Response to Reviewer 2

We thank the reviewer for their feedback and constructive criticism, which have helped us significantly improve our manuscript. We have taken care to address each comment with a direct response. The text from your comments are shown in black and our responses are shown in blue. Responses include the manuscript text that was changed, removed, or added.

This manuscript presents a methodology to compare and reconcile aerosol observations from various platforms, so called ISARA algorithm, developed for the ACTIVATE mission data. It provides a useful and rather comprehensive example of attempting to achieve closure between diverse in-situ and remote sensing measurements. The study clearly addresses a complex problem and contributes relevant information for aerosol measurement harmonization. However, it would benefit from clarifying its broader motivation, better articulating its limitations, and improving the language and structure in some sections.

My main concern is that the manuscript abstract, and introduction do not seem to precisely describe the motivation of the study. The introduction frames the problem mainly as a data closure challenge, potentially giving the impression that a general solution (i.e., ISARA) will be offered. For example the sentence "Despite the important findings from these studies, systematic and streamlined closure of aerosol data sets has not been yet achieved." However, the ISARA algorithm appears heavily tuned to specific conditions, relying on a priori information about aerosol composition, size, and shape, additionally being very limited in the atmospheric and aerosol conditions where it can be applied. This is perfectly understandable, but a clearer explanation of how broadly applicable the algorithm is would strengthen the outcome. The introduction could therefore more broadly discuss the complexity of in-situ vs remote-sensing aerosol validations, compilations and simulations – as for setting the scheme. I hope that authors consider this, and could slightly streamline the introduction and structure of the paper to better reflect the content.

In summary, this manuscript addresses a complex and relevant topic in atmospheric aerosol measurement by providing a detailed example of integrating multiple observation platforms. The methodology is simple but thoroughly described, and the analysis is well presented, but the applicability and limitations of the ISARA algorithm require clearer discussion. The manuscript would benefit from a refined motivation and improved language in certain sections. I recommend minor revisions before acceptance at AMT.

We thank the reviewer for their thoughtful and constructive feedback. In response to the concerns raised, we have revised the abstract and introduction to better articulate the overall motivation of the study. Specifically, we now clarify that this work focuses on assessing consistency across platforms as a necessary step toward achieving rigorous external closure, rather than offering a universal solution to the closure problem. We also explicitly acknowledge the limitations of the ISARA framework, including its dependence on assumptions about particle shape, composition, and the applicability to specific aerosol regimes. These clarifications are intended to better align the manuscript's framing with its scope and contributions. We believe these revisions strengthen the manuscript and more accurately convey the goals and context of our analysis.

Minor Rather Technical Comments: Abstract:

- L6: Suggest removing "aircraft" as the methodology can apply to other in-situ data platforms as well. Or can it? Please consider this also when writing introduction. Thank you for the comment. We do agree that the framework offers a great deal of flexibility in the in-situ platform used. We have removed "aircraft" from this sentence and made subsequent changes within the introduction as well.

Introduction:

- L33: Remove the word "parameter". Thank you for the comment. We have addressed this error. We also removed the example to discuss specific parameterizations later in the text.

- L51: Consider adding 1–2 sentences summarizing key findings from past studies for context. Thank you for the comment. We have added the following sentences to the text to provide more information on the findings of a few of these

studies: “These studies consistently find that the extinction and backscatter coefficients derived from in-situ instruments are systematically low compared to those derived from HSRL-2.” and “In particular, Pistone et al. (2019) found poor agreement between in-situ- and RSP- derived total SSA.”.

- L52: Clarify “these instrument data”. Which instruments are being referred to? Thank you for the comment. We have clarified the statement to be “... remote sensing data’

- L57: Suggest rephrasing to “...using a Nafion membrane dryer in the sampling line”. Thank you for the comment. The text has been altered according to the comment and is now: “...using a Nafion dryer or heating in the sampling line”.

- L86: Check for a possible extra “is”. Thank you for the comment. The extra is has been removed from this sentence.

- L91: Sentence structure is unclear-both content and grammar could be improved. Also, please clarify if the methodology is expected to be broadly applicable or limited to specific platforms and conditions. Thank you for the comment. The methods described in this paper are open source and flexible due to using MOPSMAP to handle the optical properties. The sentence has been changed to be: “While the current study focuses on the consistency analysis between in-situ- and remote sensing-derived aerosol properties of the more common spherical aerosol particles, it is hoped that the framework described in this study serves as an open source foundation that can be easily expanded and used to fully understand the information train between all manner of measurements and therefore enable systematic closure of field campaign aerosol data.”

Measurements:

- L198–202: Some grammar and sentence construction issues—please revise for clarity. Also, consider using micrometers (µm) instead of nanometers (nm) for >1 µm sizes. Thank you for the comment. We have updated the units to µm where there weren’t ranges going into the nm scale or significant figures would require multiple digits after the decimal place. We have clarified the sentence in question, which now is “The effective upper size cut is $D = 5$ µm for all 2020 data.”

