Initial Remarks

We thank the reviewers for their thorough evaluation and the constructive feedback, which helped to clarify our arguments and improve the overall quality of the paper! Please find our answers and corresponding modifications to the manuscript below.

Original reviewer comments are printed in bold

Answers are printed in regular font

Paragraphs literally inserted into the manuscript are printed in italic font.

Review I:

This study "Retrieval of ultra-violet aerosol absorption from radiation measurements in young wildfire plumes" by Tirpitz et al. presents a novel retrieval framework to determine the imaginary refractive index (k) of biomass burning aerosols in the ultraviolet and visible spectral ranges (310-440 nm) using airborne actinic flux measurements from the FIREX-AQ campaign. The application of the VPC (VLIDORT for PhotoChemistry) radiative transfer model to dense wildfire plumes represents a technically advanced and carefully executed effort. The reported k values at 387 nm (0.02–0.03), an absorption Ångström exponent of 4 ± 1, and the identification of a bleaching half-life (~13–17 hours) provide valuable quantitative constraints on the chemical aging of brown carbon. Extending the spectral retrieval down to 310 nm is particularly impactful, as it captures the strongest absorption region of BrC. The authors also provide a transparent error propagation analysis and discuss several limitations of their inversion framework.

Overall, this is a strong and important contribution. The following comments are intended to further strengthen the physical interpretation and robustness of the retrieval.

- **The use of the 1D VPC framework is well justified for plumes that are horizontally extensive. However, the Williams Flats fire reached a maximum AOD of 11.3 at 400 nm. At such high optical depths, horizontal photon transport and 3D radiative effects can become significant even for wide plumes. The manuscript accounts for horizontal inhomogeneity through AOD perturbation within the uncertainty analysis. It would nevertheless strengthen the study to provide a quantitative sensitivity test demonstrating how the retrieved k values respond to enhanced representation of 3D effects, particularly in the densest**

plume regions. For example, would increasing the assumed horizontal variability or inhomogeneity term materially change the inferred k? Clarifying the expected magnitude of potential 3D-induced bias would increase confidence that the retrieval is not compensating for multi-dimensional radiative effects.

We agree that this would be valuable. We experimented with 3D model simulations (such as SHDOM, https://doi.org/10.1175/1520-0469(1998)055<0429:TSHDOM>2.0.CO;2), but estimating the impact of 3D RT effects on our retrievals is challenging, in part because of a lack of input data. Drawing quantitative conclusions is thus difficult within the scope of the presented study. However, as described briefly in the manuscript, we are confident that 3D RT effects are not the limiting factor for the results presented in this paper, based on findings in an earlier study (Tirpitz, 2025, https://doi.org/10.5194/acp-25-1989-2025). We extended chapter 3.1 accordingly, to make this clearer:

For plumes that are several kilometers wide and for solar zenith angles 75 deg, Tirpitz (2025) showed that the application of a 1D radiative transfer models such as VPC, is highly successful: Comparisons between modelled and measured actinic fluxes showed that 3D radiative effects can occasionally be significant for individual data points (10 s temporal resolution) in areas with large extinction gradients (plume edges). However, transect-averaged measurements agreed very well with the model, confirming minimal 3D effects in this approach.

Regarding the reviewer’s comment “However, the Williams Flats fire reached a maximum AOD of 11.3 at 400 nm. At such high optical depths, horizontal photon transport and 3D radiative effects can become significant even for wide plumes”: Our experience is different: In the (near-)UV the absorption is relatively strong. This is in our favor, as (in contrast to scattering), absorption radically reduces the horizontal sensitivity range of models and observations. As discussed in the paper (L336), the largest impact of 3D radiative effects is expected in plumes of intermediate AODs. Furthermore, in a plume of AOD = 11.3, there is so little light, that the retrieval uncertainty is typically dominated by the actinic flux measurements (see e.g. Figure S1, Transects 14,15,16,17).

1. **The retrieval assumes homogeneous spherical particles with uniform composition. Wildfire smoke, however, frequently consists of internally mixed particles (e.g., BC cores with organic coatings) and potentially non-spherical morphologies. Previous studies have demonstrated that moving from homogeneous-sphere assumptions to more physically representative mixing-**

state treatments (e.g., core-shell configurations) can lead to substantial differences in simulated radiative quantities (Tiwari et al., 2023). While a full reanalysis under a core-shell or fractal framework may be computationally intensive, it would be helpful to include a qualitative discussion of the expected direction and magnitude of bias introduced by the homogeneous-sphere assumption. Because particle morphology and mixing state influence not only absorption efficiency but also the scattering phase function and asymmetry parameter (g), which are essential for accurate radiative transfer calculations, it would be helpful to clarify whether part of the retrieved spectral dependence of k could reflect structural assumptions rather than purely chemical absorption changes.

