Dr. Wang, thank you for the early and thorough response!

My overall impression is that the initial response provided by the authors does not resolve the original problems with the methodology and I struggle to see the validity and utility of this method. Fundamental problems that render the inferred particle sizes unusable remain. These issues deal, to a great degree, in how the authors interpret and use a single figure from Rieger et al. 2018. The authors take that figure as definitive and prescriptive (a cursory look at Rieger's Fig. 6 leaves no ambiguity that assuming a distribution width of 1.6 with an error of ± 0.2 throughout the atmosphere is categorically wrong), fail to recognize the difference between the fine and coarse modes, and fail to address the real variability of particle size distribution parameters as measured by the University of Wyoming's optical particle counters (UWY OPC; this was the dataset Rieger et al. used to create their Fig. 6). If the authors were to expand the uncertainty in the distribution width beyond 1-σ, which only accounts for 68% of the atmospheric variability, then the uncertainty in the inferred particles becomes even more egregious. Finally, even with their overly conservative error estimates the particle size profile in Rieger et al.'s Fig. 6 is still better than what the authors predict; if the authors take Rieger's distribution width as canonical then why not use Rieger's radius profile instead of inferring it from OMPS. I think, in many cases, using Rieger's radius profile would yield a narrower solution space. Overall, it is difficult to see the utility of this method and its shortcomings are not accurately brought forward to inform the reader of its limitations. Despite the numerous fundamental flaws, as described below, the authors fail to communicate any of these issues or limitations to the reader.

The authors' response to my comments are in black, my additional comments are in blue. If the authors resubmit would they kindly address both of my communications in their revised manuscript?

Major Concerns with this Manuscript

1. As stated in my original review this methodology is not new. Variations go back to at least 1982. The authors agreed with this but suggested that the novelty lay in the application to OMPS data collected in the aftermath of 2 recent eruptions. The concerns I have with this are:

    (a) Application of an old method to a new dataset is, by itself, not interesting. The official reviewers and editor have the final say in this, but to me it is not interesting.

    (b) The authors used this method to look at the size evolution after 2 recent eruptions. However, this was done without a thorough evaluation of the accuracy of the method. I note that another reviewer had similar concerns.

    (c) The authors failed to address the instability of the solution space and account for the natural variability of aerosol size distributions within the atmosphere. The current model is based on the assumption that the distribution width is fixed and can never change. This not only neglects the natural variability but also the uncertainties in the OPC data. I still view this as a fundamental flaw and explain why below.

    (d) As explained in Taha et al. 2021, OMPS' 510 nm channel should only be used between 20–24 km and only in the northern hemisphere. The authors used this data throughout the profile in both hemispheres. Could the authors please explain to the reader why the recommendation of Taha et al. was not followed?

2. "We concur with your assertion that the distribution width may vary, but numerous in situ measurements have constrained the range of widths and models of the size distribution for ambient condition (Rieger et al., 2018)."
   The claim that "numerous in situ measurements have constrained the range of widths and models of the size distribution" is not true. These datasets describe the natural variability, but in no way limit the variability. This is an important point: Fig. 6 of Rieger et al. (plot of average OPC profiles) is *not* prescriptive, rather it is descriptive. Further, the uncertainty in distribution width, as provided by Rieger et al., of ± 0.2 is just the standard deviation of the OPC's estimated widths *at a single altitude* (20 km): this error becomes *much* larger lower and higher in the atmosphere. Further, this only represents 68% of the real atmospheric variability (at 20 km). The consequence of this is that 32% of the real distribution widths at 20km will be greater than ± 0.2 (again, this plus/minus value becomes significantly larger the further you get from 20 km). The problem is we do not know which points are closer to 1.6 and which are farther, which makes estimating the mode radius uncertainty challenging. A further complication is the uncertainty in the OPC's estimated errors, which further expands the solution space. In short, the static ± 0.2 value is not correct, it introduces an unrealistic constraint on the solution space (even at 20 km), introduces systematic bias, and the reader is left with no recognition that any of these issues exist.

3. "In the context of this retrieval method, assuming a fixed distribution width is a necessary step and a common approach used in current retrieval algorithms..."
   This may be common practice, but we are under obligation to evaluate the impact that our assumptions have on our results. The original manuscript ignored this almost entirely and the authors' response to my initial comment failed to fully recognize the impact these assumptions have *and* leave the reader with the impression that these problems do not exist.

4. "...as the reviewer notes, 1.6 is a good estimate for the distribution width."
   That is correct. A value of 1.6 is a good estimate, but that does not mean it is accurate. As stated above, accounting for the natural variability of these widths expands the solution space significantly and this must be accounted for before the reader can have confidence in this product.

5. "All remote sensing systems make assumptions about size distributions as noted above."
   I apologize if this comes across as being "nit picky", but this is not true. My only point here is that if the authors decide to include a comment similar to this in the revised version to please be more nuanced in their meaning.

