Response to Reviewer 3

We thank the reviewer for their feedback and constructive criticism, which have helped us significantly improve our manuscript. We have taken care to respond directly to each comment. The text of your comments is shown in black, and our responses are shown in blue. The responses include the manuscript text that was changed, removed, or added.

The paper by Dmitrovic et al. describes a Python toolkit (ISARA) aimed at achieving closure between lidar and polarimeter observations of ambient aerosol properties on one hand, and in-situ measurements of the PSD, scattering and absorption on the other hand, which are limited by drying and by inlet effects. The toolkit is validated through in-situ observations (internal consistency), simulations (synthetic consistency) and remote sensing observations (external consistency). It is my feeling that the stated goals of the paper are of high importance, but that they are not sufficiently developed beyond the technical stage to be proposed for a publication. I would invite the authors to consider a more scientific and less technical approach in writing this paper, and to allow the time and the effort that this can represent at this stage I suggest to REJECT the manuscript. I will be willing to help with reviewing a revised manuscript in case the the below MAJOR points are addressed in full.

Thank you for the comment. We agree that the goals of the paper are of high importance. We also agree that the science focus of the paper needed some expansion. We therefore added a detailed analysis of more aerosol properties that was not described in the previous draft, including comparisons against RSP-derived coarse-mode properties that have never been evaluated in previous studies. Since our results show good agreement to within measurement uncertainties compared against advanced remote sensors (lidar and polarimeter observations) we do not agree that the paper is not developed past the technical stage. We further feel that the technical approach is an important development on its own, since it is a foundation that can be used to directly replicate our findings using the code and data provided. Although our results already show significant scientific promise, they can also be readily improved and expanded upon in the future. We show consistency between remote and in situ aerosol products for several ambient aerosol properties that have not been evaluated by previous studies, and we indicate which properties are not consistent. We present a comprehensive analysis using spherical particle models. It is outside the scope of this paper to perform additional analyses of more complex particle types such as dust and smoke since they require us to use modeling of non-spherical and heterogeneous particle shapes and to revisit many of the assumptions made in the remote sensing and in-situ retrievals themselves. Future work will involve demonstrating how we can leverage the additional capability of libraries like MOPSMAP to evaluate these more complex particle types. We have made extensive revisions of the manuscript to address the scientific shortcomings and to address each of your specific comments below. As such, we feel that the paper should not be rejected and we welcome the reviewer to reconsider our revisions.

MAJOR POINTS:

1. The paper takes a technical stance to the task, e.g. insisting on unnecessary details such as the use of the python programming language and the MOPSMAP scattering code. I would invite to present an algorithm, and to discuss its assumptions, limitations and uncertainties, rather than a piece of software. Concerning the scattering calculations, given that the authors only assume spherical particles, MOPSMAP is only a library allowing Mie scattering calculations and any other Mie scattering code would achieve the same results, therefore the scientific approach is to indicate that the framework of Mie theory is being used, and not to indicate which scattering library is used. The usage of the MOPSMAP code could most probably be of interest in the case of non-spherical scattering, but non-spherical scattering is not envisaged here.
   Thank you for the comment. As discussed above, we agree that there needed to be more description of the science behind aerosol properties. The structure of the paper has been changed accordingly. We have added Section 2.1 to add more scientific background to our technical description. In the revised draft, we now clearly explain the assumptions, limitations, and uncertainties associated with this method. Furthermore, the analysis performed in this study can be easily replicated by accessing the software and data products associated with this study, which we believe is important in its own right, since we intend ISARA to be used as an open source toolkit for field campaign scientists to use.

2. There has been previous literature attempting the same task, and this needs to be accounted for and mentioned. I bring here for example the IRRA approach of Tsekeri et al (AMT, 2017). I am sure that by searching the literature the authors can also find other references on the same topic. The authors should explain how their paper fits within the existing

literature, and how similar or different it is from other approaches, indicating pros and cons of each. Note that "previous works" are also cited at line 547, but no references are given. Thank you for the comment. We have added more detail in the introduction and in the methods section that indicate how this work is a successor to previous works. While Tsekeri et al. (2017) did do a retrieval algorithm using in-situ dry measurements and probes, their algorithm requires the lidar data as part of this algorithm. This is distinctly different from the approach taken in this work that does the retrieval independently of the lidar measurements and then compares the aerosol properties afterward. We have made note in several places through the text that the coarse-mode aerosol properties are prescribed and the size-distribution of the coarse-mode is taken from wing mounted probe measurements and cited studies that have done analysis of coarse-mode aerosols with wing mounted probes.