We use “Mie-equivalent” values in our study, as these are most relevant for satellite retrievals and applications in CTMs which also use classical Mie models.

We would also like to note that morphology effects such as lensing are strongest, when there is a transparent shell around a highly absorbing core (such as water around black carbon). In our case, the air is dry and condensation is unlikely. The most likely scenario is a BC core with BrC shell. Since in the UV-Vis, BrC and BC absorption are of similar order of magnitude, the impact of the mixing state is expected to be relatively weak.

We performed sensitivity studies that show an estimate of the magnitude and direction of particles that have core-shell morphology or are externally mixed, compared to that our homogeneous sphere assumption:

Comparison to homogenous particle Mie calculations	**Bias in imag. RI magnitude (387 nm)**	**Bias in imag RI angstrom coeff.**
Core-shell	10%	-0.15
Externally mixed	-10%	0.3

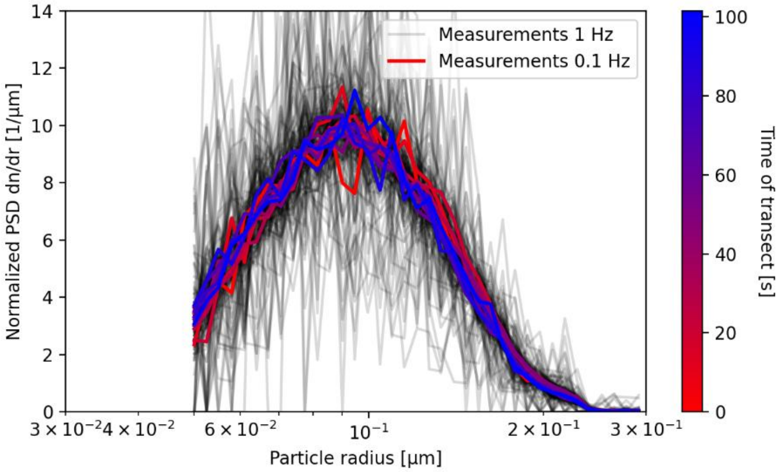
To address this issue in the manuscript we added a following paragraph in Section 3.2 “Representation of plume aerosol”:

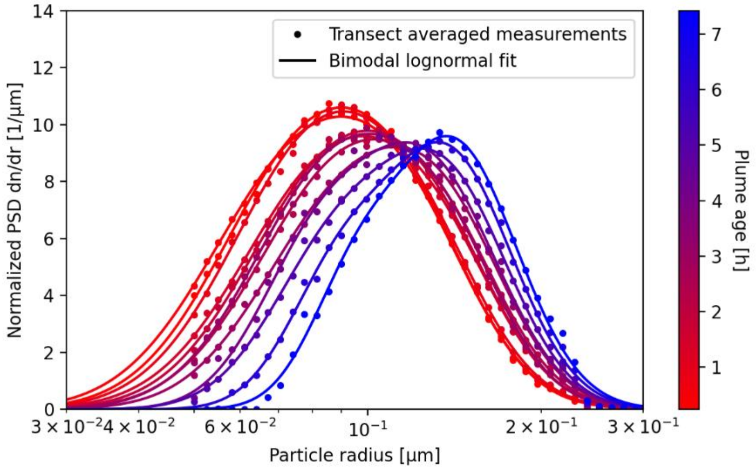
It should be noted that the retrieved imaginary RI represents a "Mie equivalent" value. RI values for actual particles may differ due to specific mixing state and particle morphology. At the same time, this Mie-equivalent RI value is "radiatively correct" in the sense that the Mie code will yield optical bulk properties (extinction cross-section, SSA, scattering phase function) that are close to reality. Furthermore, the homogeneous sphere representation underlying the Mie-equivalent approach is *often used in satellite retrievals and chemical transport models. This makes our results directly transferrable.*

Nevertheless, we performed sensitivity studies for more complex particle representations, to estimate direction and approximate magnitude for the biases induced by our homogeneous sphere assumption. For particles with a core-shell morphology (core is BC, shell is BrC, BrC/BC volume ratio and PSD same as in the homogenous case), our retrieval overestimates the particle average imaginary RI by ~10% and underestimates its Angstrom coefficient by ~0.15. For particles that are externally mixed (pure BC particles, pure BrC particles), our retrieval underestimates the particle average imaginary RI by ~10% and overestimates its Angstrom coefficient by 0.3. These numbers are within the uncertainties of our retrieval.

