

We would like to thank Dr. Boone for his close reading of the manuscript and providing valuable feedback. Our responses are provided below (blue) to the reviewer’s comments (black). Because some of Dr. Boone’s comments are quite long we abbreviate them with the ellipsis (i.e., ...) when subsequent text is further elaboration on a single, overarching, point.

These numbers are typical values ascribed to background sulfate aerosols, but as I said above, they are not representative of the PSD you would need to reproduce measurement involving scattering using a monomodal size distribution. . .

This is correct and an excellent point. In general the stratospheric aerosol community have operated under the notion that particle size distributions (PSDs) are monomodal. While OPC observations clearly indicate that the distributions are bimodal (sometimes tri-modal), inferences of PSD parameters from remote sensing observations are almost always forced into the single-mode regime due to lack of information (we do note Dr. Boone’s recent work on solving for bimodal PSDs using SAGE III/ISS and ACE-FTS spectra). This has led to many in the field referring to “background” PSDs of $r_m=75$ nm and $\sigma=1.5$. . . which in truth refers only to the first mode. Further, Dr. Boone is correct that a major thrust of this paper is that these larger modes cannot be ignored except under certain conditions (e.g., wavelength and number density dependent). We agree with Dr. Boone that a paradigm shift needs to be made.

Before proceeding we would like to make the point that the text quoted by Dr. Boone (i.e., line 126 of the original manuscript) was in reference to determining the lookup table (LUT) resolution required to sufficiently mitigate the impact that the resolution has on the inferred PSD parameters. Indeed, under the current LUT design this results in LUT resolutions that are too high (i.e., the impact, as shown in Fig. 4, is likely much lower). However, Dr. Boone’s point is still valid that a single-mode distribution of $r_m = 75$ nm and $\sigma=1.5$ is not the best representation of background conditions. The text in the manuscript was updated to reflect this.

. . . My interpretation would be that the analysis approach tends to allow a number of solutions at larger r_m to leak into the solution set. If it happens in the synthetic data, I expect it would affect the analysis of real measurements as well (i.e., create a bias toward larger r_m). Is there any way to adjust the analysis to reduce that bias?

Dr. Boone’s interpretation of the interplay between r_m and σ is correct regarding the inverse relationship and how an overestimation of one may offset the other. Unfortunately the information content within the SAGE data itself is too limited to further reduce these biases. We do note that, per the discussion in the original manuscript, the bulk of these biases are within $\pm \approx 15\%$ when the measurement uncertainty is within 5% (not uncommon for SAGE extinction spectra). This bias decreases as particle size increases. We do not view this as a bad situation.

As discussed in the original manuscript we evaluated several extinction ratio combinations as well as variations on the LUTs’ PSD ranges. While the various LUT variations (see Table 2 of the original paper) resulted in PSD estimates that were, generally speaking, in good agreement, the LUT parameters presented in the paper are what yielded the minimum bias.

The H_2SO_4 content in the droplet will be governed by thermodynamic equilibrium. The H_2O vapor pressure for the droplet should equal the ambient partial pressure of H_2O in the atmosphere (Steele and Hamill 1981, [https://doi.org/10.1016/0021-8502\(81\)90054-9](https://doi.org/10.1016/0021-8502(81)90054-9)). If one knows the am-

bient temperature and H₂O concentration, an estimate of the H₂SO₄ content can be calculated (Bernath et al 2023, <https://doi.org/10.1016/j.jqsrt.2023.108520>). If there is higher H₂O pressure in the ambient air, the droplet should take up H₂O to find equilibrium (thereby decreasing wt% H₂SO₄). Conversely, if the H₂O pressure in ambient air is lower, there would be evaporation from the droplet to achieve equilibrium.

This is a fair and valuable point. The intent of this section was to demonstrate the impact of assuming an incorrect H₂SO₄ content. Therefore, we see no need to make a functional change to the algorithm. However, we incorporated a similar caveat in the paper and now include the suggested references.

Another reinforcement of fine mode parameters supposedly being representative of background aerosols. A caveat of some sort would be appropriate, unless you truly believe the numbers are representative (despite what your later results suggest).

Addressed above for a previous comment. Here, the reference to “background conditions” was removed. Since the purpose of this section is to evaluate the impact of an incorrectly assumed composition there is no further modification to the text.

I don’t disagree with what was done in this section, but it may underestimate the impact of smoke on the PSD parameters. Unlike the liquid droplet sulfate aerosols, smoke particles are not constrained to be spherical, which could significantly affect its scattering characteristics. Also, aged smoke particles apparently turn glassy, which would presumably change its optical constants, possibly outside the range of combined BC and BrC employed in the study, although for all I know turning glassy might push the optical constants closer to sulfate aerosol values, which would reduce the systematic errors.

We agree that the composition, shape, etc. of stratospheric smoke is largely unknown. We acknowledge this uncertainty in the paper. Because of this uncertainty we did not make any changes to the paper. Indeed, the final sentence of this section (“Therefore, we must provide a cautionary note when using data that may be contaminated by the presence of smoke.”) was provided to raise this awareness.