3. The stated goal is a "rigorous external closure" (see line 4). I find this to be a real contradiction with the state-of-the-art achieved by the paper itself, given that it is a "preliminary consistency" and a "preliminary effort" (see lines 19 and 72). This may question whether more work is needed before a paper is submitted to AMT. The sentence at lines 546-548 seems to confirm this impression. Thank you for the comment. We have removed the word "preliminary" from this line since we have added several novel comparisons in the revised draft. These results agree within the uncertainties of the measurements. This sentence is now written as "While this study focuses on spherical, sulfate-dominated aerosol mixtures, its overall success demonstrates that ISARA has the potential to support systematic and streamlined closure of aerosol datasets across diverse field campaigns and aerosol regimes.". The focus of this work is to prove the functionality of ISARA under conditions that are generally realistic but relatively ideal in terms of ignoring the possible presence of non-spherical aerosols within the marine environment. Furthermore, we state that rigorous external closure over a wide range of all possible marine and terrestrial scenarios will require future studies and changes to some of the assumptions made during this study, but will not require a fundamental change to the underlying software (i.e., ISARA).

4. The computations are done for a specific atmospheric scenario only, with a mixture of sulfate and organics in the fine mode and sea salt in the coarse mode (lines 104-109 with a clear mention that "this will limit the scope of this study"). Whereas this is fine in itself, it removes generality away from the ISARA approach and from its stated goal of a generally applicable approach. It is important to state the limited scope from the onset of the paper, rather than add the limitation further down the line. Moreover, given that ACTIVATE operated also in Bermuda (line 136) why is the influence of dust aerosols not also investigated? Thank you for the comment. We have added the following text to the abstract to list the assumptions used in this study more clearly from the start: "While this study focuses on spherical, sulfate-dominated aerosol mixtures, its overall success demonstrates that ISARA has the potential to support systematic and streamlined closure of aerosol datasets across diverse field campaigns and aerosol regimes.". Furthermore, while dust may have been observed during ACTIVATE, it is outside the scope of this paper to analyze these more complex aerosol types until we are confident in the comparisons using more typical aerosol types. We have reiterated this point throughout the text.

5. After clarifying the issues affecting in-situ observations, the authors claim (lines 89-90) that "the ISARA attempts to overcome these limitations by estimating the contribution of coarse-mode particles". It is unclear how this can be achieved, and I would state that this is not possible without a dedicated measurement for the coarse particles (e.g. using an open-path OPC). A CDP is later mentioned at lines 226-234, therefore the problem may perhaps simply be of being explicit and honest from the onset about the fact of using a dedicated measurement for the coarse mode. Thank you for the comment. We agree that this text was misleading and have changed the text to clarify that we are using dedicated probe measurements. In this case, we preferentially used the CAS, but relied on the CDP or FCDP where the CAS was not available.

6. Figure 9: ISARA shows underestimation. Figure 10: low level of closure. This does not look good for the ISARA method Thank you for the comment. We do not feel that this underestimation is a critical weakness because the comparison shown yields results within a reasonable uncertainty. We have made this point evident throughout the text by adding

the corresponding retrieval uncertainties and standard deviations where possible. This algorithm represents a solid beginning, and future improvements are likely possible.

7. The sentence “a potential reason for detecting low-RH aerosol is the presence of smoke from fires” (lines 588-589) is in contradiction with the assumed marine environment, but is also presented in a way that makes it appear unjustified. Please note that water vapour is a combustion product, hence fires should contribute to increasing absolute humidity and not to decrease it. The whole narrative at lines 590+ appears incoherent: note that figure 13 shows fires and not smoke aerosols as stated here. It is suprising also to read that aerosols “are becoming drier due to colder temperatures” given that lowering the temperature (with constant WVMR) has the effect of increasing the RH. I agree with the sentence on line 593 that “further work will be done” and I feel that it must be done before re-submitting the paper. We agree that this text was erroneous and we have removed it. We now focus attention to the presence of winter conditions, which is far more important than any amount of water vapor generated from combustion. This is especially true because no smoke aerosols were detected in the profile and because the profile was located far from any source of smoke. If we expanded the scope of this study to include more atypical particles, the paper would be incredibly long and unfocused. Instead, we aim to demonstrate that this method works well for spherical aerosols found in marine environments, and that the overall approach is also useful for future research.

8. Overall good agreement (line 602) feels an unjustified statement here. The same applies to the “successful retrieval” (line 622). The evidence is not so clear-cut. Thank you for the comment. After performing extensive revisions throughout the paper, we feel that overall these statements are justified but have made it a point to indicate which properties were shown to be consistent and which were not.