2. **The particle size distribution is constrained by in situ measurements and fitted to a bimodal lognormal distribution, which is commendable. However, the retrieval uses transect-averaged PSD parameters. In young wildfire plumes, rapid coagulation and condensation can modify particle sizes on relatively short timescales. If within-transect variability or systematic growth with plume age is not fully represented, some of the radiative impact of size evolution could mathematically project onto the retrieved imaginary refractive index.**

   We had to average PSDs to achieve reasonable noise levels considering the very short measurement times inside the plume (see upper Figure below). Based on 10s-averaged observations we did not observe any significant in-transect variability (upper Figure). The PSD changes systematically with plume age (see lower Figure). This is accounted for in the retrieval by using the PSD of the current transect and thus the current plume age.
   We added these figures and a describing text to the supplement to provide support data addressing this issue.





It would therefore be helpful to include a sensitivity analysis in which PSD parameters are perturbed within observed in situ ranges to assess the stability of k(λ).

Please note that we basically perform these sensitivity analyses in the context of the error propagation. The uncertainty in the in situ measured PSD is linearly propagated through the retrieval and thus directly accounted for.

Additionally, clarifying whether PSD parameters show systematic dependence on plume age would help ensure that the inferred bleaching signal is not influenced by size evolution effects.

As shown in the figures above, it influences inferred bleaching signals, but this is accounted for, as explained above. Maintaining a constant PSD for all retrievals yields a seemingly even faster temporal decay in the imaginary RI.

3. **Section 4.1 notes that retrieved VAC and SSA show significantly less variability than in situ measurements, which is attributed to spatial averaging inherent in remote sensing retrievals. This is a plausible explanation. However, the discrepancy appears substantial. It would be useful to present residual diagnostics showing where the radiative transfer model deviates most strongly from the observed actinic flux. For example: Do residuals increase in regions where in situ variability is highest? Is there any systematic misfit in dense or highly heterogeneous plume segments? Such diagnostics would help clarify whether the reduced variability reflects representativeness differences or potential over-constraining of the inversion (e.g., fixed real refractive index and parameterized PSD).**

   We believe the reviewer is referring to Section 4.3 and Figure 7, where we compare VACs for the entire Shady fire and the shading indicated the transect-to transect variability, which is large for the in-situ measurements.

   We recognize that our explanation of this variability (line 437) may have been misunderstood and have reformulated this sentence to: "*This indicates that in situ measurements averaged over a single plume transect may suffer from biases due to variations associated with the specific height at which the plume was probed and variability in the plume geometry during the transect. Therefore, these values are not necessarily representative for the entire plume cross-section.*"

   We also want to point out that the residual analysis of the retrieval requested by the reviewer is implicitly shown as the retrieval errors in Figure 5E, as the magnitude of the residual is one of the main components of this error.

   We are confident that transect-averaged results are not impacted by over-constraining the retrievals for different reasons: The PSD variability between transects is small (see lower PSD figure above). The in-transect variability of our retrieval results can be high (Figure 5, panel B+C), which indicates that variability is in fact captured. In addition, the retrieval reacts to small scale variability, speaking strongly against it being over-constrained. We also believe that over-constraining the retrieval would increase variability among transects. For instance, a fixed real refractive index and a rigid PSD when real variations in these parameters are present, will result in compensatory variations in the imaginary refractive index.

4. **The retrieval constrains the spectral dependence of the imaginary refractive index using a two-parameter Ångström-type power law (k_0, α_k). While this parameterization is widely used, laboratory and field studies have sometimes**

reported deviations from strict power-law behavior in the near-UV, particularly below ~350 nm. Could the authors comment on the sensitivity of the retrieved α_k and bleaching timescale to this spectral constraint? A brief discussion or limited sensitivity test allowing small deviations from the power-law form would help clarify the robustness of the inferred aging signal.

