Below please find our specific responses to the reviewer RC2. The format is the reviewer comments in italics followed by our reply. Because of the similar remarks in CC2 we have included references to CC2 below. We also responded to CC2 earlier.

RC2:
Overall, we appreciate the reviewer's focus on manuscript improvement.

GC1) Finding re: number density independence (text interpreting Figure 1, lines 88-89). The first of the stated findings in the Abstract (3rd sentence), reached from interpreting Figures 1a and 1b, is not sufficiently demonstrated. The text on lines 88-89 states "This figure shows that the particle size is only a function of CR, and is independent of the number density." There are 2 stated findings in the sentence about particle size, and both are questionable, unless clarified to a specific context.

The upper Figure (1a) shows the curve in color ratio with particle size from Mie calculations, essentially presenting how much larger the aerosol extinction is at the shorter of the two wavelengths, compared to the longer wavelength, comparing 510nm & 745nm aerosol extinction to that at the reference wavelength of 869nm. The lower Figure (1b) shows how a set of assumed number concentrations translate into aerosol extinctions at 510nm and 869nm wavelengths, for a range of assumed median sizes.

The reasoning for why the Figure shows this shows one can conclude the number concentration is independent of the number density is far from clear. The methodology in the paper assumes this to be the case, within a particular range of particle sizes (e.g. particle sizes sufficiently scattering at the corresponding wavelength[e.g. above some threshold value in extinction-cross-section at that wavelength]). But the text is not correct to state that can be inferred from what is shown in the Figure. I suggest to delete that text on lines 88-89, and re-write the 3rd sentence of the Abstract that states this to be a finding of the study (lines 10-11).

Thanks for your suggestions. We will correct the text as the reviewer suggests.

GC2) Statements re: methods too general or unclarified The sentence from GC1) is an example of several statements within the manuscript (including within the Abstract) where results are stated too generally, with insufficient communication of the specifics. Given that this manuscript is within a specialist journal such as Atmospheric Measurement Techniques, the scientific writing on the methods needs to be quite precise. Whilst I understand that the text describing equation 1 is aiming to present the basis of the Bourassa et al. (2007,2008) method, the explanation on lines 75-83 need to be improved. For example the sentence on line 76 states "In computing the color ratio of aerosol extinction, the number density cancels out". Whilst that could be OK within a paragraph describing a methdological description, here this appears more prominently, and out of that context. My suggestion here is simply to delete this, expression, since it is part of the methodology already described comprehensively in Bourassa et al. (2007,2008). See specific revisions SR9 and SR10.

>See response to SR 9, 10 below

GC3) Comparisons to balloon-borne laser particle counter measurements (Section 3.1). This is the other part of the text where the method needs to be better explained (given this is submitted for an Atmospheric Measurement Techniques paper). The text on lines 125-126 need to provide the location of the sounding compared to, and the specific size-cut for the particle number shown in the black line in Figure 3b. (this information to be re-stated also in the Figure caption).
The terminology can be confusing because the Wyoming laser-OPC (WL-OPC) was developed at Boulder (see Ward et al. 2014) and the new lightweight OPC system is called L-OPC (see Kalnajs and Deshler, 2022). The cavity-laser OPC is described in Ward et al. (2014), with multiple size channels, down to 75nm radius (75, 150, 250, 500nm, and 1.0, 2.5, 5.0, and 15.0 microns). For these comparisons to the OMPS-LP aerosol extinction, I am assuming the 75nm radius channel is shown, but this is important considering also that the original OPC40 and OPC25 only measured to 150nm particle radius (see Deshler et al., 2019). Please add, within the text on lines 125-126, and the caption to Figure 3, the minimum particle size for the size-resolved number concentration shown. Given the Mie scattering curves will of course vary for the different wavelengths considered, the minimum size is an important issue here.
Related to this a suggestion is to add a dashed line for the R>150nm number concentration (and possibly also the 250nm line, in dot-dashed or so).

We agree with these excellent suggestions. We modified the text as suggested.