When you are working with extinction ratios, it is not clear how number density (N) is being derived...

Another reviewer raised a similar concern. Subsection 3.2.3 was added to discuss this. In short, the PSD parameters within the solution space were used to calculate the 1 μm extinction coefficient with N set to 1 cm⁻³ and the observed 1 μm extinction coefficient was divided by the calculated values, which indicates the difference in scale (i.e., N). Equation 2 was also updated to explicitly show that N plays a role.

This behavior is actually associated with resonances in the Mie scattering for certain particle sizes. Note that the curve for 384 nm in Figure 13 has two peaks, one near where r₂ is roughly equal to the wavelength (around 384 nm), and one where r₂ is roughly twice the wavelength (around 768 nm). Similarly, the curve for 520 nm has a peak where r₂ is near the wavelength (520 nm). If you were to extend the plots to higher wavelength, you would presumably see a second peak

somewhere around 1040 nm.

Exactly. I updated our statement to adopt some of your language to more clearly communicate this phenomenon. Thank you for the suggestion.

Droplets with different sizes will have slightly different H₂O vapor pressures because it will be impacted by the degree of curvature of the surface, but I would expect the composition of two droplets under the same conditions (temperature and ambient H₂O concentration) to have relatively comparable compositions.

Our original statement was unclear so thank you for raising this. The intent of this question was “what if 1 mode is sulfuric acid and the other is smoke?” The text was updated to remove this ambiguity.

We can also note from Figure 17 that the values of r_e from this study are systematically higher than the v7.0 results, which is perhaps another indication that this approach yields a high bias, particularly at smaller mode radius, as was (to me) suggested by the sensitivity study results in Figure 7.

Agreed, there is a systematic offset between the 2 products. However, the 2 are within the specified uncertainties of the v7.0 products (as discussed within the paper). When doing these types of comparisons the question always becomes: which one is correct? The best we can say is that the 2 are within the reported uncertainty.

It is surprising that you would choose not to mention Australian Black Summer (or Australian New Year, as it is also called) wildfires from 2019/2020. . .

Dr. Boone is correct in that this was a missed opportunity. We have added a brief discussion regarding the Australian fire as well as the recommended warning to the reader.

No comparison is made between the SAGE II results (Figures 15 and 16) and the SAGE III/ISS results (Figures 18 and 19). . .

The requested comparison was performed and a single figure (for the southern hemisphere) was added. Additional discussion was added to the paper as well. Briefly, we performed the requested hybrid intercomparison wherein we calculated PSD parameters using the SAGE III/ISS data with the 5/6/15 conditions as well as condition 0. Differences between these 2 products were compared. In general, the agreement between the 2 versions is within $\approx 30\%$ for all products within the main aerosol layer. However, there are larger differences (especially for number density) as discussed in the text.

The difference between the peak altitude near the equator versus that at higher latitudes is not a direct indication of aerosol sedimentation. As was mentioned earlier in the paragraph, Brewer Dobson circulation will naturally transport air to lower altitude as it moves poleward, even in the absence of sedimentation. To get a sense of the descent, you should observe the location of the > 300 nm plume at a particular latitude (e.g., 30 S or 45 S) and note how it changes over time.

Good point. The context of this paragraph points the reader to Brewer-Dobson circulation,

so the mention of sedimentation was a non-sequitur on my part. This statement was revised to remove ambiguity.

It depends on how you define the extent of the atmosphere, I suppose. For measurement purposes, I would define the overall height of the atmosphere according to the range over which atmospheric extinction contributes significantly to the signal. With that consideration, I would say you are looking through hundreds of kilometers, not thousands.

Statement revised.

This term was never explicitly defined. Looking at Equation 2, some people might interpret distribution width as σ , while others might interpret it as $\ln(\sigma)$. The latter choice might be the more logical interpretation. σ would be literally interpreted as the width of the distribution in $\ln(r)$ space. σ is defined in the paper as the geometric standard deviation, and the text shifts into referring to it as distribution width without comment.

The text was updated to remove ambiguity.

Ideally, when determining a statistical projection of the solution set, your ‘basis set’ would span the full set of expected conditions. At low temperatures and/or high H₂O concentrations, wt% H₂SO₄ can be 50% or lower (Bernath et al 2023, <https://doi.org/10.1016/j.jqsrt.2023.108520>). These would fall outside the chosen basis set.

Agreed. However, as discussed above, inclusion of a sulfuric acid estimate would go a long way in mitigating these assumptions. The SAGE III/ISS water vapor product is currently being released and the temperature product is still in development. However, incorporating this capability would be a significant upgrade to the current algorithm.

Traditionally, it would be written as “one can infer” Change made.

Typo. “...how well the two matched”? Corrected

LUTs Corrected

situations -> situation Corrected

$\sigma_2=1.05$ Corrected

$k_{520}/k_{1020} \leq 1.4$ Corrected

And Figures 15 and 16 [Corrected](#)

influence -> influenced [Changed](#)

deposition -> sedimentation? [Changed](#)

were -> was (plume is singular). [Corrected](#)

2 -> two [Corrected](#)