MINOR COMMENTS:

5. “In-situ instruments cannot efficiently sample coarse-mode particles due to limitations in the inlet cutoff diameter” (line 65). This statement is incorrect given that there are several open-path airborne instruments such as the CDP, the CAPS, etc. that overcome these limitations (initially developed for cloud particles and later extended to use in aerosol layers). It is well-known that the FAAM research aircraft has successfully sampled coarse and giant dust particles up to 300 um diameter with such probes (see e.g. Ryder et al, ACP, 2015; Marenco et al, ACP, 2018). Thank you for the comment. We have changed the text throughout the paper to clarify that we are using the coarse-mode measurements in the same way as the sources you have cited.

6. The definition of the fine-mode (0.09-1 um dry diameter) and coarse-mode (> 1 um ambient diameter) regimes (lines 83-84) is weird given that there could be an overlapping zone between fine and coarse (particles with dry diameter < 1 um and ambient diameter > 1 um would be in both regimes). I would suggest to consistently refer to either the dry or the ambient diameter for discriminating the two modes. Thank you for the comment. This was an error on our part. We have changed the word dry diameter to ambient diameter.

7. Line 6: the symbol κ is undefined. At the end of page 14, the relationship between κ and f(RH) must be explained. Thank you for the comment. We have decided to remove the explicit parameterization of κ from the abstract and moved it to the introduction, which is expanded further in Section 2.4. In this section, we explain that is related to scattering and absorption coefficients through the growth factor equation abd relative humidity.

8. Joint flights (line 135): it is unclear which two airplanes performed joint flights. Thank you for the comment. Because we address this directly in the following paragraph, we have removed the word “joint” from this sentence.

9. “The novel vertically-resolved aerosol particle number concentration” (line 184): I suggest to add the word “estimate” to this statements, given that number concentration from lidar is estimated based on assumptions. Thank you for the comment. We have added the “estimate” to this sentence.

10. Inlet cutoff at 5,000 nm (line 199): indicate how the cutoff diameter was determined. Thank you for the comment. This information was determined by using ground station measurements collocated to the aircraft measurements. Although it is outside the scope of this paper to describe this process, we have cited McNaughton et al. (2007) that evaluated this cutoff diameter.

11. For random variables such as the scale factor (line 365), the κ (line 367) and IRI (line 370), the random distribution used must be given. For the scale factor, you can observe the random distribution in Fig. 5. For the random distributions of and IRI you can observe the random distribution in Fig. 5. We feel that showing the random distributions of these variables would require an additional separate figure that is unnecessary given the two figures already present with this information embedded.

12. Nephelometers (line 202) are being mentioned before being introduced (line 213 and following). I suggest that the full instrument set must be introduced before discussing their details such as installing a cyclone. Thank you for the comment. We have added Section 2.1 to address this concern.

13. Equation 1 using RH of 20 and 80% is inconsistent with having observations at 40 and 85% (line 215). Please clarify. We have changed the text and redefined f(RH) more clearly in Eq. 15. The measurement of humidified scattering was done at 85% RH, however, we use γ to calculate f(RH) for the 80/40% RH

14. Line 248: “the methods standardized by the merging tool”. It is unclear what the authors refer to. Thank you for the comment. We agree that this text was ambiguous. We have changed the sentence to be: “Data in their native resolution are averaged to 45 seconds using the NASA merging tool.”. A description of how the merging tool works is found in https://www-air.larc.nasa.gov/missions/etc/onlinemergedoc.pdf, which is cited in the previous sentence of the text.

15. Symbols: there is an unclear use of symbols N_a and n_0 (seeming to refer to the same variable). Also, in lines 293 and 294 C_calc refers to different variables, hence I suggest using different symbols. Thank you for the comment. We have corrected our terminology throughout the paper to make the distinction between N and n^o clear. We have also corrected the C_{calc} term to have the “abs” or “scat” subscript where appropriate.

16. Equations 3 and 7: the bounds of the integral should be log(Dmin) and log(D_max), and not dlog(Dmin) and dlog(Dmax). Thank you for the comment. We have corrected this error.

17. Line 270: remote sensing sensors are normally considered more sensitive to the surface area and not the volume. Please correct. Thank you for the comment. As another reviewer pointed out, this statement was not fully correct. The sensitivity of the remote sensor to surface area or volume depends on the observational wavelength. We have added more detail with the following text: “ In general, remote sensors are not as sensitive to particle number concentration as they are to particle surface area and volume concentrations. For particles larger than the remote sensor’s observing wavelength, the remote sensor is most sensitive to particle cross-sectional area (i.e., surface area concentration). For particles smaller than the observing wavelength, the remote sensor is most sensitive to volume concentration.”.

18. Stitched data (line 279): this must be documented. Thank you for the comment. We have explicitly documented this step through the following text: “After this step, the SMPS and LAS size distributions are stitched at a diameter of 94 nm, which is the upper bound of the size-range by the SMPS and the lower bound of the LAS size-range.”.