6. "However, Rieger et al. (2018) Fig 6 shows that not all distribution widths are likely, and 1.6 is a reasonable choice."
   Rieger et al is not prescriptive and using a static value of 1.6 is demonstrably wrong. For example (looking at Rieger's Fig. 6), 1.6 falls near the mean value at 35 km, but at that altitude the distribution width ranged from $\approx$1.2–2.25 (far outside the ± 0.2). At 10 km the mean width ranged from $\approx$1.6–$\approx$2.7 (it went off the scale so I cannot tell). Finally, at 22.5 km the width ranged from $\approx$1.1–1.6. The point I'm trying to make is that while a value of 1.6 is reasonable for a rough estimate we have to understand that there is a high probability of the width being very different from 1.6 and that the assumed standard deviation (i.e., the ± 0.2 value) is highly dependent on altitude (especially under perturbed conditions). Further, Rieger's Fig. 6 only shows the mean distribution width $\pm 1{-}\sigma$, which leaves 32% of

the atmospheric variability unaccounted for). In short, the assumed distribution width (1.6) is incorrect, the assumed uncertainty of this width (0.2) is incorrect, and both values were applied statically throughout the profile without regard to the actual atmospheric variability as shown in Rieger et al. 2018 or in the UWY OPC record. This introduces insurmountable errors in the estimated radius, which makes the product unusable.

7. "To summarize, Figs. 1, 2 quantify the expected impact of CR uncertainty and distribution width uncertainty on size using values from Taha et al., (2021) and Rieger et al (2018). For 1.6 distribution width and color ratios between 2 and 4, the maximum size uncertainty is 20%..."
   Thank you for these informative figures. They certainly convey a lot of information. However, I still have some concerns about this analysis for the following reasons: 1. as explained above the distribution width is not consistent at 1.6 (this is even shown in Rieger et al.'s Fig. 6); 2. the uncertainty of with distribution width is not consistent throughout the profile (as shown in Rieger et al.); 3. the error used in your distribution width is fixed at ±0.2, which is only 1-σ (at 20 km), meaning this leaves ≈32% of the variability unaccounted for (since the standard deviation expands as you move away from 20 km this ±0.2 value accounts for less and less of the variability); 3. Taha et al. 2021 recommend only using the 510 nm channel between 20-24 km and only in the northern hemisphere (presumably the uncertainty becomes untenable outside these regimes). Even though the errors used in the calculation that went into Fig. 1 of your response are overly conservative, we still get a spread in radii that exceeds the values presented in Rieger's Fig. 6. For me, it is hard to determine what value has been gained by using this method instead of just using the statistical representation of the OPC data as Rieger did.

8. "Note the Rieger et al. (2018) also provides size distribution widths for coarse mode particles and the fine mode and coarse mode distribution widths are similar as are the mean distributions (1.6)."
   This is a very challenging problem. The coarse mode in Rieger et al. is actually the second mode in a bimodal distribution. To reduce this bimodal system to a single mode, and call the second mode's distribution width representative of the overall width, is not correct. Further, Rieger et al. did use the same "representative" value of 1.6 for the second mode, but the 2 profiles (fine mode vs coarse mode) are substantially different. Remember: Rieger et al. is not prescriptive and their 1.6±0.2 values are *not* valid throughout the profile and cannot be taken as a one-size-fits all approach. Details must be paid to the real variability in the atmosphere and not limited to a previous author's assumptions. Again, I agree that dealing with this second mode is challenging, but the argument put forth by the authors (quoted above) is categorically wrong. The consequence of their assumptions is that not only are the inferred particle radii under background conditions wrong, but they are even worse following major events.

9. "Section 3.2 aims to assess the impact of varying scattering angles on retrievals. This analysis is also related to Reviewer comment 3, and partly evaluated the error source from that. Yes, the comparison essentially compares the color ratio between OMPS-LP and SAGE III/ISS, but the purpose is different. Additionally, it is valuable to include a section comparing the retrieved particle sizes, rather than directing readers to seek out the color ratio comparison from other sources. We will make the goals of this section clearer."
   The entire paper, to this point, has been dedicated to inferring particle sizes from OMPS

color ratios. Introducing particle estimates from SAGE data in order to "..assess the impact of varying scattering angles..." provides an unforeseen transition. This will be confusing to the reader. My main concern here is that taking the additional step to convert SAGE data to particle sizes introduces an unnecessary layer of obfuscation that only decreases the validity of this comparison. It would be far more meaningful to do a direct comparison of the SAGE and OMPS extinction products as was done by Taha et al. 2021. In short, it is difficult for me to see how this section benefits the paper and it seems this type of comparison has already been done.

Again, I thank the authors for taking the time to read my comments and look forward to their response.