Review of "Using OMPS-LP color ratio to extract stratospheric aerosol particle size and concentration with application to volcanic eruptions", manuscript prepared for AMT by Wang and co-authors.

This manuscript describes an adapted application of an existing method to derive aerosol particle size distribution parameters from measurements of aerosol extinction from the OMPS-LP limb-profiling sensor (e.g. Taha et al., 2021).

The method is applied to analyse vertical and meridional variations in microphysical aerosol properties in background stratosphere conditions and from two volcanic case studies, the 2019 Raikoke and 2022 Hunga-Tonga large-magnitude explosive eruptions,

For the Raikoke case, the derived number concentrations are evaluated comparing to in-situ optical particle counter measurements from high-altitude balloon soundings from North America. The OMPS-derived particle size and number concentrations are compared also to similar colour-ratio analysis from the SAGE-III sensor.
The Figures 9 and 10 represent an important and valuable first analysis of the size variations evident across the vertical profile of the Hunga-Tonga stratospheric aerosol enhancement.

The paper is certainly appropriate for AMT, and the analysis of the Hunga-Tonga will be of particular interest with the unexpectedly strong aerosol optical depth. The microphysical parameters are relevant the two hypotheses for the effect -- that the aerosol scattering is amplified by the co-emitted water vapour, and whether there was additional primary emitted aerosol (from the vaporised seawater and/or in-plume-oxidised sulphate).

I have seen that three other revieweers have submitted reviews on the manuscript already, and although I have not referred to their comments when carrying out this review, my comments here are focused on improving the Introduction and Methods section, and in relation to the current interpretation of Figure 3.

Given that this is a manuscript submitted to the specialist journal Atmospheric Measurement Techniques, the methods section requires substantial improvement, and whilst I understand most interest will of course be towards the main scientific results re: the particle size variations, the section 2 requires better summary explanation of the methods.

In some places the scientific writing style needs to be improved for an article in a peer-reviewed journal, avoiding "our algorithm" etc., written in a more formal/impersonal tense. A particular substantial revision, is re: equation 1 of the paper, which as currently written does not convey the approximate nature of the relationship assumed when inferring the large-scale variations in particle number and size.

That said, the article is certainly publishable in Atmospheric Measurement Techiques once the text has been sufficiently improved. The variations shown for the Hunga-Tonga and Raikoke aerosol clouds will be of substantial interest to the stratospheric aerosol community.

Please see below a list of specific revisions to improve the text, and please also check for where there may still be other parts of the text where the wording could be improved.

## General Comments

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 colour 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).

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

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).

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.

List of specific revisions
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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.

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.

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 sulphuric 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."

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

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.

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.

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.

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.

SR9) Section 2, line 76-78

Add ", at wavelength $\lamda$, " after "The aerosol extinction" and add subscript lamda 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.

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 re-structuring
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.

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.

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

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