The overestimation shown in the 16-18km altitude-range could potentially be due to only some proportion of those R>75nm particles being measured, even at the shorter of the two OMPS-LP wavelengths. I appreciate this is a retrieval, but then the issue of what particle sizes are represented within the two aerosol extinction metrics within the color-ratio particle size method probably justifies considering an uncertainty-range across more than 1 of the size channels (particle size cuts) in the comparisons.

We added an uncertainty calculation section in the revised manuscript as follows:

Adjusting the size distribution will affect the cross section and the Mie phase function. To investigate the OMPS-LP aerosol color ratio sensitivity to the assumed phase function, we perturb the phase function parameters by ± 10% and run the OMPS-LP retrieval algorithm at a range of scattering angles observed during a single orbit, similar to Chen et al. (2018). OMPS-LP V2.0 aerosol retrieval algorithm assumes a gamma aerosol size distribution using fitted parameters of α = 1.8 and β = 20.5 where effective radius reff = 0.185μm. Their conversion formular (Chen et al., 2018) is as follows

$$r_{eff} = \frac{(\alpha + 2)}{\beta} \quad (5)$$

which is used to calculate the aerosol phase function (see Fig 1). For a width W=1.6, we can get a modal radius rm = 0.1μm by the following formular,

$$r_{eff} = r_m \exp\left(\frac{5}{2} ln^2 S\right) \quad (6)$$

Figure 2 shows that the phase function is more sensitive to β changes than α. A 10 % change of β can produce ± 10 and 15% change in 510 nm and 869 nm phase functions, respectively, while a 10 % change of α results in a ± 3 and 5% change for both wavelengths. Chen et al. (2018) noted that increasing α increases the peak of the differential size distribution while increasing β shifts the peak distribution to a larger particle radius. Figure 2b shows that the phase function perturbations produce anti-correlated but lesser changes in aerosol extinction. The changes caused by β perturbations are mostly within 5% for both wavelengths and 3% for α. The structural change in the extinction coefficient along the orbit is caused by scene reflectivity (Chen et al., 2018). The effect on extinction ratio (510/869) is shown in Fig. 2c for the same variations of phase functions. The color ratio perturbations are mostly within 3% between scattering angles of 65-125° and 5% outside that range, except for the very small scattering angles, which might be caused by scene reflectivity changes. Let's pick a typical color ratio of 3 with W = 1.6 to calculate the uncertainty of retrieved particle size. The size range would be 0.096 ~ 0.103 µm for 3% change in color ratio perturbations and 0.094 ~ 0.106 µm for 5% change in color ratio perturbations. It means the 3% and 5% change in color ratio perturbations lead to < 4% and < 7% difference in retrieved aerosol particle size, respectively.

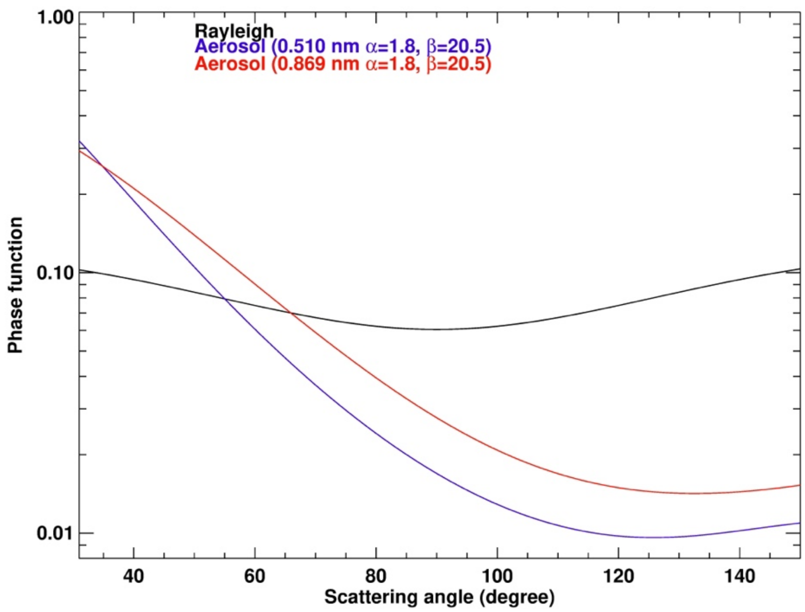

Figure 1: Plot of the aerosol phase function used for OMPS LP retrieval algorithm, which assumes gamma size distribution at 510 (blue) and 869 nm (red). Rayleigh phase function is also shown (black).

