a real refractive index >1.55), and their rapid coating on BC cores. Nitrate coated BC particle can yield the sizes observed in the spikes as well as produce a spectral signature that mimic BrC absorption, leading to a potential overestimation of BrC absorption.

Your current error propagation assumes that residuals are unbiased and random; it does not account for systematic structural misfits. Therefore, your uncertainty estimates may be too optimistic, and the retrieved BrC properties could be systematically biased.

Required action:

You must add a quantitative discussion (or sensitivity analysis) estimating the potential bias in your retrieved BrC absorption properties when considering nitrate coated BC. At a minimum, use published values of nitrate optical properties and a simple core shell Mie calculation to estimate the change in single scattering albedo and absorption that would be misattributed to BrC. If you conclude that the bias is smaller than your reported uncertainties, you must state that explicitly. If it is larger, you must revise your conclusions accordingly.

These revisions may require additional data analysis and modelling, but they do not require new field measurements. If you are unable to perform the sensitivity analyses, you must clearly state this as a major limitation in the conclusions and recommend that future work addresses these issues.

The editor's comment about nitrate coating is interesting, as we have not considered this as a possible explanation yet. We therefore performed an additional sensitivity calculation that is now described in a new paragraph in the manuscript:

"Furthermore, we investigated potential impacts of ammonium nitrate (NH_4NO_3) coating on the aerosol optical properties, as reported, e.g., by Yuan et al. (2020). High-resolution

Aerosol Mass Spectrometer measurements (see FIREX-AQ database, Aknan and Chen, 2023) during the three analyzed flights imply (NH_4NO_3) volume fractions of 1.9-2.4%. Assigning all particulate nitrate to NH_4NO_3 gives conservative upper bounds of 2.4-3.3%, with a transect-averaged maximum of 7%. Core-shell Mie calculations using 2.5%, 3.5%, and 7% volume fractions of non-absorbing NH_4NO_3 shells (n=1.55) changed absorption by at most 6% and single-scattering albedo by equal or less than 0.01. As illustrated in Supplement S2, these changes are about an order of magnitude smaller than the changes introduced by the size uncertainties assumed in our error propagation scheme (see Section 3.6) and thus have no significant impact on the presented retrieval results."

The results are also illustrated in Figure S2 below. The optical properties resulting from the core-shell calculations (colored lines) show a deviation from the non-shell calculation (black solid line) that is much smaller than the propagated uncertainties (blue envelope).

We agree and are aware that the wavelength-dependent residual structure and the inconsistencies between up- and downwelling actinic flux observations may point towards unknown model inadequacy. This is why we included the residual in the error propagation as a measure for potential model inadequacy, even though propagating uncertainties in relevant model inputs and CAFS measurement errors should alone describe the uncertainty in case of an adequate model. We believe that this is the best feasible approach and would like to emphasize the following:

1. The propagation of uncertainties from the model input parameters considers correlations accurately by propagating full covariances.
2. The propagation is (probably too) conservative in other aspects:
    a. During the averaging of transects we do not assume statistical noise (no division by n-1 as common for a statistical error-of-mean calculation) but average the uncertainties.
    b. Since all relevant model input parameters and CAFS measurements are associated with errors, the propagation of the residual artificially inflates the retrieval errors, by potentially double counting errors arising from CAFS noise or model inputs.

After all, despite the various simplifications, the model reproduces the measurements well within the specified uncertainties. We are thus confident, that our results are reliable within the specified errors.

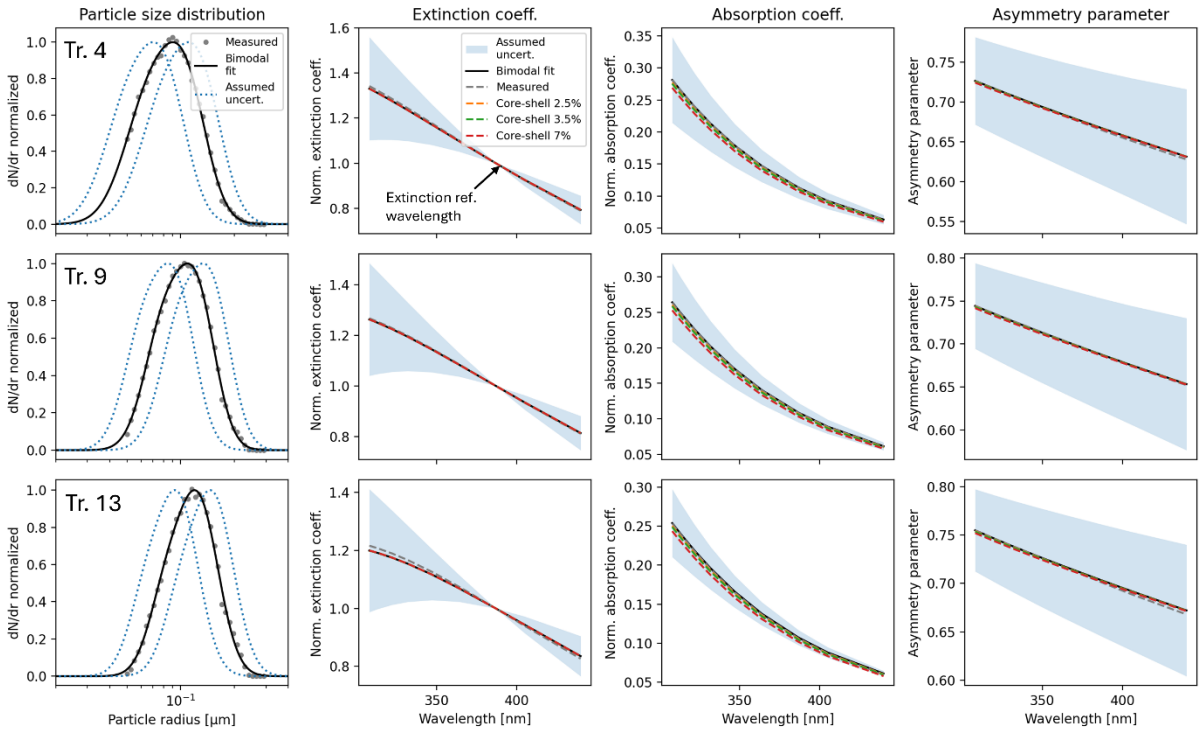


Figure S2. Sensitivity of calculated aerosol optical properties to two aerosol model simplifications: (1) the use of a bimodal PSD instead of the originally measured PSD and (2) NH_4NO_3 shells occupying 2.5 %, 3.5 %, and 7 % of total dry particle volume (based on FIREX-AQ aerosol composition measurements; see main text Section 3.2). Panel rows show three Shady Fire plume transects (4, 9 and 13) representative for different plume ages. The first column of panels shows the originally measured PSDs from the laser aerosol spectrometer (grey dots), the bimodal fit used in the retrieval (black solid line), and the particle radius uncertainty assumed for our error propagation (blue dotted lines). The remaining panels show the optical properties (extinction coefficient, absorption coefficient and asymmetry parameter) for the measured PSD (grey dashed line), the bimodal fit (black solid line), core-shell configurations (colored dashed lines), and the assumed particle radius uncertainty (blue shaded region).