19. Full range of particle sizes (line 279): this is incompatible with the inlet cutoff. Please clarify. Thank you for the comment. We felt that this sentence was redundant and unclear, so we have removed it. This information is discussed directly above in the previous paragraph.

20. Figure 1 caption: I suggest to use percentiles instead of minimum and maximum to reduce the influence of outliers. Thank you for the comment. We have changed Figure 1 to reflect the 10th and 90th percentiles and updated the corresponding caption.

21. CRI averaging (line 291): it is unclear over which dataset the averaging is being done. Thank you for the comment. In the following sentence, we now state: “For a CRI to be valid for averaging, all three of the computed scattering coefficients must be within 20% of the corresponding measured dry scattering coefficient $\left(\frac{|C_{\mathrm{scat,calc}}-C_{\mathrm{scat,RH=40}}|}{C_{\mathrm{scat,RH=40}}} < 0.2\right)$ and all three of the calculated absorption coefficients must be within $1\,\mathrm{Mm}^{-1}$ of the measured absorption coefficients $\left(|C_{\mathrm{abs,calc}} - C_{\mathrm{abs,RH=40}}| < 1\,\mathrm{Mm}^{-1}\right)$”. This now unambiguously defines what conditions are required for a CRI to be considered valid for averaging.

22. Co-location (line 401). As observations are done on-board the same platform, they are certainly co-located, therefore this is unclear. Thank you for the comment. For clarity, we have removed the word “collocation” from this sentence.

23. Line 404: I suppose that data are removed when the cost function is larger than a threshold, not lower. Please correct. Thank you for the comment. We have corrected this error and replaced the “<” with “>” in addition to removing the extra “to be” in the text.

24. “coarse mode AOD is limited to < 0.1”: why? As stated in the text directly above this quote we state “To limit the presence of coarse-mode aerosol particles in this analysis”. We have added the following text to the preceding paragraph to better motivate this decision: “Because a significant presence of coarse-mode particles would be atypical within the altitudes that are sampled by the Falcon, we attempt to limit the amount of coarse particles in the columns of compared data.”.

25. LDR (line 407): clarify if you refer to VLDR or PLDR, because they bare not the same. The threshold at 0.13 (line 410) is not so small, at least for VLDR, and could indicate dust presence. Thank you for the comment. We have clarified the mathematical definition of LDR. Because the HSRL-2 description already stated that this is aerosol-specific data, we feel that an additional distinction between PLDR and VLDR is not needed.

26. Successful retrieval rate (line 482): please clarify if the retrieval rate means that a solution is found, or that the solution found is close to the observations). Thank you for the comment. This is simply the number of successful retrievals over the number of attempts, where the success is based on the maximum thresholds in the deviation of scattering and absorption. The text has been clarified by adding “$\left(\frac{\text{number of successful retrievals}}{\text{number of attempts}}\right)$” to this sentence.

27. Lines 483-486. The authors raise an important limitation and they should perform a more in-depth analysis to disentangle the causes. Note that for measurement noise (mentioned by the authors) this should be possible to address with the current data. Concerning the issue of non-sphericity, this can be run using MOPSMAP. Thank you for the comment. We agree that more in-depth analysis could be done for future studies, but we feel it would distract from the focus of the paper on more typical marine cases. We have also added a discussion to further support the low signal issues that were observed.

28. Acronyms (NRMSD, MRB, NMAD) must be explained when first used. Thank you for the comment. We have removed the discussion of MAD from the text as it is redundant to NRMSD. Additionally, we have ensured that all consistency statistics are defined when first used.

29. Line 565: specify that you refer to case #10 of table 5, because there are two flights on this date. Thank you for the comment. As mentioned above, we have removed the extra case from this table and updated all the text accordingly to be consistent.

30. "are likely organic and sulfate-dominated mixtures": this sentence is unexplained and unsupported by evidence Thank you for the comment. We have added the following text to support this statement: "This information indicates that the fine-mode particles being sampled were anthropogenic in origin and are likely sulfate-dominated mixtures with organic aerosol species. This is what would also be expected for this marine environment based on climatological evidence (Braun et al., 2021).".

31. "marine environment" (line 588): This statement seems inconsistent with the Hysplit trajectories showing continental influence. Thank you for the comment. We do not feel that these are contradictory statements. A notable feature of the marine environment of the Northwest Atlantic is the persistent influence of anthropogenic outflow. We have clarified this in the previous response.

32. "the data from case 12 are also shown in Table 5" (line 597): why? Thank you for the comment. We agree that showing these data here was unnecessary and we have removed this case from the included tables.

33. "1064 m": it is actually 238-4499 m Thank you for the comment and for catching this typo. We have corrected the value to be 4261 m.

34. Table 5 caption should read "case studies 10 and 12" Thank you for the comment. We have changed this table to include only data from case study 10, which is now case study 7 in the revised draft.