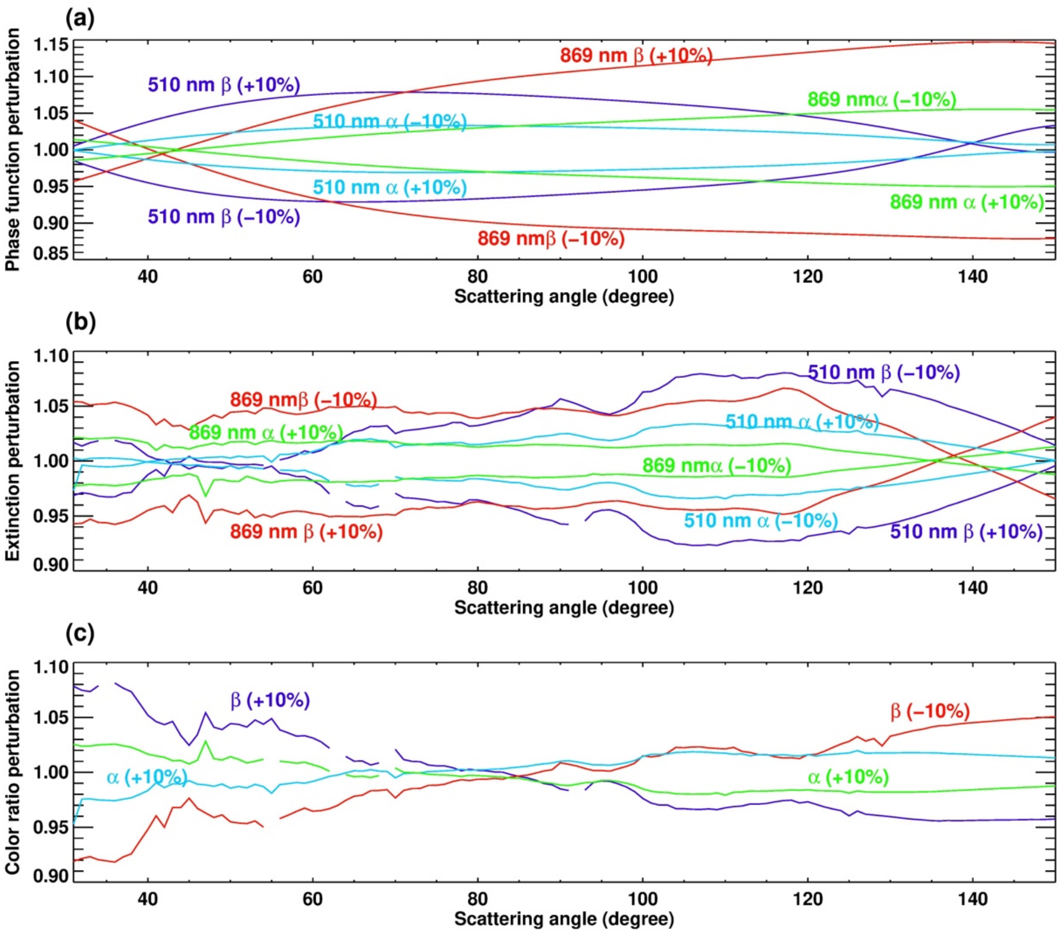

Figure 2: plot of simulated phase function (a), aerosol extinction (b), and color ratio (c) perturbations as caused by gamma parameter changes of ± 10%. Perturbations are shown relative to OMPS LP operational retrieval at various scattering angles. Scattering angles represent the range of are in the northern hemisphere, while large scattering angles are in the southern hemisphere (Taha et al., 2021). The aerosol extinction was retrieved using a single orbit on 12 September 2016 at 20.5 km altitude.

List of specific revisions

SR1) Abstract, lines 8-9 -- This initial text "We have developed an algorithm" might make some readers assume the MS is to explain some development of a new algorithm, but the MS is rather applying an existing method already developed for the OSIRIS data (Bourassa et al., 2007, 2008), to OMPS measurements, comparing to in-situ measurements for two recent volcanic case studies. For this initial sentence better to put the object of the sentence (derive the size...) at the start of the sentence, with also referring more generally to "aerosol microphysical parameters" rather than the specifics in this initial sentence.