In fact, we initially started by retrieving a second-order angstrom dependence:

$$ln\,(k)\ =ln\,(k_0)\ +\ \alpha_1\,ln\left(\frac{\lambda_0}{\lambda}\right)\ +\alpha_2\left(\frac{\lambda_0}{\lambda}\right)$$

We found that, at least for the limited spectral range investigated in the paper, the single angstrom dependence is sufficient. It is also beneficial to simplify interpretation, consolidation and visualization of the results. Adding the second order degree of freedom only has a small impact on the retrieval results. We added a sentence to the manuscript to clarify this:

“*Adding higher order terms to equation 2 had only a small impact and was thus not implemented.*”

5. **Table 1 indicates differences in fuel type and combustion characteristics across the sampled fires. The manuscript reports similar bleaching timescales (13–17 hours) and notes no statistically significant differences among fires. It would be interesting to show fire-specific half-life fits (with uncertainty ranges) to clarify whether the apparent similarity reflects true physical consistency or limited statistics. A short discussion on whether fuel type, combustion efficiency, or initial chemical composition might influence bleaching rates would enhance the interpretation.**

This is an interesting suggestion. Unfortunately, we do not have enough data to derive statistically significant fire-to-fire variability and its relationship to detailed fire characteristics. Instead, we use the fact that the three fires shown in Figure 8 occurred in similar ecosystems to provide a statistically significant timescales over a larger duration than possible with each individual fire.

We added a clarifying statement in the caption of Figure 8: *“The dependence of the volume absorption cross-section at 300 nm on plume age is shown for three fires, which burned under similar conditions in similar ecosystems (Pearson correlation coefficient 0.5)”*

6. **A brief discussion of how vertical variability observed during aircraft sampling is represented (or averaged) in the 1D radiative transfer framework would further clarify the robustness of the inversion.**

   This is explained in detail in Section 3.5 (Fixed model inputs). Extinction is vertically resolved using measured Lidar profiles, while aerosol properties are vertically constant (in situ observations, literature values). Any inaccuracies in this description will be reflected in the retrieval error.

Review II:

In this work the authors made use of airborne actinic flux spectra taken by the Charged-coupled device Actinic Flux Spectroradiometer (CAFS) during the 2019 FIREX-AQ campaign over the NW USA. These observations were used in combination with the VLIDORT for PhotoChemistry (VPC) radiative transfer model (RTM), capable of actinic flex spectra modelling, to infer smoke absorption properties. Their approach involves constraining the VPC model with ancillary information from FIREX-AQ measurements in combination with available relevant information on particle size distribution and vertical distribution. The core of the analysis consists of varying the RTM a-priori aerosol absorption properties so that coincident CAFS actinic flux measurements are matched by the calculations. The assumed spectral imaginary component of the refractive index required to explain the CAFS actinic flux observations, is deemed to be an accurate representation of the actual aerosol absorption properties. This inversion approach is applied to three (out of 90) fires observed during the FIREX-AQ 2019 campaign. The authors have carried out a well-documented radiative transfer inversion exercise that yields imaginary refractive index and single scattering albedo spectra. The reported results can, in turn, be used to evaluate other measurements of aerosol absorption parameters collected using both ground based as well as satellite measurements.

This is a very important contribution. It provides valuable information on micro-physical and macro-physical properties of carbonaceous aerosols specific to the type of burning material. The paper is very well written. Results are clearly presented and the figures are for the most part adequate.

A couple of minor comments:

The three-panel figure 1 looks incomplete to this reviewer. The y-axis label in the top three-panel figure is missing, and the size of the actual figures is quite small. One can assume the y-axis title is the same as in the center figure, but it is best to directly

specify it. The cartoon representation (bottom figure) seems unnecessary. The distinct spectral contrast in the UV and visible regions can be described in the text, giving up more space for a fully documented 2-panel figure 1.

Figure 1 is intended to help the reader understand the scientific topic of this manuscript and the underlying concepts, not as quantitative result of our study. We understand the concern about the missing axes in the top panel (we initially had the same concern) but adding them would make the figure unwieldy and complex.

Yes, we could omit panel C, but we found that this figure is the most effective way to explain to the reader that BrC absorption dominates radiative transfer in the UV. This is an important concept for our study, and ensuring a reader clearly understands this, merits a little space in the paper.

We will ensure that the figure will be displayed large enough in the final version of the paper, so that it can be easily read.

My second comment, also a minor one, is about the conclusion section of the paper. I suggest removing the sub-section titles in the conclusions part of the manuscript. This section should consist of a set of interconnected paragraphs that offer a full comprehensive summary of the work presented.

We thank the reviewer for this comment. We have removed the subtitles and updated the paragraph transitions.