- L202: Nephelometers and absorption measurements are introduced here without prior explanation. Reorganizing or cross-referencing earlier sections may improve flow. Thank you for the comment. We have added the Section 2.1 to provide a primer on particle properties and general background for these measurements.

- L213: Was the PSAP measurement also conducted at <40% RH? Please specify. Thank you for the comment. The PSAP sample stream is dried by heating the optical block to 35°C. The text has been updated to include this sentence.

- L218: Replace “variety of errors” with a more descriptive phrase identifying specific artifacts or correction needs. Thank you for the comment. We have replaced “variety of errors” with “transmittance and flow errors”.

- L223: How much data was excluded by the >1 Mm^{-1} cutoff for scattering and absorption? Justify the threshold. Thank you for the comment. This comment led to us evaluating the 1 Mm^{-1} and determining that 0 was a better value for absorption. This yielded a 10x increase in the data available for retrieval. This threshold was changed accordingly to 0 Mm^{-1}. The results and conclusions have been updated accordingly.

- L245: Clarify what is meant by "most useful analysis" of profiles extending >1 km. Thank you for the comment. The profiles are most useful for comparison with the RSP-derived products. We have added the following text to clarify this information: "As such, the external consistency analysis is most useful from vertical profiles where the in-situ platform samples the column of air above an arbitrary ground point. Vertical profiles where the extent is more than 1 km are most useful for comparing with the column-averaged aerosol particle properties derived from the RSP-measurements.".

- L257: Consider rephrasing "1-second data" as "native time resolution data" for clarity. Thank you for the comment. We tried to clarify this with the following text "The data that are in their native resolution are averaged to 45 seconds using the NASA standard merging tool."

- L282, L286, L334: "ACTIVATE region" should be defined more precisely—preferably with coordinates or a map. Also specify which areas or conditions were excluded and why. Thank you for the comment. We have clarified the region that ACTIVATE was bounded by within the text. The following sentence was added to Section 2.2: "The extent of the North Atlantic region that was sample during ACTIVATE was within bounds of 58–78°W and 28–42°N.". We have described the cloud filtering process and the focus on when the Falcon (i.e., the in-situ platform) was performing vertical profiles in coordination with the King Air. It is outside the scope of this work to describe the methodologies that guided the ACTIVATE mission.

- L289: Clarify "mid-point particle diameter." Why is geometric mean diameter not used? Mid-point particle diameter is synonymous with geometric mean diameter. The text has been clarified as follows: "Note that this process uses the mid-point (i.e., the geometric mean) particle diameters from each SMPS and LAS channel.".

- L285–295: The assumption of sulfate-only aerosol seems oversimplified. Given the limited RRI range and the potential presence of organics or other compounds, the assumptions behind the ISARA-derived optical properties need better justification. Also, specify how wavelength conversions were done and what values of AAE or SAE were used. Thank you for the comment. We agree that the limited RRI does limit the scope of this study, however, looking at Li et al. (2023), it would appear that most mixtures of aerosol particles will have an apparent real refractive index (RRI) that falls in the 1.51–1.55 range. As such we have updated the text with the following: "Furthermore, external mixtures of many aerosol species have an apparent RRI that falls between 1.5 and 1.58 (Li et al., 2023). If an external mixture of aerosols is dominated by a RRI outside of this range, it is likely that the assumptions of a spectrally flat CRI and sphericity are not longer valid.". Additionally, AAE and SAE were not used. We calculate the coefficients at their native measurements. For example, dry scattering coefficients were calculated at 450, 550, and 700 nm wavelengths and dry absorption coefficients were calculated at 470, 532, and 660 nm wavelengths. The final ambient properties were again recalculated using MOPSMAP at each of the HSRL-2 wavelengths.

- L326: Missing preposition—please review. Thank you for the comment. This sentence has been changed to the following: "After calculating the set of forward-modeled $C_{\mathrm{scat,wet}}$, we use γ to derive the "measured" $C_{\mathrm{scat,wet}}$ (i.e., $C_{\mathrm{scat,RH=80}}$).".

- L334: Again, clarify the threshold inconsistency between L223 and here. How much absorption data were actually usable above 1 Mm^{-1}? Thank you for the comment. As discussed in the response above, we have re-evaluated the use of 1 Mm^{-1} minimum threshold. Additionally, this sentence was not directly connected to the figure so it has been corrected as follows: "It is observed that aerosol particles in the ACTIVATE region generally have low absorption (IRI $\leq$ 0.01) and low hygroscopicity ($\kappa \leq 0.1$).".

– L615: The manuscript states that the region was chosen for “diversity of aerosol and meteorological conditions,” yet the assumptions made to enable closure seem to contradict this. Consider rephrasing or qualifying this claim. Thank you for the comment. We agree that as written this is somewhat contradictory. We have attempted to resolve this contradiction by focusing on the large volume of data available. The text has been changed as follows: “...large volume of statistically-rich aerosol measurements collected over three years of operations. Although the ACTIVATE region does feature a variety of aerosol and meteorological conditions, many of the ACTIVATE missions were carried out in cloud-free conditions without detectable influence from dust or smoke, making the data set well-suited for the consistency analysis performed in this study.”.