Please change "We develop an algorithm that uses the aerosol extinction at two wavelengths..." instead to "We apply an existing method to derive aerosol microphysical parameters from OMPS dual-wavelength aerosol exinction measurements, to analyse particle size variations *for two recent stratospheric volcanic case studies." or similar.*

Good point. We will modify the text as suggested.

SR2) Abstract, lines 9-10
With the above re-wording, the first part of the 2nd sentence of the Abstract is already integrated into the above re-worded 1st sentence, and the latter part re: SAGE should be stated later, being clear this is SAGE-III on ISS.

We will correct.

SR3) Abstract, line 10 -- This 3rd sentence of the Abstract also needs to be re-worded, with the current text "We show that the color ratio between two wavelengths is insensitive to number density." suggesting the manuscript is presenting this as a finding from the study.
Figure 1 is very useful in showing the variation with particle size and relative magnitudes of monochromatic aerosol extinction at 3 specified wavelengths, for a log-normally distributed liquid aqueous sulfuric acid solution aerosol population (for an assumed refractive index spectrum and water content/weight-percent). However, as is explained in General Comment 1 above, the text on lines 88-89 is not correct in stating the Figure shows the color ratio is independent of number density. I understand this is an assumption within the methods explained in the Bourassa et al. (2007, 2008) studies, and for a particular range of particle sizes, it's a reasonable assumption to make. However, the Abstract should not present as a finding from the study. Also, the "and thus" in the 2nd part of the sentence (with the current wording) does not follow, and suggests a misunderstanding re: what is assumed in the method, with the wording "We show the color ratio between two wavelengths (e.g. 510nm/869nm) is insensitive to aerosol concentration, and thus can be used to derive aerosol size assuming a log-normal size distribution."

Within the re-constructed sentence, the color ratio metric also needs to better communicated, a suggested re-wording to have "of aerosol extinctions at" in place of "between", and change "(e.g. 510nm/869nm)" instead to "(510mm and 869nm)". My specific suggestion for the 2nd part of this sentence is to delete "and thus can be used to", and also change "We show that" instead to a re-worded sentence similar to below:"The color ratio between aerosol extinction at two wavelengths is used to derive from satellite measurements large-scale particle size variations within volcanic aerosol clouds in the stratosphere."

We will make these changes.

SR4) Abstract, lines 12 -- Re-word "With the size and extinction, we can compute a number density consistent with both wavelengths". The particle size is being inferred from the two-wavelength aerosol extinctions, not measured directly. With the re-constructed preceding sentences (from SR3), suggest to instead have this sentence be to explain some greater specifics in the method, prior to its application for profiling the size distribution of the Raikoke and Hunga Tonga aerosol clouds. A suggested re-wording "With the size and extinction, we can compute..." instead with "As a further microphysical Consistent with the two extinction

Good suggestions, we will implement these.

SR5) Abstract, lines 13-14 -- Please re-word this sentence to be more specific re: the Comparisons between the particle size variations from OMPS-LP and those derived from SAGE-III on ISS, giving some specifics about how well the two size products compare.

It is difficult to describe these comparisons in a single sentence, but we will modify the sentence to read: Our size and concentration estimates are also in good agreement between 24 and 18 km with measurements made by the Stratospheric Aerosol and Gas Experiment on the International Space Station (SAGE III/ISS) over multiple scattering angle ranges.

SR6) Introduction, lines 21-22 -- change "have been connected to short-term changes in climate" instead to "major eruptions causing substantial short-term changes in climate", or similar. The first sentence (of this 1st paragraph of the introduction) mentions the volcanic impacts on climate, but this adaptation of the sentence is then consistent with the strong (but temporary) radiative forcings after historical very large-magnitude explosive tropical eruptions.

We agree and clarify this.

SR7) Introduction, line 25 -- Please re-word this sentence to be clearer what is meant by "dust" in this context ("Volcanic ash, Pyro-CB smoke and dust") -- presumably it is cosmic dust (i.e. meteoric aerosol) that is meant here, right? Suggest to have the re-worded text provide this information in relation to citing a recent observational studies for each of these non-sulphate stratospheric aerosol constituents. Suggestions are Vernier et al. (2016), for volcanic ash, Khaykin et al. (2020) for pyro-Cb smoke, and Schneider et al. (2021) for meteoric aerosol. With this re-wording the text on line 26, suggest to cite paper also for the volcanic ash Heating (Muser et al., 2020), and cite the Yu et al. (2019) for the wildfire smoke heating effect. The cosmic dust (meteoric smoke) may be present at sizes smaller than for wildfire smoke and volcanic ash, also some components dissolving into the sulphuric acid solution aerosol particles (see James et al., 2023), the heating effect is primarily associated with the smoke and ash.

No, we meant mineral dust which has been observed in the lower stratosphere by research aircraft. We agree that references would be helpful here and will include them in the revision.

SR8) Introduction, lines 32-34
Re-word "has provided global monitoring of the stratospheric aerosol layer since 1975" because the SAGE and SAM-II record only began in 1979 (see McCormick et al., 1977). The original testing of the SAM sensor on the Apollo Soyuz mission was in 1975, but the global monitoring only began in 1979. Re: the text on the solar occultation methods, the so-called "onion-peeling" retrieval method was originally developed in the 1960s (Edward Ney group at the University of Minnesota), and it will help retain the heritage of the instrument development to cite the Pepin (1969) report that first documented the technique, and the Rosen et al. (1969) which shows (Figure 4) the only peer-reviewed paper showing the initial application of the methods to balloon-borne solar extinction measurements, and later further developed for application to the

first satellite measurements of the stratospheric aerosol layer (the SAM, SAM-II, McCormick et al., 1979), and then SAGE, SAGE-II and SAGE-III series of satellite measurements.

Nice history, CC1 also provided some references on the technique that we will include in the revised manuscript.

SR9) Section 2, line 76-78
Add ", at wavelength l, " after "The aerosol extinction" and add subscript l symbol to the two wavelength-dependent symbols in equation 1 (sigma and AR). In addition, please change the abbreviation "AE" for aerosol extinction, as this may initially confuse some readers, given the related size-associated aerosol metric "Angstrom exponent". Please change all occurrences of "AE" instead to either "k" (used in the Bourassa et al. (2007, 2008) papers) or other terminology (the letter b is used, subscript "ext" to denote aerosol extinction in Seinfeld & Pandis (2006). It also needs to be stated explicitly that these calculations are based on an assumed log-normal size distirbution, with also the associated water content (sulphuric acid and water composition). As explained in General Comment GC2, please delete the two short sentences lines 77-78 as this statement does not follow from what is shown. The information and associated assumptions are explained within context in the Bourassa et al. (2007,2008) papers.

We will implement these suggestions.

SR10) Section 2, lines 82-83
As mentioned in General Comment GC2, the text "...but as the two wavelengths approach each other" is too colloquial and needs to be formalised. Also, the "Any two distinct wavelengths can be chosen" at the start of the sentence also does not need to be stated. Suggested restructuring to better explain the issue. Suggest to re-word the original text: "Any two distinct wavelengths can be chosen for CR, but as the two wavelengths approach each Other Fig. 1a shows that the CR gradient decreases and thus the uncertainty in the retrieved size increases." instead to "For OMPS-LP wavelengths closer to the reference 869nm wavelength, the gradient in the colour ratio curve is less steep, causing an increased uncertainty in the retrieved particle size." It is worth noting also that the 869nm aerosol extinction measurement is "cleanest" retrieval in relation to effects from other species, and this must also be considered in relation to how the errors/uncertainty differ between different color-ratio wavelength-pairs.

We will implement these suggestions.

SR11) Figure 1 caption, lines 431-435. Although it is not stated explicitly, my understanding is that these calculations are based on a log-normally distributed aerosol, with geometric standard deviation of 1.6, following the specification within SASKTRAN. This should be stated in the caption to Figure 1.

Yes

SR12) Section 2, line 368 -- formatting error re: Loughman et al. (2015) reference.

Corrected. Thanks.

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